

Torrefaction and Steam Explosion of Selected Biomass for Biofuel Production

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By

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Abstract

Lignocellulosic biomass has demonstrated great potential as feedstock for pellet production, notwithstanding the fact that the industrial production of pellet is faced with some economic challenges. Pretreatment research has been focused on identifying, evaluating, developing, and demonstrating promising approaches that enhance the physiochemical and mechanical properties of biomass. Assessment of biomass pretreatment processes depends on a parameter called the severity factor, which is defined as the combined effect of temperature and residence time. This study summarizes pretreatment processing methods, namely, steam explosion and torrefaction of selected biomass feedstocks (sawdust and oat straw), processing characteristics, and product properties. The study also discusses the economics of densifying biomass, outstanding challenges, the potential for industrial applications of biomass-pretreated products, and global trends in biomass utilization. Steam explosion pretreatment of sawdust and oat straw was conducted under 3 stages: mild, medium, and severe conditions. The effects of temperature, time, and moisture content on the physiochemical and mechanical properties of pellets formed were examined. The p -values of the regression models for all the response variables (dimensional stability, tensile strength, and pellet density) studied were significant ($p < 0.05$), except for the pellet density of steam-pretreated oat straw pellets. The interaction of these three factors did not significantly affect the response variables of oat straw pellets. Microstructural examination of the pellets from steam-pretreated biomass revealed that the material contained particles that were more closely bonded and featured a cemented surface with fewer pores when compared to particles from untreated oat straw and sawdust.

Microwave-assisted torrefaction was conducted on white spruce sawdust (WSS) at temperatures of 200 °C, 250 °C, and 300 °C and retention times of 5 min, 7 min, and 9 min in an inert

environment. Torrefaction pretreatment is a mild form of pyrolysis that has the potential to produce high-quality raw material for making biofuel that serves as a replacement for coal in the bioenergy industry. The torrefaction process produces a solid carbon, commonly known as biochar, and condensable (torrefaction liquid (TL)) and non-condensable gases. In this study, torrefaction characteristics were investigated to observe its effects on the thermal and physiochemical properties of the pellets produced. During the torrefaction process, a significant mass loss associated with the decomposition of hemicellulose was observed. The hemicellulose content drastically reduced to approximately 1.8% from 19.25% and the cellulose content was reduced by approximately 10%, while the lignin gained approximately 35% as the severity increased. This led to an improvement in the higher heating value (HHV), hydrophobicity, bulk, particle density, pellet dimensional stability, and pellet density. However, the pellet tensile strength decreased as the torrefaction severity increased. Therefore, to enhance the tensile strength of the pellets, the introduction of a binder was necessary. Torrefaction liquid and sawdust were used as additives at different proportions during pelletization. The addition of binders (torrefaction liquid and sawdust) to the pellet formulation increased the tensile strength of the torrefied WSS by approximately 50%. The OH groups in the biomass break down to a limited degree due to dehydration. This hinders the formation of H bonds, thereby increasing the chances that the pretreated biomass will become hydrophobic. The Scanning Electron Microscope (SEM) graphs showed that the torrefied WSS pellets demonstrated more firmly glued surfaces with fewer pores spaces when set side by side with the raw pellets. The thermogravimetric analysis conducted showed that the torrefaction of WSS slightly reduced its thermal stability.

Technoeconomic analysis of six case scenarios (wood pellet production from raw sawdust in a pellet plant (I), straw pellet production from raw oat straw in a pellet plant (II), integrated torrefied treated sawdust pelletization plant utilizing the torrefaction liquid (TL) as the pellet binder (III), integrated torrefied treated oat straw pelletization plant utilizing the torrefaction liquid (TL) as a pellet binder (IV), integrated steam treated sawdust pellet plant (V), and integrated steam treated oat straw pellet plant (VI)) was carried out to develop a process model for pellet production from sawdust and oat straw that uses torrefaction and steam explosion pretreatments prior to pelletization. The breakdown of equipment purchase cost showed that the torrefaction reactor is the most expensive unit with about 51% of the purchase cost. Scenarios 1, 2, 5, and 6 generated positive net present value at baseline model. Facility-dependent and feedstock costs were the major significant contributors to the pellet production cost. The minimum selling price of pellets obtained from Scenarios 1-6 were \$113.4/t, \$118.7/t, \$283.4/t, \$298.7/t, \$200.5/t, and \$208.4/t, respectively. The profitability of pellet production as determined by the net present value (NPV), internal rate of return (IRR), and payback period was found to be sensitive to variations in feedstock cost.

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I will forever and ceaselessly remain thankful and grateful to God Almighty, whose infinite mercies saw me through to complete this study successfully.

Dedication

I dedicate this Thesis to GOD Almighty and to my late sister Onyenwoke Ebere Mercy (Bento).

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List of Abbreviations

| | |
|-------|---|
| AAFC | Agriculture and Agri- food Canada |
| AACC | American Association of Cereal Chemists |
| ADF | Acid detergent fibre |
| ADL | Acid detergent lignin |
| ANOVA | Analysis of variance |
| AOC | Annual operating cost (US\$) |
| ASAE | American Society of Agricultural Engineers |
| ASABE | American Society of Agricultural and Biological Engineers |
| ASTM | American Society for Testing and Materials |
| BIMAT | Biomass Inventory Mapping and Analysis Tool |
| CFRC | Canadian Feed Research Centre |
| d.b | Dry basis |
| DFC | Direct fixed capital cost (US\$) |
| DS | Dimensional stability |
| DTG | Differential thermogravimetric |
| EMC | Equilibrium moisture content |
| FC | Fixed carbon (%) |
| GC/MS | Gas chromatography-mass spectrophotometer |
| GDP | Gross domestic profit |
| HHV | Higher heating value |
| H/C | Hydrogen to carbon ratio |
| HP | High pressure |

| | |
|------|--------------------------------------|
| HSI | High Steam Industries |
| IRR | Internal rate of return (US\$) |
| MC | Moisture content (%) |
| MLTC | Meadow Lake Tribal Council |
| NDF | Neutral detergent fibre |
| NPV | Net present value (US\$) |
| NREL | National Renewable Energy Laboratory |
| NT | Non treated |
| OH | Hydroxyl groups |
| OPEX | Operational expenditure (US\$) |
| O/C | Oxygen-Carbon ratio |
| PBP | Payback period (yr) |
| PC | Purchase cost |
| pH | Potential of hydrogen |
| PSP | Pellet selling price |
| RT | Retention time |
| SC | Start-up cost |
| SEM | Scanning electron microscope |
| SF | Severity factor |
| SK | Saskatchewan |
| SPF | Spruce–pine–fir |
| TCI | Total capital investment |
| TEA | Techno-economic analysis |

| | |
|------|---|
| TGA | Thermogravimetric analysis |
| TDT | Thermal decomposition temperature |
| TL | Torrefaction liquid |
| TOP | Torrefaction before pelletization process |
| TPC | Total plant cost (US\$) |
| TPDC | Total plant direct cost (US\$) |
| TPIC | Total plant indirect cost (US\$) |
| VM | Volatile matter (%) |
| w.b | Wet basis |
| WC | Working capital |
| WSS | White spruce sawdust |

List of Nomenclature and Symbols

| | |
|------------|---|
| δ_x | Tensile strength (horizontal) stress (Pa) |
| F | Load at fracture (N) |
| d | Specimen diameter (m) |
| l | Specimen thickness (m) |
| M_Y | Percent mass yield of the torrefaction operation |
| M_T | Mass of the torrefied sample |
| M_I | Mass of the untreated sample |
| W_L | Weight loss |
| E_Y | Energy yield |
| H_1 | HHV of untreated sample |
| H_2 | HHV of treated sample |
| M_{SR} | Percent mass of solid yield |
| M_{TL} | Percent mass of liquid yield |
| M_{GY} | Percent mass of gas yield |
| M_{LS} | Mass of liquid sample |
| wt% | Weight percentage |
| P | Probability value (%) |
| L | Transportation distance |
| A | Area |
| R | Biomass collection radius |
| Π | Pi |
| C_t | Biomass transportation cost |
| C_f | Fixed cost of transportation |
| C_v | Variable cost of transportation |
| C_{ne} | Estimated capital cost |
| Q_{ex} | Baseline pellet plant size |
| C_{ex} | Empirical capital cost of a baseline pellet plant |
| Q_{ne} | Pellet plant size |
| I_0 | Initial investment in the pellet plant |

| | |
|-------------------|--|
| CFA_t | Annual cash flow |
| SVN | Salvage value |
| R | Discount rate |
| N | Year with N terminal time |
| R_0 | Severity factor |
| T | Reaction temperature ($^{\circ}\text{C}$) |
| t | Residence time (min) |
| d_{gw} | Geometric mean diameter (mm) |
| S_{gw} | Geometric standard deviation of particle diameter (mm) |
| W_i | Mass on the i^{th} sieve |
| Vol_{14} | Volume of pellets after 14 days (mm^3) |
| Vol_0 | Volume of pellets directly after pelleting (mm^3) |

Chapter 1

Introduction and Objectives

1.1 Background of Study

Globally energy demand is anticipated to increase by approximately 28% from 2015 to 2040 (EIA 2017). Energy security and the climate change concerns are key factors to searching for sustainable and renewable energy resources (EIA 2017, Owusu and Asumadu-Sarkodie 2016).

The extraction and utilization of fossil fuels have been known to cause detrimental effects to the environment because these processes emit greenhouse gases (GHGs) and synthetic chemicals which cause global warming. These gases and synthetic chemicals include carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O), and fluorinated gases - hydrofluorocarbons (HFCs), perfluorocarbons (PFCs), and sulfur hexafluoride (SF₆). The usage of about 80% of fossil fuel resources will result in a 70% increase in GHG emissions (Shahrukh et al. 2016). Therefore, there is an urgent need to reduce dependence on fossil fuels and achieve a sustainable renewable energy supply (Tumuluru et al. 2011). The utilization of lignocellulosic biomass is seen as a solution to cut down the dependence on raw materials from fossil resources and their effect on climate change (Clauser et al. 2022).

Wood residues have been recognized as a renewable energy source for quite some time, and their use in the Canadian forestry industry has come a long way. Forest residues such as bark, wood chips, branches, treetops, and sawdust have become feedstocks for electricity cogeneration facilities and advanced steam facilities (Onyenwoke et al. 2022). The worldwide demand for wood pellets for heating and power generation is increasing steadily. It is expected that by the year 2025, the global demand for wood pellets will increase to about 52 Mt, which is about 3 times its value in 2010 (Statistica 2022).

Currently, Canada is second only to the USA in wood pellet production worldwide as a result of their abundant biomass supply, effective logistics, and established industry (wood pellet market 2021).

Aside from woody biomass, Canada also generates an abundance of agricultural residues annually, where notably Saskatchewan is a major agricultural province (li et al. 2012, Sarker et al. 2020). Straw biomass is traditionally left on the field after grain harvest to improve soil organic matter. Straw biomass can be more completely utilized as a feedstock to produce biofuels and bioproducts (Kim and Dale 2004). The use of such abundant biomass sources that produce low net carbon dioxide emissions can be part of a solution to decreasing greenhouse gas levels associated with the production and use of fossil fuel resources (Kim and Dale 2004).

The challenges associated with the utilization of raw biomass that can limit their industrial usage include high moisture content, low bulk density, poor grindability, hydrophilicity, and inherently low energy value. These problems affect the storage, handling, transportation, and energy efficiency of this raw biomass (Azargohar et al. 2019a; Azargohar et al. 2019b; Haykiri-Acma et al. 2016; Medic et al. 2012). To overcome these limitations, a variety of pretreatment methods have been developed.

Ever since the Industrial Revolution around 150 years ago, most of the electricity used in the United States has come from burning fossil fuels. These fuels, such as coal and oil, are all made of carbon (Wilkerson 2018). The bulk of the coal driving the Industrial Revolution and contributing to global warming today has been deposited during the Carboniferous Period (359–299 million years ago), resulting in a significant drawdown of atmospheric carbon dioxide at that time. The interesting similarities between biofuels and coal is that both start with biomass, both require heat and pressure under anoxic conditions, both generate a product with value as a fuel

(Feulner 2017). The temperatures of coalification are frequently reported as approximately 100–170 °C for bituminous coal (Taylor et al. 1998) and 170–250 °C or even 200–275 °C for anthracite (Taylor et al. 1998; Daniels et al. 1994). The temperatures of pretreatment of biomass ranges from 160-300 °C (Onyenwoke et al. 2022; Onyenwoke et al. 2023a). But obviously, there are significant differences between coalification and torrefaction because coal has much higher heating value (HHV), as well the coal slag left after burning coal. The calorific value of biomass pellets is 10% lower than medium coal (Feulner 2017). The coalification takes a significant time (on order of a few million years), whereas torrefaction happens in shorter period (hours).

One solution being proposed is, oddly enough, to continue burning carbon for energy like we have been doing – but with one difference: instead of fossil fuels, burn carbon that comes from plants (Feulner 2017). These aim to modify the physiochemical properties of raw biomass to make it more suitable for biofuels production and are themselves part of the overall energy balance and life cycle of any process that produces energy from extant biomass (Onyenwoke et al. 2022; Onyenwoke et al. 2023a; Cristóbal et al. 2016; Näyhä 2012; Clauser et al. 2021).

1.2 Potential of Biomass Production in Saskatchewan

Biomass resources comprise of forest and agricultural residues. It also includes industrial and municipal solid wastes. Saskatchewan (SK) Ministry of Agriculture reported that in 2013, Canada produced about 69.6 Megatonnes (Mt) of wheat, canola, barley, and oat, with Saskatchewan production at 33 Mt (Iroba 2014). The amount of grain straw produce is about 30-40% of the grain mass harvested. Wood waste comprises 12% to 24% of every log entering a mill, depending on the type of finished product. Major waste components include sawdust & planer shavings from sawmills, and bark. The weight of wood waste available for new projects in

SK is 590, 000 tonnes/yr (oven dry tonnes). Currently, no market for forest biomass exists in SK, which is piled and burnt at roadside or spread back into cut blocks after harvest (Saskatchewan Ministry of Energy and Resources, 2022). Oat straw and sawdust are the selected biomass feedstock for this research. There is a need to develop processes that exploit locally produced biomass, to optimize transportation costs vs benefits in Canadian Prairies. In addition, these residues are inexpensive, abundant, and readily available resources (Kim and Dale 2004). About 40% of Canada's cultivated farmland is found in Saskatchewan (Government of Saskatchewan 2022); its diverse agriculture and agri-food sector has an international reputation for consistently supplying high-quality food and ingredients.

1.2.1 Potential of oat straw production in Saskatchewan

Found centrally in the Canadian Prairies, Saskatchewan is one of the largest exporters of agri-food products in Canada. With a large productive land base and a strong commitment to crop development research, Saskatchewan is playing a vital role as a major food supplier to the world's rapidly growing population. Canada is a major supplier of oat globally. The global yield of oats in 2021 amounted to about 22.68 Mt (Shahbandeh 2022a). Oat is the second most cultivated cereal crop in Saskatchewan Canada behind wheat, with a yield of oats in Canada in 2020 of approximately 4.6 Mt (Shahbandeh 2022b). This represents an increase from about 4.23 Mt produced in 2019 as shown in Figure 1.1.

Saskatchewan is the largest producer of oat in Canada, accounting for about 50% of Canada's oat production. Figure 1.2 shows Saskatchewan oat production in 2016-2020. Although animal consumption accounts for the majority of oat use globally, because of the awareness of the nutritional benefits of whole grains, the human food market for oats is growing. Most Canadian

oats are used for human consumption (POGA 2022). Approximately 30-40% of the whole oat plant is made up of straw (POGA 2022), After grain harvesting, most crop residues are left on the field. Some of these residues have been used for livestock feeding, bedding, insulation and mulching. Oat straw can serve as a feedstock (raw material) for producing biofuel and bioproducts. The use of agricultural residues such as oat straw for energy production relieves the pressure on food resources compared with the use of the more readily convertible starch-based biofuels, while also adding value to low-value crop residues (Adapa et al. 2011).

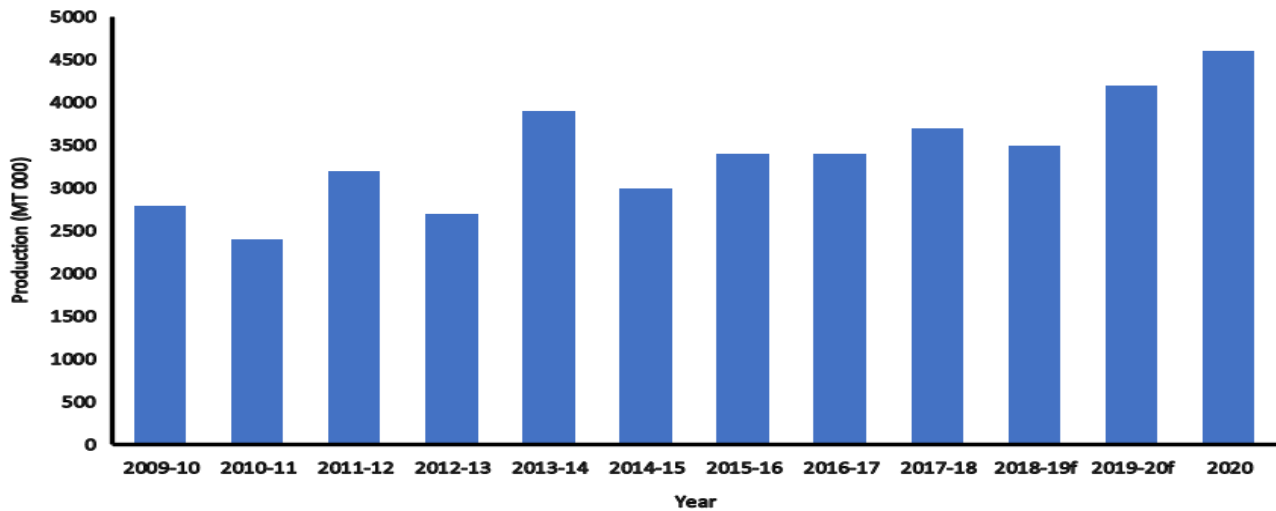


Figure 1.1. Canada oat production. Source: Statistics Canada; AAFC forecast.

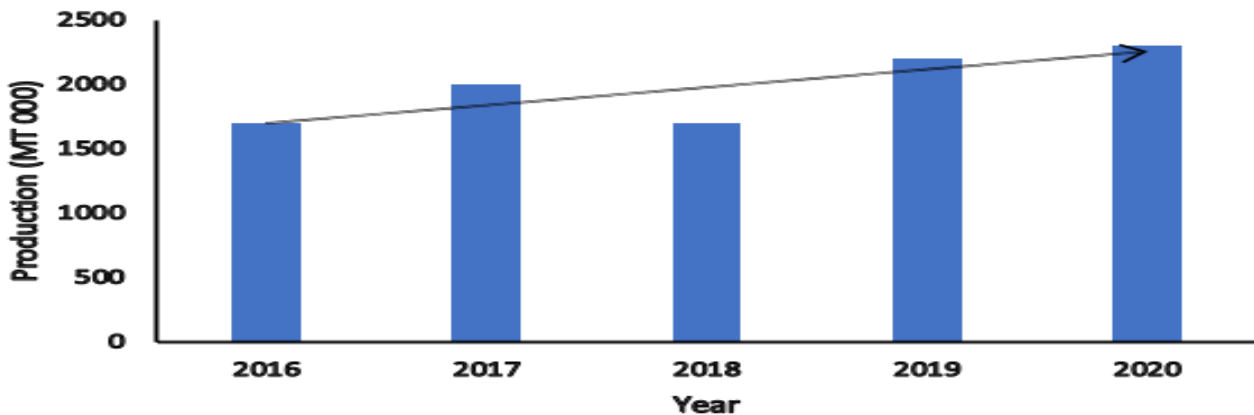


Figure 1.2. Saskatchewan oat production.

1.2.2 Potential of sawdust production in Saskatchewan

Approximately 350 million hectares of land in Canada is forest, making Canada the third largest in terms of forest area in the world (Statists Research Forestry in Canada 2022) as shown in Figure 1.3. The forestry industry is an important contributor to the country's economy. In 2020, the wood product manufacturing sector had a nominal GDP of approximately C\$12.6 billion, which makes it the most lucrative of the forestry industries (Statists Research Forestry in Canada 2022). The sawmill industry produces softwood and hardwood lumber, with sawdust and wood chips as waste products. Softwood lumber represented 98% of all lumber production in 2020 (Capuano 2023). Canada is one of the world's largest producers and exporters of softwood lumber. In 2020, 84% of its softwood lumber production was exported to the U.S. (Capuano 2023).

Saskatchewan has four big sawmills and several smaller sawmills. Approximately 1.3 million m³ (545 million board feet) of spruce–pine–fir (SPF) lumber are produced annually from these mills (Haykiri-Acma et al. 2016). The main wood production and consumption centres are in Meadow Lake, Prince Albert, and Hudson's Bay (Jackson 2022). Mistik Management Ltd., a forest management firm, owns two wood and forestry facilities: NorSask Forest Products and Meadow Lake Mechanical Pulp Inc. Between the two mills, they harvest the area's spruce, fir, pine, etc., and process about 1.3 million cubic meters of timber annually. The hardwood goes to Paper Excellence Meadow Lake Mill (a pulp and paper mill) and about half-a-million cubic meters of softwood comes to Meadow Lake Tribal Council (MLTC) (Jackson 2022). Each year, 56,000 tonnes of sawdust, bark, trimmings, and uneven chips from making lumber was continuously fed into the sawmill's 50-year-old beehive burner. It is essential to find an economically and

ecologically sound pathway to transform these residues into raw materials to produce valuable energy (Onyenwoke et al. 2023b).

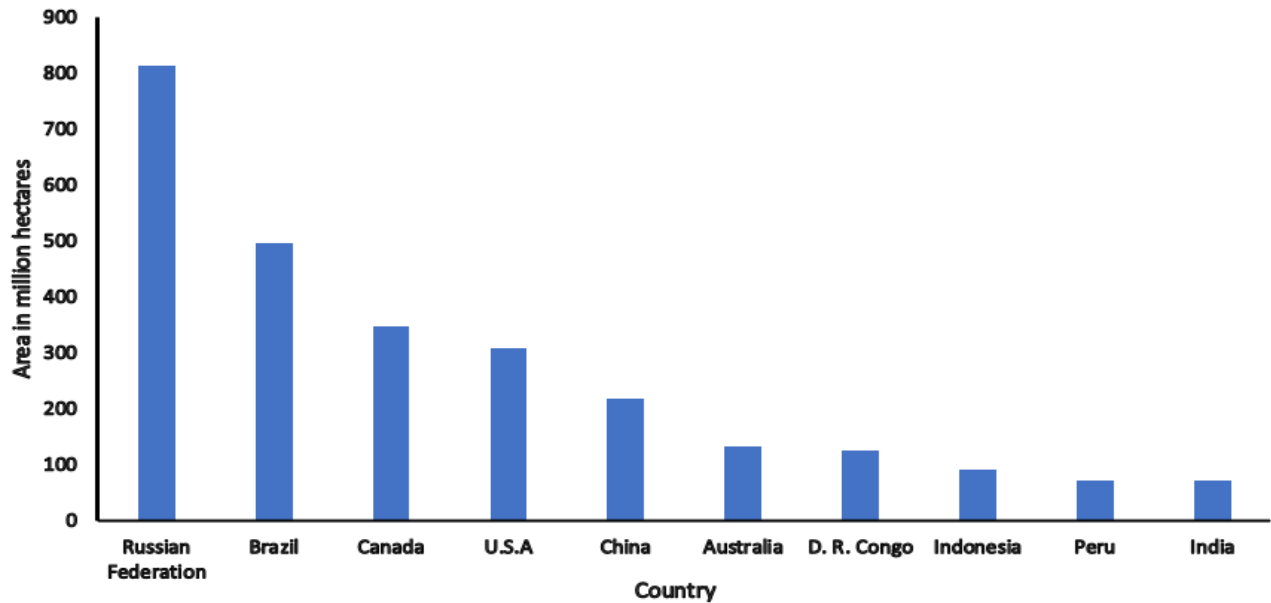


Figure 1.3. Ten countries with the most forest area in 2020.

1.3 Biomass Pretreatment

Pretreatment generally means the breakdown or modification of the naturally resistant biomass composition (lignin) that limits or prevents the accessibility of enzymes to the other biomass components (hemicellulose and cellulose). Pretreatment of biomass is necessary for the development of bioprocess and products. Over the years, many different pretreatments have been investigated on a wide variety of feedstock types and have been generally classified into biological, physical, chemical, and physicochemical pretreatments. The economics of pretreatment strategies must be considered when selecting a pretreatment method (Bin and Charles 2008).

Table 1.1 summarizes the advantages and disadvantages of various lignocellulose pretreatment methods. Torrefaction and steam explosion pretreatment options are of interest in this study. Chapter 2 discusses in detail these two pretreatment options.

Table 1.1. Advantages and disadvantages of various lignocellulose pretreatment methods. (Chen et al. 2017; Mupondwa et al. 2017; Capolupo and Faraco 2016; Iroba and Tabil 2013, Tumuluru et al. 2011).

| Pretreatment Methods | | Merits | Demerits |
|-----------------------------|---|---|--|
| Chemical | Ionic liquid | The product has no toxic gases, it has a large temperature range | The ionic liquids are expensive |
| | Alkaline hydrolysis | Uses relatively low temperature and pressure | Uses chemicals, requires a long residence time |
| | Acid hydrolysis | High sugar conversion, high delignification | Formation of inhibitors and corrosion of equipment |
| | Organosolv | Pure lignin, cellulose, and hemicelluloses were obtained | High-cost, recycling of solvent |
| Microbial | Wood-rotting fungi | No formation of toxic substance, little or no energy input, degrade lignin and hemicellulose | Low hydrolysis rate, capital intensive due to large area requirement |
| Physical | High-energy electron radiation | The degree of cellulose polymerization is reduced | Very expensive |
| | Mechanical extrusion | Less cellulose crystallinity and reduced hazardous process. | Requires high energy and has little or no effect on lignin |
| | Ultrasonic | Improve accessibility and reactivity of cellulose | Low delignification |
| | Microwave | Simple operation, energy-efficient, short time | High cost due to the high energy required |
| Physicochemical | Steam explosion | Less expensive, lignin transformation | Requires high temperature and pressure |
| | Ammonia fibre explosion | No inhibitory substance formed, increase accessibility | Not efficient for feedstock with high lignin |
| | Supercritical CO ₂ explosion | Increase surface area of cellulose, no formation of toxic/inhibitors, moderate process conditions | High cost |
| Thermochemical | Torrefaction | Simple operation, no chemicals required, short time | High cost due to the high energy required |

1.4 Technoeconomic Analysis of Cellulosic Biomass as Feedstock for Pellet Production

Technoeconomic analysis is a tool that examines parameters of interest in the research and development of a given technology with respect to economic performance. Despite the abundant supply of cellulosic biomass feedstocks (Kumarappan et al. 2009), the concept of cost-competitive pellet production is still largely unexplored. Biomass-based facilities have several limitations that have hindered their development. The nature and yield of the biomass produced from various feedstocks vary significantly, and this is one of the vital factors affecting their large-scale practical use in a biomass-based facility. The chemical structure of these feedstocks has been identified as a major constraint to cost-effective biofuel production (Mupondwa et al. 2017). Also, biomass production cost, feedstock transportation, and densification cost contribute significantly to the high expenditure component of the downstream production; therefore, the cost implications of biomass production, transportation, and pretreatment are limitations to the commercialization of pellet production.

Previous research has analyzed the economics of biomass-based energy from the aspect of generic models (Dassanayake and Kumar 2012; Kumar et al. 2008; Shahrukh et al. 2016). The cost of sawdust pellets production has been estimated by Mani et al. (2006b) to be US\$ 51/t at a plant capacity of 45 kt. Similarly, Shahrukh et al. (2016) performed a technoeconomic analysis of pellets produced from steam-pretreated biomass feedstock. They reported that the cost of production varies from US\$95 – 105/t for regular pellets and US\$146 – 156/t for steam pretreated pellets at a facility capacity of 190 kt and 250 kt, respectively. Pirraglia et al. (2013) carried out a techno-economic analysis of an industrial scale 100 kt/yr biomass torrefaction system in the US, reporting a production cost of \$199/t, and concluding that high capital cost represented the most sensitive factor affecting the net profit value (NVP), followed by biomass cost.

Sensitivity analysis provides information on how the variation of the economic and process assumed variables affect the economic performance and the sensitive factors of the design. The sensitivity analysis of a variable (such as the demand for a product, cost of raw material, product revenues, or energy costs) could guide future research and development efforts that, in turn, maximize the potential of the different biorefinery scenarios.

1.5 Research Gap

Among the processes involved in transforming lignocellulosic biomass to useful biofuel at large scale, pretreatment of feedstock is seen as the most critical as it may have a negative impact on the overall cost of biofuel production. Because of the high economic cost and severity of most of these pretreatment methods, their applications at industrial scale may be limited.

Currently there is an issue in the practicality of torrefaction systems and products for industrial applications. The problems concerning the promotion of systems and products and balancing energy efficiency and fuel quality remain unsolved. During torrefaction, numerous reaction products are formed. Their yield strongly depends on torrefaction conditions (temperature and time) and on biomass properties. These torrefaction gases contain a tremendous amount of energy.

Introducing a closed recycling system will be economically viable for torrefaction system, it is critically important that this energy be utilized efficiently in the system (torrefaction and densification).

1.6 Research Objectives

The main objective of this study is to investigate the effect of steam explosion and torrefaction pretreatment strategies to improve the accessibility of the energy potentials and enhance the densification process of sawdust and oat straw as feedstock for biofuel production.

The specific objectives of this work include:

1. To investigate steam explosion pretreatment of oat straw and sawdust to improve their quality as feedstock for biofuel pellets;
2. To investigate torrefaction pretreatment of oat straw and sawdust to improve their quality as feedstock for biofuel pellets;
3. To investigate the combined effect of pretreatment and binders on the quality of pellet formed; and
4. To carry out a comparative techno-economic analysis of pellet production from sawdust and oat straw using different pretreatment methods.

1.7 Organization of the Thesis

This thesis is organized and formatted in accordance with the guidelines for manuscript-based theses of the College of Graduate and Postdoctoral Studies, University of Saskatchewan. The thesis is composed of eight chapters, five of which are research manuscripts. Chapter 1 is the introduction and objective of this study. Chapter 2 has been presented during the Canadian Society of Bioengineering conference in July 2019. The manuscripts discussed in Chapters 3 (addressing specific objective 1), Chapter 4 (addressing specific objective 2), Chapter 5 (addressing specific objective 3), and Chapter 6 (addressing specific objective 4). In each manuscript-based chapter, the contribution of the Ph.D. candidate and the contribution of the

paper to the overall study are discussed in the manuscript. Chapter 7 is the general discussion and contributions to knowledge of this thesis, Chapter 8 is the summary, conclusions, and recommendations for future research. A list of the references in all the chapters is provided after Chapter 8. The appendix contains supplemental data.

1.8 Manuscript Content of the Thesis

The Ph.D. research presented in the manuscripts has resulted in discovering the best conditions to employ and the development of new approaches for steam explosion and torrefaction pretreatment of biomass for pellet production. In Chapter 2, a comprehensive literature review on pretreatment processing methods especially steam explosion and torrefaction is presented, including multiple biomass feedstocks such as forest and agricultural residues, processing characteristics, and product properties. The study also discusses the economics of densifying biomass, outstanding challenges, the potential for industrial applications of biomass-pretreated products, and global trends in biomass utilization. Steam explosion pretreatment of sawdust and oat straw to improve their quality as biofuel pellets was investigated in Chapter 3, where the effects of temperature, time, and moisture content on the mechanical properties of biomass pellets was carefully studied. This study evaluates the physical and mechanical characteristics of the pellets formed from steam treated sawdust and oat straw such as tensile strength, moisture uptake rate, bulk density, pellet density, and dimensional stability. In chapter 4, the effect of torrefaction on the physiochemical properties of white spruce sawdust for biofuel production was presented. This study evaluated the physiochemical characteristics of the pellets formed from torrefied sawdust such as tensile strength, moisture uptake rate, bulk density, pellet density and the scanning electron microscope (SEM) analysis. The combined effect of pretreatment and

binders on the quality of pellet formed from oat straw was illustrated in Chapter 5. Torrefaction liquid (TL) and sawdust were introduced during pelletization as binding agents in the pellet formulation. The chemical components and thermochemical analysis of the pellets were measured. These include ultimate analysis, proximate analysis, and high heating value. Finally, technoeconomic analysis of torrefaction and steam explosion pretreatment and pelletization of oat straw and sawdust plant was conducted in Chapter 6. The economic parameters such as total revenue produced by the plant and payback time are the focus that attract investors to the industry.

Chapter 2

Steam Explosion and Torrefaction of Forest and Agricultural Residues for Bioenergy/Biofuel Applications

A similar version of this chapter has been presented at the CSBE/SCGAB AGM and Technical Conference 2019 in Vancouver, BC, Canada and is available online as Abstract only:

- Onyenwoke, C. L.G. Tabil, and T. Dumonceaux. Steam Explosion and Torrefaction of Forest and Agricultural Residues for Bioenergy/Biofuel Applications. CSBE/SCGAB AGM and Technical Conference in Vancouver, BC, Canada. Identifier: CSBE19122.
<https://library.csbe-scgab.ca/all-publications/4915:steam-explosion-and-torrefaction-of-forest-and-agricultural-residues-for-bioenergy-biofuel-applications>.

Contribution of the Ph.D. Candidate

The Ph.D. candidate, Onyenwoke, C. collected the data, resources, and organized the manuscript, while Drs. L.G. Tabil and T. Dumonceaux revised and organized the manuscript. Planning of experiment/study was done by the Ph.D. candidate, Onyenwoke, C. and the supervisors Drs. L.G. Tabil and T. Dumonceaux.

Contribution of the Paper to the Overall Study

This paper provides information with regards to previous studies on steam explosion and torrefaction pretreatment and bioenergy production from oat straw and sawdust. It gives insight into the gaps in the research and how they could be filled. Therefore, a comprehensive literature review with the objectives of the study has been conducted and presented in this section.

2.1 Abstract

The use of fossil resources for energy and products has resulted in environmental, economic, and social issues and has encouraged intensive research on alternative raw materials for energy and chemical production. Alternative energy production can be achieved using various renewables such as wind, water, and sun, but industries based on sustainable materials, chemicals, and fuels depend on lignocellulosic biomass. Lignocellulosics are abundantly available, relatively widely distributed worldwide, and may alleviate the conflict in use between food and energy. The use of residues from agriculture, such as oat straw and forest residues for energy production addresses the food vs. fuel dilemma and adds value to existing crops. Pretreatment research has been focused on identifying, evaluating, developing, and demonstrating promising approaches that enhance the enzymatic hydrolysis of the pretreated biomass at lower enzyme dosages and shorter conversion times. Assessment of biomass pretreatment processes depends on a parameter called the severity factor, which is defined as the combined effect of temperature and duration of pretreatment. This study summarizes pretreatment processing methods, namely, steam explosion and torrefaction, multiple biomass feedstocks such as forest and agricultural residues, processing characteristics, and product properties. The study also discusses the economics of densifying biomass, outstanding challenges, the potential for industrial applications of biomass-pretreated products, and global trends in biomass utilization.

2.2 Introduction

The world is currently faced with the challenge of reducing dependence on fossil fuels and achieve a sustainable renewable energy supply (Tumuluru et al. 2011). As energy prices have fluctuated, and as concern about climate change has grown, companies and governments have

looked increasingly to bioenergy as an economical and environmentally friendly alternative to fossil fuels as well as for energy security (NRC 2016). Alternative energy production can be achieved using various renewables such as wind, water, and sun, but industries based on sustainable materials, chemicals, and fuels depend mostly on lignocellulosic biomass (Sarkar et al. 2012). Increasing the use of biomass for energy can help reduce greenhouse gas emissions and produce a competitive market with fossil fuels whose price is increasing steadily. The growing interest in biomass as a solid fuel includes combustion to produce steam electrical power and commercial plant uses as well as gasification to produce combustible gas and syngas. Lignocellulosics are abundantly available, relatively evenly distributed worldwide, and may alleviate the conflict in use between food and energy (Rabemanolontsoa and Saka 2016; Adapa et al. 2011). Oat is a crop known worldwide, and its production stands at about 25 Mt. Canada is one of the leading suppliers of oat globally. Oat continues to be a major cereal crop in Saskatchewan, with average annual production in recent years more than 1.5 Mt, on approximately 600,000 ha (1.5 million ac). Oat production in Saskatchewan has been steady. Recently, demand has been active from the United States food market, as well as the livestock market. The majority of oat trade is with the United States, with Canada being their major supplier (Sask. Govt. Oat Production and Markets Factsheet 2017) Agricultural crop residues such as oat straws are considered as suitable feedstocks for biofuel (Tavakoli et al. 2009).

The Canadian forest industry has a long history of using wood residues as a renewable energy source. Forest residues such as bark, sawdust, wood chips, branches, and treetops have become feedstocks for sophisticated steam and electricity cogeneration facilities, lumber drying kilns, and new bioenergy products such as wood pellets, syngas, cellulosic ethanol, biodiesel, bio-oil, biocarbon, and much more (CCFM 2018). Saskatchewan's main tree species are white spruce,

jack pine, black spruce, and trembling aspen. Saskatchewan has four large sawmills and a variety of smaller sawmills, with a combined annual production capacity of over 1.3 million m³ (545 million board feet) of spruce–pine–fir (SPF) lumber. White spruce (*Picea glauca*) is one of the most widely distributed boreal forest conifers in Canada. Its distribution in Saskatchewan stretches across the province’s entire boreal forest region. It can be found on moist, well-drained, silty soils. In Saskatchewan white spruce accounts for 16% of the total annual allowable cut and 28% of the coniferous annual allowable cut (Saskatchewan Forest Products Buyers’ Guide 2019).

The massive production of the crop and forest product results in a large amount of waste biomass residue; after grain harvesting, most crop residues are left on the field. Among the abundance of biomass, only a small fraction is utilized as animal feed, animal bedding, and mulching. Straw biomass is a suitable origin of biofuels and bioproducts (Kim and Dale 2004). The use of such carbon sources that are accessible in substantial quantities and could be utilized in a carbon dioxide neutral way is a sensible solution to major sustainability problems of society (Kim and Dale 2004).

Some of the characteristics of raw biomass materials compared to fossil fuel resources render raw biomass difficult to use on a large scale including low bulk density, high moisture content, hydrophilic nature, and low calorific value. These limitations greatly impact the logistics, storage, and final energy efficiency of biomass (Azargohar et al. 2019a; Medic et al. 2012; Haykiri-Acma et al. 2016). Several pretreatment strategies such as physical, chemical, physicochemical, and biological pretreatment methods have been utilized to overcome this barrier caused by lignocellulose matrix to cellulose degradation. Because of the high economic cost and severity of most of these pretreatment methods, their applications at industrial scale are very limited (Ade-Omowaye et al. 2001; Bagby 1982; Bazhal et al. 2003; Focher et al. 1998).

2.3 Lignocellulosic Biomass Composition

The composition of lignocellulosic biomass varies because of its source. Lignocellulosic biomass shows great potential as feedstock for pellet production.

2.3.1 Chemical structure and composition

Lignocellulosic biomass derived from agriculture and forestry, which includes agro-industrial residues, forest-industrial residues, energy crops, municipal solid waste, and other materials, are readily available bioresources to be used as feedstock for biorefineries that supplement oil refineries as sources of fuels and platform chemicals (Jönsson and Martín 2016). Cellulose, hemicellulose, and lignin constitute the major components of lignocellulosic biomass. About 75% of lignocellulose is comprised of polysaccharides (cellulose and hemicellulose), which can potentially be converted into monosaccharides for fermentation. Unlike cellulose, which has long unbranched fibrils entirely made up of glucose, hemicellulose is a branched polymer composed of various 5- and 6-carbon sugars (Agu et al. 2017; Marriot et al. 2016). Lignin acts as a binder which is covalently bound to and interlinked with hemicellulose and cellulose. While lignin is typically considered to be a barrier to the production of biofuels from the carbohydrate component of biomass, it also has energy value, and can be converted to a variety of value-added products (Karimi et al. 2013) The main source of lignocellulose is plant secondary cell walls, the thick, strengthening layer of the cell wall that is arranged inside the primary wall after cell elongation has terminated. The main constituents of plant secondary cell walls are hemicellulose, cellulose, and lignin, and these vary in proportion in different feedstocks. The composition of lignocellulosic material strongly depends on its source. There is a significant variation in the lignin and (hemi) cellulose content of lignocellulosic depending on its source, whether from hardwood, softwood, or grasses. The cellulose, hemicellulose, and lignin contents in basic

agricultural residues are listed in Table 2.1. The contents of the structural components are from various materials (Potumarthi et al. 2013).

Table 2.1. Lignocellulose composition of agricultural biomass. (Iroba and Tabil 2013; Sarkar et al. 2012; Sun and Cheng 2002).

| Agricultural residues | Cellulose (%) | Hemicellulose (%) | Lignin (%) |
|--------------------------------|----------------------|--------------------------|-------------------|
| Barley straw | 40 | 20 | 15 |
| Canola straw | 42.39 | 16.41 | 14.15 |
| Coastal Bermuda grass | 25 | 35.7 | 6.4 |
| Corn cobs | 45 | 35 | 15 |
| Corn stalk | 35 | 15 | 19 |
| Corn straw | 42.39 | 16.41 | 14.15 |
| Cottonseed hairs | 80-95 | 5-20 | 0 |
| Fresh bagasse | 33.4 | 30 | 18.9 |
| Grasses | 25-40 | 35-50 | 10-30 |
| Hardwood stems | 40-55 | 24-40 | 18-25 |
| Leaves | 15-20 | 80-85 | 0 |
| Newspaper | 40-55 | 25-40 | 18-30 |
| Nutshells | 25-30 | 25-30 | 30-40 |
| Oat straw | 41 | 16 | 11 |
| Paper | 85-99 | 0 | 0-15 |
| Primary wastewater solids | 15–20 | NA | 24–29 |
| Rice hull | 36 | 15 | 20 |
| Rice straw | 32.1 | 24 | 18 |
| Sawdust | 55 | 14 | 21 |
| Softwood stems | 45-50 | 25-30 | 25-35 |
| Solid cattle manure | 1.6-4.7 | 1.4-3.3 | 2.7-5.7 |
| Sorghum straw | 33 | 18 | 15 |
| Sorted refuse | 60 | 20 | 20 |
| Swine waste | 6 | 28 | NA |
| Switchgrass | 45 | 31.4 | 12 |
| Wastepaper from chemical pulps | 60-70 | 10-20 | 5-10 |
| Wheat straw | 30 | 50 | 15 |

2.3.1.1 Cellulose

Cellulose is the most abundant polysaccharide on earth; annually, about 75 billion tons of cellulose are produced and consumed (Potumarthi et al. 2013). Cellulose is one of the main components of wood and many other lignocellulosic materials and typically represents 40–50 % of woody biomass dry weight. Cellulose with $(C_6H_{10}O_5)_n$ formula is a linear polymer of glucose with a repeating unit of cellobiose (consisting of two molecules of D-glucose connected by $\beta(1 \rightarrow 4)$ glycosidic bonds) (Karimi et al. 2013).

In lignocellulosic material, the cellulose molecules are in the form of fibrils, and these fibrils are composed of microfibrils. Microfibrils, in turn, are composed of elementary fibrils that are further associated with hemicellulose and lignin. Each microfibril is suggested to contain approximately 36 glucose chains (Ding and Himmel, 2006). The microfibrils consist of three groups of glucose chains; true- crystal chains (core chains), subcrystalline chains (transition chains), and “sub- crystalline or noncrystalline” chains (surface chains) (Ding and Himmel 2006). The cellulose microfibrils are known to be independent, the ultrastructure of cellulose is mainly due to the presence of covalent bonds, hydrogen bonding and van der Waals forces. The hydrogen bonding determines the straightness of the chain within the cellulose microfibril, the order (crystalline) or disorder (amorphous) may be brought into the structure by interchain hydrogen bonds (Laureano-Perez et al. 2005). The true-crystal chains are the most resistant part of cellulose for chemical and biological hydrolysis.

The hydrolytic enzyme attack on cellulose depends on its structural features, which include: the surface area, the crystallinity of the cellulose, degree of polymerization, and the lignin seal surrounding the cellulose fibres, which leads to the structural resistance of cellulose (Jeoh et al.

2007). For the enzyme activity to proceed during hydrolysis, the interaction between the enzyme molecules and the surface of cellulose particles needs to be well established.

2.3.1.2 Hemicellulose

Hemicellulose, also known as polyoses, a heteropolymer of polysaccharides and polyuronides, can be found in almost all lignocellulosic materials along with cellulose. Polyoses are the linking material between cellulose and lignin. Unlike cellulose, hemicellulose consists of different monosaccharide units. Also, the polymer chains of hemicelluloses have small branches and are amorphous. Because of amorphous morphology, hemicelluloses are partially soluble or swell in water. The polysaccharide part of hemicellulose contains different polymers of hexosans (mannan, glucane galactan, and ramnose) and pentosans (xylan and arabaninose), while the polyuronide parts of the molecule contain hexuronic acids along with methoxyl, acetyl, and free carboxylic groups (Karimi et al. 2013; Potumarthi et al. 2013).

Among the different hemicellulose polymers, the most common are xylans and glucomannans, with xylans being the most abundant. Xylans are the main hemicellulose components of secondary cell walls constituting about 20–30% of the biomass of hardwoods and herbaceous plants (Gírio et al. 2010). Hemicelluloses in agricultural biomass like straw and grasses are composed mainly of xylan, while softwood hemicelluloses contain mostly glucomannan (Agbor et al. 2011). Also, the hemicellulose comprises, in a small amount, acetyl groups esterified to some OH groups of its different sugars. Hemicellulose is sometimes called xylan due to the abundance of xylose. Due to its branched structure, hemicellulose structure is not typically crystalline but rather amorphous. This makes this biopolymer more soluble in water and has a higher susceptibility to hydrolysis (Sánchez et al. 2013). The carbohydrate component of lignocellulosic biomass (cellulose and hemicellulose) can be converted into biofuels such as

ethanol by different microorganisms. Hemicelluloses, among other components of lignocellulosics, are the most thermo-chemically sensitive (Agbor et al. 2011; Hendriks and Zeeman 2009). Hemicelluloses in plant cell walls have been proposed to be reduced by 50% to increase digestibility since they are thought to 'coat' cellulose-fibrils (Agbor et al. 2011).

2.3.1.3 Lignin

As the use of lignocellulosic biomass has increased due to its potential for producing biofuels and bio-based chemicals, researchers have been challenged with developing a better knowledge of lignin structure, quantity, and potential uses. Lignin has always been seen as a waste product from the deconstruction of plant cell walls, in attempts to separate polysaccharides that can be hydrolyzed and fermented into fuel or other valuable commodities (Lupoi et al. 2015).

Apart from cellulose and hemicellulose, lignin is one of the most abundant polymers in nature and is present in the cell wall and extracellular matrix. It is an amorphous heteropolymer which has three different phenylpropane units (p-coumaryl, coniferyl and sinapyl alcohol) that are held together by different kinds of linkages. Lignin is sometimes referred to as glue between hemicellulose and cellulose components. The primary purpose of lignin is to give the plant structural support, which is the mechanical strength properties, impermeability, and resistance against microbial attack (makes the plant resistant against diseases and biodegradation by microorganisms) and oxidative stress (Agbor et al. 2011; Hendriks and Zeeman 2009; Karimi et al. 2013; Potumarthi et al. 2013).

Lignin is hydrophobic and highly resistant toward chemical and biological degradation. Lignin associates very closely with cellulose microfibrils, thereby making it a significant deterrent to enzymatic and microbial hydrolysis of lignocellulosic biomass (Hendriks and Zeeman 2009).

Chang and Holtzaple (2000) indicated that enzymatic digestibility is inversely correlated with lignin content.

Delignification (extraction of lignin by chemicals) process of biomass involves swelling, disruption of lignin structure, increases in internal surface area, and increased accessibility of cellulolytic enzymes to cellulose fibres. Although not all pretreatments result in substantial delignification, the structure of lignin may be altered without extraction due to changes in the chemical properties of the lignin. The pretreated biomass becomes more digestible than the raw biomass even though it may have approximately the same lignin content as non-pretreated biomass (Agbor et al. 2011).

2.3.1.4 Extraneous materials

Lignocellulosic materials contain varying quantities of other materials such as ash, proteins and pectins, depending on its source (Coyne et al. 2013). A large number of compounds are available in lignocelluloses, known as extraneous materials, which can be extracted by employing polar and nonpolar solvents. The composition and content of these materials vary among plant species. Extraneous materials are categorized into extractives or non-extractives based on their solubility in water (Karimi et al. 2013). The essential parts of extractive components are resins (fats, fatty acids, resin acids, and phytosterols), terpenes (isoprene alcohols and ketones), and phenols (residue and byproducts of lignin biosynthesis). The non-extractives parts of the extraneous materials are made up of inorganic components such as alkali earth carbonates, oxalates, starches, pectins, and proteins. Mainly, rice straw is covered by a layer of silica, which results in different behaviour in the pretreatments compared with other similar biomass, e.g., wheat straw (Binod et al. 2010; Karimi et al. 2013). Usually, the extraneous parts are not considered to play much of a role in lowering cellulosic biomass biological conversions, and fewer attempts have

been made in this direction. Although it might be intriguing, it would be difficult to see the effect of these components, since they are in large numbers and mostly present in low amounts (Karimi et al. 2013).

2.4 Pretreatment Technology for Lignocellulosic Biomass

Pretreatment technologies are aimed to increase accessibility to biomass by saccharifying enzymes and thereby the yields of fermentable sugars. In general, pretreatment methods fall into four different categories, including physical, chemical, physicochemical, and biological (Haghighi et al. 2013). There are many factors to consider for effective pretreatment of lignocellulosic biomass. Low capital and operational cost are one of the factors to be considered for the pretreatment process to be effective. Handling or preconditioning steps before the commencement of the pretreatment process should be reduced. It should produce no or small amounts of sugar and lignin degradation products that inhibit the growth of fermentative microorganisms or the action of hydrolytic enzymes, and it should require a low energy input or be performed in a manner that energy utilized could be used for other purposes such as secondary heating (Chandra et al. 2007; Agbor et al. 2011). Other factors such as regeneration/cost of catalyst, generation of higher-value lignin co-products, and obtaining hemicellulose sugars in the liquid phase form the basis of comparison of different pretreatment options (Galbe and Zacchi 2007; Mosier et al. 2005a; Agbor et al. 2011). All these factors are considered so that pretreatment results balance against their impact cost on downstream processing steps and the trade-off with operational cost, capital cost and biomass cost (Lynd et al. 1996; Mosier et al. 2005b).

Canadian research inventiveness in lignocellulosic bioconversion and pretreatment technologies involve collaborations extending over researchers from universities, Agriculture and Agri-Food Canada (AAFC), National Research Council, Natural Resources Canada, Environment Canada, and many other industrial organizations like Iogen, FPInnovations, NorSask, and Lignol Technologies. Many of these Canadian research innovations have also involved collaboration with the US Department of Energy, in particular Oak Ridge National Laboratory. These entities all have an interest in advancing the science and technology of biomass conversion (Mupondwa et al. 2017).

Pretreatment research has been focused on identifying, evaluating, developing, and demonstrating promising approaches that enhance the enzymatic hydrolysis of the pretreated biomass at lower enzyme dosages and shorter conversion times. Over the years, many different pretreatments have been investigated on a wide variety of feedstock types and have been generally classified into biological, physical, chemical, and physicochemical pretreatments. A combination of physical parameters, such as temperature or pressure, and biological or chemical treatments, can also be used in the pretreatment process. Some of the pretreatment methods have been reported to increase the digestibility of different raw materials efficiently; however, their viability at a larger scale represents a significant drawback. The mechanism for making cellulose more accessible to enzymes depends on the pretreatment employed and the nature of the raw material.

Theoretically, a simple pretreatment process produces a disrupted, hydrated substrate that is readily hydrolyzed, but the formation of sugar degradation products and fermentation inhibitors is avoided (Figure 2.1). The assessment of biomass pretreatment processes depends on a parameter called the "severity factor," which can be described as the combined effect of

temperature, acidity, pH, and duration of pretreatment. Studies on biomass pretreatment have used the severity factor for comparing pretreatment results even though an accurate measure of the severity is not provided. In other words, it is used for rough estimates (Galbe and Zacchi 2007).

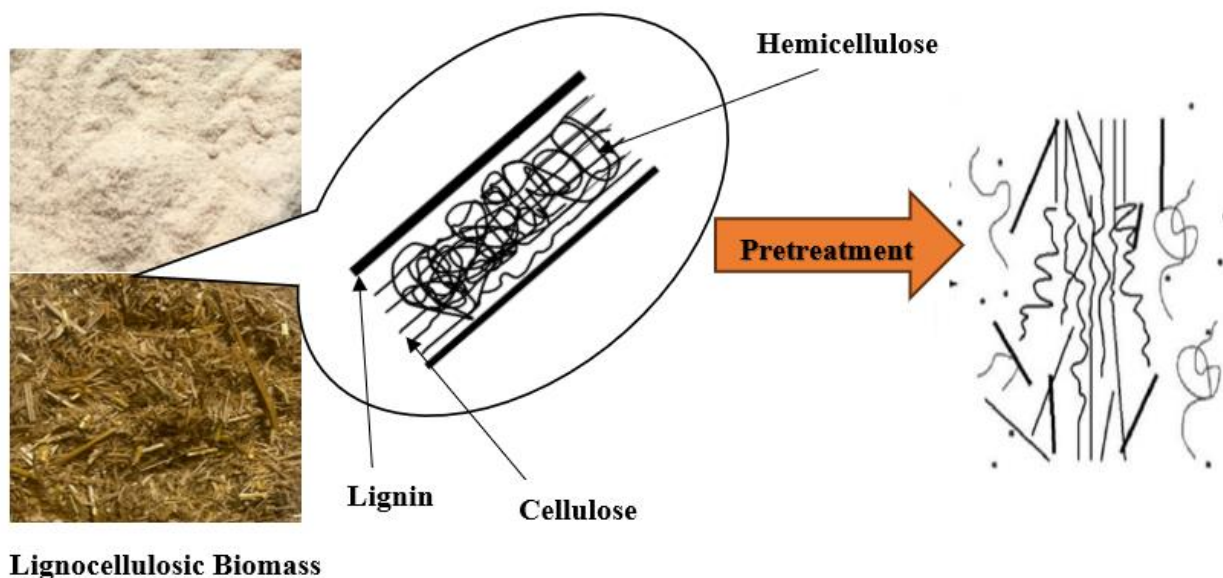


Figure 2.1. Schematic of goals of pretreatment on lignocellulosic material (redrawn from Luque et al. 2011).

Pretreatment can also be classified based on pH, namely, acidic, alkaline, and neutral pretreatments (Galbe and Zacchi 2007). This classification is focused on chemical pretreatment only. It does not encompass other pretreatment methods such as physical or biological pretreatments. Generalized classification of pretreatment methods groups them into; physical, chemical, biological, and multiple (combinatorial) pretreatment. In combinatorial pretreatment methods, physical parameters such as temperature or pressure or a biological step are combined with chemical treatments and are termed physicochemical or biochemical pretreatment methods.

Combinatorial pretreatment strategies are generally more effective in enhancing biomass digestibility, and often employed in designing leading pretreatment technologies (Agbor et al. 2011).

2.4.1 Physical pretreatment

The crystallinity of cellulose prevents the disruption of lignocellulose material. The basic step to disrupt biomass crystallinity is by size reduction. Several studies reported how biomass particle size distribution influences biomass conversion to biofuel (Cardoso et al. 2013; Drieimeier et al. 2011; Zhang et al. 2013; Putro et al. 2016). Particle size reduction increases the accessible surface area of biomass particles and facilitates inter-particle bonding. Size reduction increases the specific surface area of biomass and reduces cellulose crystallinity and the degree of polymerization; however, it also depends on biomass characteristics (Saadaoui et al. 2013; Adapa et al. 2011; Iroba et al. 2014a). The energy requirement for mechanical comminution of biomass materials is dependent on the final screen size of the biomass particle, characteristics, and the moisture content of biomass (Barakat et al. 2013; Agbor et al. 2011; Sarkar et al. 2012; Kumar et al. 2009; Islam and Matzen 1988). The effect of steam explosion pretreatment on corn stover particle size for improving enzyme digestibility was studied by Liu et al. (2013). It was found that the quantity of byproduct was higher, and sugar recovery was lower for biomass particles with a larger size. However, sugar conversion and yield were higher during enzymatic hydrolysis. With the increase of particle size, specific surface area, as well as crystallinity decreases (Liu et al. 2013; Putro et al. 2016). Khullar et al. (2013) studied the effect of particle size on the enzymatic hydrolysis of pretreated *Miscanthus*. The highest total conversion of biomass was obtained according to particle size, starting with the smallest particle size (0.08 mm), then followed by the particle size of 2 mm, and finally the lowest conversion was achieved

at particle size of 6 mm (Khullar et al. 2013; Putro et al. 2016). Different types of physical pretreatment processes are used to improve the biodegradability of lignocellulosic biomass or enzymatic hydrolysis.

2.4.2 Torrefaction

Torrefaction is a thermal biomass pretreatment process, where biomass is thermally degraded in an oxygen-free environment under atmospheric pressure and at reaction temperatures of 200–300 °C (Chen et al. 2015a; Zhang et al. 2018). Torrefaction is also known as mild pyrolysis, which has been explored for the pretreatment of biomass to increase heating value and hydrophobicity.

Torrefaction reduces moisture content and increases the calorific value of the biomass, which increases the density of biomass energy. Torrefaction is a feasible process for improving the properties of biomass as fuel (Sadaka and Negi 2009; Tumuluru et al. 2011). Zwart et al. (2006) compared alternative biomass to liquid routes, which include chipping, torrefaction, pelletization, and pyrolysis. The most efficient and commercially feasible way was found to be based on torrefaction, followed by pyrolysis and pelletization (Kumar and Sharma 2017). The first heating of biomass during the torrefaction process, makes the unbound water evaporate. Continuous heating leads to the removal of bound water through the process of chemical reactions (Tumuluru et al. 2011).

It is assumed that during the thermo-condensation process, most of the bound water is removed, and this occurs above 160 °C when the formation of CO₂ begins (Zanzi et al. 2022). Exothermic reaction and decomposition of the hemicellulose begin at further heating between 180–270 °C, which causes the biomass colour to change due to evaporation, CO₂, and a large quantity of acetic acid and phenols. The enthalpy of these compounds is relatively low, leading to an

increase in the energy density of the biomass (Zanzi et al. 2022). The process becomes completely exothermic at temperatures higher than 280 °C, causing a significant increase in the quantity of CO₂, phenols, acetic acid, and other higher hydrocarbons produced (Zanzi et al. 2022; Tumuluru et al. 2011).

2.4.2.1 Principles of torrefaction

Torrefaction of lignocellulosic materials given their distinct compositions and structures, cellulose, hemicellulose, and lignin, generates different thermal decomposition characteristics. Hemicelluloses tend to decompose more than lignin and cellulose. The thermal decomposition temperature (TDT) of hemicellulose is the lowest among the three constituents at the range of 220 and 315 °C Figure 2.2. Cellulose decomposes at temperatures between 315 and 400 °C. Lignin gradually decomposes across temperatures ranging from 160 to 900 °C (Lu et al. 2012; Tumuluru et al. 2011; Chen et al. 2015b). These decomposition reactions destroy hydroxyl groups (OH) and increase the density and specific heating value of the product. The destruction of hydroxyl groups also leads to creating a hydrophobic product (Hakkou et al. 2006).

In general, three performance indices have been used to evaluate torrefaction performance: solid yield, enhancement factor of higher heating value (HHV), and energy yield (Chen et al. 2015a; Zhang et al. 2018).

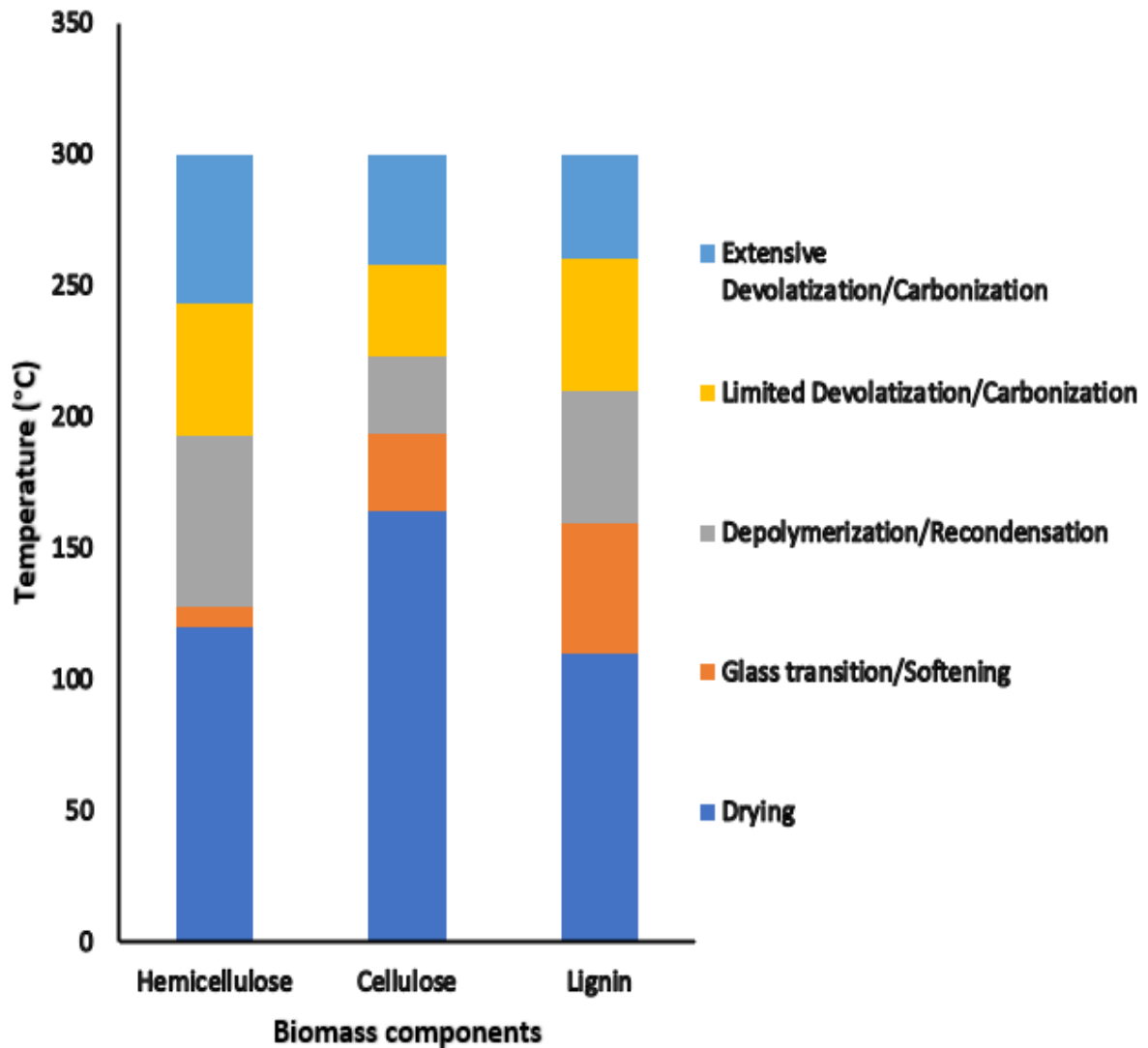


Figure 2.2. Physiochemical changes in biomass during torrefaction (redrawn from Bergman et al. 2005).

Torrefaction reduces the oxygen-carbon (O/C) ratio of biomass and reduces the energy required during milling, and simultaneously the sharp edges and splinters are removed in the milled powder. The torrefied biomass has also shown to have hydrophobic properties, which makes it storable. Torrefaction of biomass significantly influences the grindability and increases the uniformity of the final product quality (Tumuluru et al. 2011; Zhang et al. 2018).

2.4.2.2 Classification of torrefaction

Torrefaction processes, according to recent studies, can be performed in a dry or wet environment, and are referred to as dry and wet torrefaction, respectively (Figure 2.3). Biomass pretreatment occurs in the gas phase in the case of dry torrefaction, while for wet torrefaction, it occurs in the liquid or steam phase. Dry torrefaction can be classified into nonoxidative and oxidative torrefaction, while wet torrefaction can be categorized into dilute acid treatment and steam explosion. Although the torrefaction process can generally be classified into four different methods as shown in Figure 2.3, typically there are two bench-type heating measures for biomass torrefaction, including tube furnace torrefaction (named as conventional torrefaction) and microwave torrefaction (Ho et al. 2018; Gronnow et al. 2013). Nonoxidative torrefaction is the conventional method used to pretreat solid biomass. There is relatively little research carried out on the other three methods (Chen et al. 2015b).

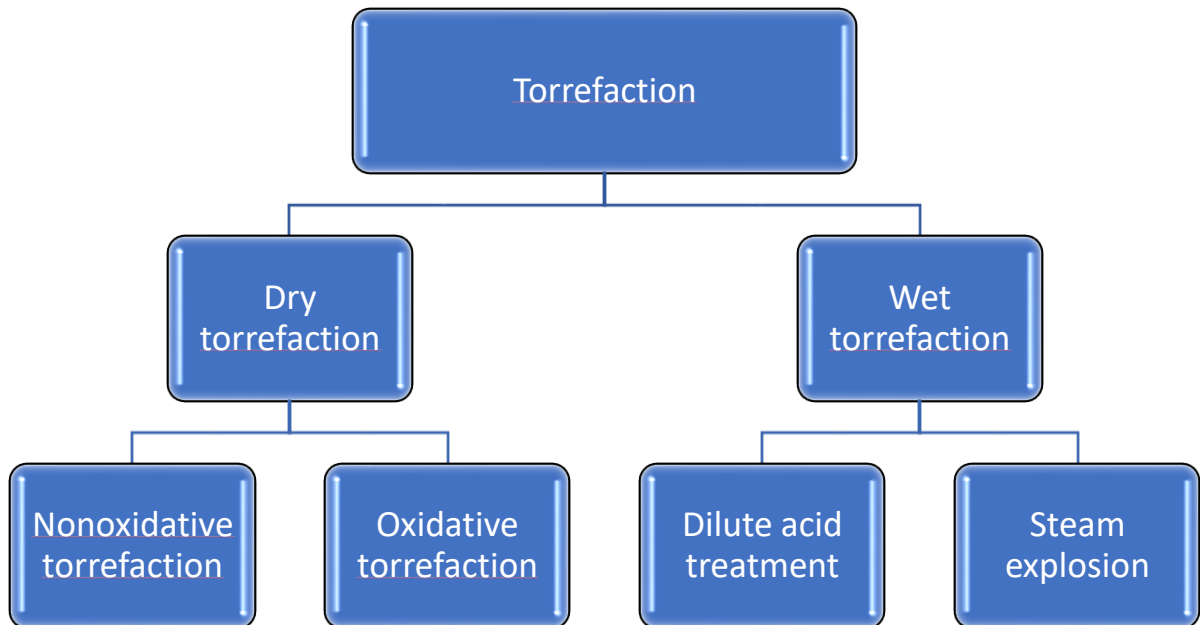


Figure 2.3. Classification of torrefaction.

2.4.2.2.1 Nonoxidative torrefaction

Nonoxidative torrefaction or conventional torrefaction is a thermal pretreatment technology to improve the characteristics of lignocellulosic biomass into an attractive biofuel (Saadon et al. 2014). The solid biomass upgrading process takes place in an inert or nitrogen atmosphere at a temperature of 200 – 300 °C for several minutes to several hours (Chen and Kuo 2010; Lu et al. 2012; Tran et al. 2013; Chen et al. 2015b). This thermal pretreatment method is an incomplete pyrolysis process, and it has also been called mild pyrolysis. The following parameters describe it: reaction temperature 200–300 °C, a heating rate less than 50°C/min, absence of oxygen, residence time less than 30 minutes at temperature greater than 200 °C, ambient pressure, and flexible biomass. The absence of oxygen in the reactor is necessary to avoid oxidation and ignition (Tumuluru et al. 2011; Chen et al. 2015b).

The two significant factors in implementing nonoxidative torrefaction biomass pretreatment are temperature and time (duration). The conventional biomass torrefaction can be classified into light, mild, and severe torrefaction based on pretreatment temperature, which features ranges of approximately 200-235, 235-275, 275-300 °C, respectively, as shown in Table 2.2 (Chen and Kuo 2010; Chen et al. 2015b). Light torrefaction reduces the moisture and decreases the molecular weight volatiles in biomass, hemicellulose which has the lowest thermal decomposition temperature among the biomass constituents is thus thermally degraded to a certain extent, while other biomass constituents are hardly or slightly affected which means the biomass loses little weight and its energy density increases very little (Rousset et al 2011; Chen et al. 2015b). Mild torrefaction intensifies hemicellulose decomposition and volatile liberation and affects the cellulose to an extent, the calorific value is increased, and the solid yield is reduced. During severe torrefaction, the hemicellulose is almost completely depleted, and the

cellulose is oxidized to a certain degree. Lignin, which has the highest thermal decomposition temperature among the biomass constituents, is slightly affected within the temperature range of torrefaction.

Table 2.2. A comparison of light, mild and severe torrefaction on biomass constituents
(Chen et al. 2015b).

| Torrefaction | Light | Mild | Severe |
|---------------------|--------------|----------------|----------------|
| Temperature (°C) | 200-235 | 235-275 | 275-300 |
| Hemicellulose | Mild | Mild to severe | Severe |
| Cellulose | Slight | Slight to mild | Mild to severe |
| Lignin | Slight | Slight | Slight |

2.4.2.2.2 Oxidative torrefaction

Oxidative torrefaction is a process of thermally upgrading the biomass in an oxygen-containing environment also under atmospheric pressure in a temperature range of 200–300 °C (Chen et al. 2015a). This is a promising method for industrial application due to its low requirements of equipment and technological conditions as well as the potential for cost reduction (Uemura et al. 2013; Zhang et al. 2019). This process is cost-effective such that additional operating expense is not incurred due to heating and supply of inert gas like the conventional torrefaction, air or a combustion flue gas is utilized as carrier gas (Chen et al. 2015b; Zhang et al. 2019).

Oxidative torrefaction not only involves ordinary torrefaction (e.g., devolatilization and thermal degradation of biomass), which occurs in non-oxidative torrefaction but also involves oxidation reactions (Uemura et al. 2013; Wongsiriamnuay et al. 2010; Chen et al. 2015b; Zhang et al. 2019). The oxidative reaction in the oxidative torrefaction is exothermic, implying that the heat

is thermodynamically generated from the torrefaction process and released to the surrounding (Chen et al. 2015b). With the presence of oxygen, the reaction rate of oxidative torrefaction is generally faster than that of conventional or non-oxidative torrefaction as a consequence of depolymerization and degradation of macromolecules in biomass, which shortens the torrefaction duration. Nevertheless, the ash content in oxidatively torrefied products may be somewhat higher when compared to those under nonoxidative torrefaction (Lu et al. 2012; Zhang et al. 2019).

Many studies have provided much valuable information on oxidative torrefaction and suggested that oxidative torrefaction is a potential route for upgrading biomass and enhancing its utilization in industry. However, it should be emphasized that detailed information concerning energy input and efficiency in an oxidative torrefaction system is still absent (Zhang et al. 2019).

2.5 Physico-Chemical Pretreatment

Chandra et al. (2007) reported that the combination of chemical and physical processes of pretreatment is called physico-chemical processes. One such process is referred to as steam explosion.

2.5.1 Steam explosion

Steam explosion is classified as a physiochemical pretreatment process, and it was first introduced and patented by Manson (1926), as an efficient process to defibrate wood into fibres (Mason, 1926). Steam explosion is also known to be a thermo-mechanicochemical pretreatment which results into the breakdown of the structural components lignocellulosic biomass by the process of heating, which forms organic acids during the process, and shearing forces leading to the expansion of the moisture (Jacquet et al. 2015). There are two stages involved in the steam-

explosion process: vapocracking and explosive decompression, which includes modification of the biomass components and improvement of the cellulose crystallinity index. These effects allow the lignocellulosic biomass structures to open and influence the enzymatic hydrolysis yield of the material.

Technically, the steam-explosion installation is composed of a steam generator that supplies a reactor, which is subjected to a sudden depressurization (Jacquet et al. 2015). During depressurization, the material is ejected from the reactor and is recovered in the explosion tank. Two factors influence the efficiency of the process: the retention time and the pressure. Several studies show that hydrolysis of the hemicellulose fraction is correlated to the residence time of the biomass in the reactor. Long residence time allows the complete hydrolysis of the hemicellulose fraction, which promotes downstream processes such as fermentation (Chornet and Overend, 1988).

Steam explosion pretreatment is carried out in a closed batch unit (Figure 2.4). The steam explosion system is composed of a steam generator (a): high pressure steam boiler, with the capacity to produce steam up to 3.45 MPa (500 psi). Custom made by HSI Hydro Steam Industries, Franklin Park (IL, USA), Model # STH-1640-30-4E. 30 KW. It also comprises a 203 mm diameter pressure vessel (chamber) with a volume and pressure capacity of 40 L (b) and up to 3.32 MPa (482 psi), respectively. It also consists of a cyclone (c) which opens at the bottom to allow discharge of treated samples. Pressure is controlled through the manual opening and closing of the main steam feeding valve (d) to the vessel and the time is controlled using a timer. Pressure could be monitored using a software pressure gauge connected to a laptop. The flow of pressure is controlled by opening and closing of the steam inlet valve using an electric/pneumatic valve.

Biomass is loaded through the ball valve located at the hopper (e) of the pressure vessel. The steam is generated inside the boiler, when the steam reaches the target reaction temperature, the ball valve is opened to allow the saturated steam to be transferred to the reactor chamber to treat the sample for a pre-specified period.



Figure 2.4. Steam explosion system at the Canadian Feed Research Centre, University of Saskatchewan.

Although hydrolysis products (mono- and oligosaccharides) are relatively stable in acidic conditions, they may undergo further reactions such as dehydration, fragmentation, and

condensation. These subsequent reactions generate a variety of products such as furfural, hydroxymethylfurfural, levulinic and formic acid, and various aromatic compounds, which are fermentation inhibitors. An increase in the retention time is correlated with a rise in the amount of these degradation products, which must be minimized. Pressure also plays a significant role in the process. The pressure is correlated to temperature and has an impact on the hydrolysis of cellulose fractions and the kinetics of the production of degradation products. Furthermore, the pressure difference between the reactor and the atmospheric pressure is proportional to the intensity of the shearing forces applied to the biomass during the explosive release (Sun et al. 2005).

2.6 Densification of Biomass

Biomass is difficult to utilize as a fuel in its natural form, as it is bulky, has a high moisture content, and is loose and dispersed. Biomass as an energy source and as a feedstock for biorefineries does not present easy, economical, and efficient transportation, handling, and storage characteristics due to large volume requirements (Tabil 1996; Mani et al. 2006a; Adapa et al. 2009). To mitigate this unfavorable condition is where densification gains extreme attention and importance. Biomass densification may be defined as the compaction or compression of biomass to eliminate inter- or intraparticle empty spaces. It also reduces the moisture content of biomass during compression. The main goal of densification is to increase mass per unit of volume of the biomass. Wood pellets are fuel made of wood shavings, bark, sawdust, and chips held together by compression or the addition of a binder. They have low moisture content and are easily transported over long distances. Densification is an important strategy for the biomass market, because it improves the convenience and accessibility of biomass due to the uniform shape and size, clean, stable fuel, enhances its volumetric calorific value, and they can be easily

adopted into the direct-combustion or co-firing with coal, gasification, pyrolysis, and biomass-based conversion reactors (Granada et al. 2002; Kaliyan and Morey 2009).

2.7 Summary

Lignocellulosic biomass is a renewable resource that can be used in the production of biofuels and platform chemicals in addition to bioenergy. As the most abundant carbon-neutral resource, using biomass as a replacement for fossil fuels can help mitigate environmental pollution. However, several physical, chemical, and structural factors can hinder the conversion of biomass into fuels and chemicals. Among the processes involved in transforming lignocellulosic biomass to useful biofuel at large scale, pretreatment of feedstock is seen as the most critical due to its negative impact on the overall cost of biofuel production. Because of the high cost and severity of most of these pretreatment methods, their applications at the industrial scale are very limited. Knowledge of the properties of biomass will be relevant in the allocation of feedstock to the appropriate end-use. An ability to determine these properties promptly is also necessary for the commercialization of biomass-based products. Thus, there is a need for high throughput methods and equipment in the characterization and monitoring of the feedstock as conventional methods have been destructive and laborious. However, the effect of various preprocessing and pretreatment methods on the lignocellulosic matrix is not well understood. Applications of preprocessing methods such as size reduction or increasing porosity, and pretreatment techniques such as steam explosion and torrefaction on agricultural biomass have demonstrated an improvement in pellet (compact) quality that can be linked to the changes in the lignocellulosic components and distribution.

Chapter 3

Investigation of Steam Explosion Pretreatment of Sawdust and Oat Straw to Improve their Quality as Biofuel Pellets

- A similar version of this chapter has been published in the journal *Energies*: Onyenwoke, C., L.G. Tabil, T. Dumonceaux, D. Cree, E. Mupondwa, P. Adapa, and C. Karunakaran. 2022. Investigation of steam explosion pretreatment of sawdust and oat straw to improve their quality as biofuel pellets. *Energies* 15(22): 7168. <https://doi.org/10.3390/en15197168>.

Contribution of the Ph.D. Candidate

The experimental work, data collection and analysis, original drafting, review and editing of the manuscript was done by the Ph.D. candidate, while Drs. L.G. Tabil, T. Dumonceaux, D. Cree, E. Mupondwa, P. Adapa, and C. Karunakaran, provided resources and editorial input. Drs. L.G. Tabil and T. Dumonceaux performed supervisory roles, while Drs. L.G. Tabil, T. Dumonceaux, D. Cree, E. Mupondwa also carried out the project administrative roles.

Contribution of the Paper to the Overall Study

This paper addressed the first objective of this study: to investigate the steam explosion pretreatment of sawdust and oat straw to improve their quality as biofuel pellets. Pretreatment of biomass prior to pelletization improves the quality of the pellets, and this study evaluates the physical and mechanical characteristics of the pellets formed from steam treated sawdust and oat straw such as measuring tensile strength, moisture uptake rate, bulk density, pellet density, and

dimensional stability. Also, the chemical components and thermochemical analysis of the pellets were measured. These include ultimate analysis, proximate analysis, and determination of high heating value. Furthermore, the results of this study demonstrated that steam explosion pretreatment of sawdust and oat straw resulted in a substantial increase in the mechanical strength of their respective pellets, which is relevant during transportation and storage. The result of this study can be implemented in the pellet industry.

3.1 Abstract

Steam explosion pretreatment of sawdust and oat straw under mild, medium, and severe conditions was conducted to improve the quality of pellets generated from these feedstocks. This work examined the effects of temperature, time, and moisture content on the mechanical properties of biomass pellets. From the analysis of variance (ANOVA) conducted, the p -values of the regression models for all the response variables (dimensional stability, tensile strength, and pellet density) studied were significant ($p < 0.05$), except for the pellet density of steam-pretreated oat straw pellets. The interaction of these three factors did not significantly affect the response variables of oat straw pellets. In addition, the higher heating value (HHV) of treated biomass increased up to a maximum of about 9.5% and 7% as compared with the non-treated sawdust and oat straw, respectively. In addition, an increment of about 3.6-fold and 3.1-fold in pellet tensile strength of steam-pretreated sawdust and oat straw was observed, respectively. Microstructural examination of the pellets from steam-pretreated biomass revealed that the material contained particles that were more closely bonded and featured a cemented surface with fewer pores when compared to particles from untreated oat straw and sawdust.

3.2 Introduction

Achieving a sustainable, viable renewable energy supply will reduce societal dependence on fossil fuels, but this remains a major challenge (Tumuluru et al. 2011). As the price of energy fluctuates, and concern over climate change has increased, bioenergy has emerged as an economically favorable alternative to fossil fuels that also provides environmental and energy security benefits (NRC 2019). Alternative energy generation can also be achieved using different kinds of renewable sources such as water, wind, and sun, but the industries based on sustainable materials, fuels, and chemicals depend mostly on lignocellulosic biomass (Sarkar et al. 2012). Accelerating the use of biomass-derived energy can reduce emissions of greenhouse gases and supply a competitive market with fossil fuel, the cost of which is increasing daily. Biomass can be used as a solid fuel by direct combustion to produce electrical power through steam generation, or through gasification, which produces both combustible gas and syngas. Lignocellulosics are copiously available, moderately evenly distributed worldwide, and may ease the conflict in use between energy and food (Rabemanolontsoa and Saka 2016). The use of agricultural and forest residues such as oat straw and sawdust for energy production relieves the pressure on food resources compared with the use of the more readily convertible starch-based biofuels, while also adding value to low-value crops and forest residues (Adapa et al. 2011). Oat is a worldwide crop, and its production stands at about 25 Mt (Saskatchewan Agriculture 2017). Canada is one of the leading suppliers of oat, and globally, oat trade makes up the majority. Oat is a major cereal crop in the Canadian province of Saskatchewan, with average yearly production in recent years of more than 1.5 Mt, on approximately 600,000 ha (1.5 million ac). Oat production has been steady in Saskatchewan. Recently, in the United States, food market oat demand has been active, as well as for the livestock market. Most of the Canadian oat trade is

with the United States, with Canada being their major supplier (Saskatchewan Agriculture 2017). In addition to the utility of oats as human food and agricultural animal feed, residues from oat production including straws are useful as biofuel feedstocks (Tavakoli et al. 2009)

Using wood residues as a renewable energy source has come a long way in the Canadian forestry industry. Forest residues such as bark, wood chips, branches, treetops, and sawdust have become feedstocks for electricity cogeneration facilities and advanced steam facilities. New bioenergy products have emerged such as syngas, cellulosic ethanol, wood pellets, biodiesel, biocarbon, and bio-oil for use in lumber drying kilns (CCFM 2018). Important tree species in Saskatchewan include white spruce, trembling aspen, black spruce, and jack pine. There are four large sawmills in Saskatchewan along with several smaller sawmills; the total annual production capacity of these mills is more than 1.3 million m³ (545 million board feet) of spruce–pine–fir (SPF) lumber (Saskatchewan Forest Products Buyers' Guide 2019). White spruce (*Picea glauca*) is a widely distributed boreal forest conifer in Canada. In Saskatchewan, this species is distributed across the expanse of the boreal forest region, mainly in moist, well-drained silty soils. Accordingly, white spruce makes up a major proportion of the total allowable annual harvest in Saskatchewan (16% of the total and 28% of the coniferous allowable cut) (Saskatchewan Forest Products Buyers' Guide 2019).

The massive production of the crop and forest products results in a great deal of biomass residue, and the majority of these residues are simply left in the field after grain harvest or in the forest, respectively. A very small proportion of this biomass is used, principally as agricultural animal feed and bedding, or as mulch. Straw biomass can be more completely utilized as a feedstock to produce biofuels and bioproducts (Kim and Dale 2004). The use of such abundant biomass sources that produce low net carbon dioxide emissions can be part of a solution to decreasing

greenhouse gas levels associated with the production and use of fossil fuel resources (Kim and Dale 2004).

Some of the difficulties associated with the exploitation of untreated biomass materials that can impede their large-scale application include low bulk density, high moisture content, hydrophilicity, and inherently low calorific value. These constraints impact the logistics, storage, and energy efficiency of these feedstocks (Azargohar et al. 2019a; Medic et al 2012; Haykiri-Acma et al. 2016). Several pretreatment strategies including physicochemical and biological pretreatment methods have been utilized to overcome this obstacle caused by lignocellulose matrix to cellulose degradation. As a result of the high economic cost and severity of most of these pretreatment application methods, their utilization on an industrial scale is very limited (Ade-Omowaye et al. 2001; Bagby 1982; Bazhal et al 2003; Focher et al. 1998).

Steam explosion is a physicochemical pretreatment process that breaks down lignocellulosic biomass by the application of high-pressure heat, which results in the formation of organic acid, and shearing forces causing moisture expansion and explosive decompression are the two stages involved in the steam-explosion process. These processes modify the biomass components through hydrolysis of hemicellulosic components (resulting in the release of mono- and oligosaccharides), alterations to the chemical structure of lignin, and enhancement of the crystallinity index of cellulose. These conditions allow the lignocellulosic biomass structures to unlock and can enhance the fermentable carbohydrate yield of subsequent enzymatic hydrolysis steps (Jacquet et al. 2015).

Biomass pretreatment modifies the structure of the biomass feedstock, which facilitates pelletization (Onu olughu et al. 2021; Sokhansanj et al. 2005). To improve the quality of biomass pellets, chemical binders combined with pretreatment are required, but these negatively impact

the total cost of densification (Mupondwa et al. 2017; Jiang et al. 2016). Several biomass pretreatments have been investigated to facilitate the pelletization of agricultural biomass. Douglas fir was subjected to high-pressure saturated steam treatment by Lam et al. (2012) to enhance its pellet quality. This revealed that the moisture absorption rate of pellets reduced from 0.0152 to 0.0125 mL/min, indicating an improvement in storability.

Retention time during steam explosion has a major effect on the amount of degradation of the products that are observed, which must be minimized. Moreover, pressure is another important parameter that is correlated to temperature and influences the kinetics of the production of degradation products and hydrolysis of cellulose fractions. Additionally, the difference between the atmospheric pressure and that of the reactor is proportional to the severity of the shearing forces applied to the biomass when the pressure is suddenly and explosively released (Sun et al. 2005). Much literature can be found related to the effects of pressure and retention time on steam explosion of biomass, but few previous studies have examined the effect of initial moisture content of the feedstock. The objective of this study is to fill this knowledge gap by determining the effects of initial moisture content of the feedstock during steam explosion pretreatment on the fuel and physiochemical properties of biomass.

3.3 Materials and Methods

3.3.1 Feedstock preparation

Oat straw and white spruce sawdust were used in this study. The oat straw was obtained from a black soil zone within the Rural Municipality of Douglas No. 436 around Maymont, SK, Canada. The variety of the oat straw is Morgan, harvested in mid-October 2019, swathed, and baled with a conventional combine. The white spruce sawdust was collected from NorSask Forest Products Inc. (Meadow Lake, SK, Canada) in June 2020. The samples were kept in a dry chamber to be

used for this study. Oat straw and white spruce sawdust samples had a moisture content of approximately 10% (w.b.) and 42% (w.b.) as received, respectively. Oat straw samples were further size reduced using a hammermill of sieve 3.2 mm. Samples were preconditioned to different moisture contents (25%, 35%, and 45% w.b.), by carefully spraying a calculated amount of water required for the targeted sample moisture content and gently stirring to obtain a homogeneous mixture. The preconditioned samples were stored in a sealed plastic bag in a conditioned room at 4 °C for a period of 48 h before determination of sample moisture content and usage for experimentation.

3.3.2 Steam explosion experiment

The experiment was conducted at the Canadian Feed Research Center (North Battleford, SK, Canada). Steam explosion pretreatments were carried out in a closed batch unit (Figure 3.1). The steam explosion system is composed of a 30-kW steam generator (HSI Model # STH-1640-30-4E) with a capacity to produce steam up to 3448 kPa (500 psi) (HSI Hydro Steam Industries, Franklin Park, IL, USA) (Figure S3.1a). It also comprises a 20.3 cm diameter pressure vessel (chamber) with a volume capacity of 40 L and pressure up to 3323 kPa (482 psi) (Figure S3.1b). It also consists of a cyclone that opens at the bottom to allow the discharge of treated samples (Figure S3.1c). Pressure is controlled through the manual opening and closing of the main steam feeding valve to the vessel and the time is controlled using a timer. Pressure was monitored using a software pressure gauge connected to a laptop. Pressure was regulated by opening and closing of the steam inlet valve using an electric/pneumatic valve.

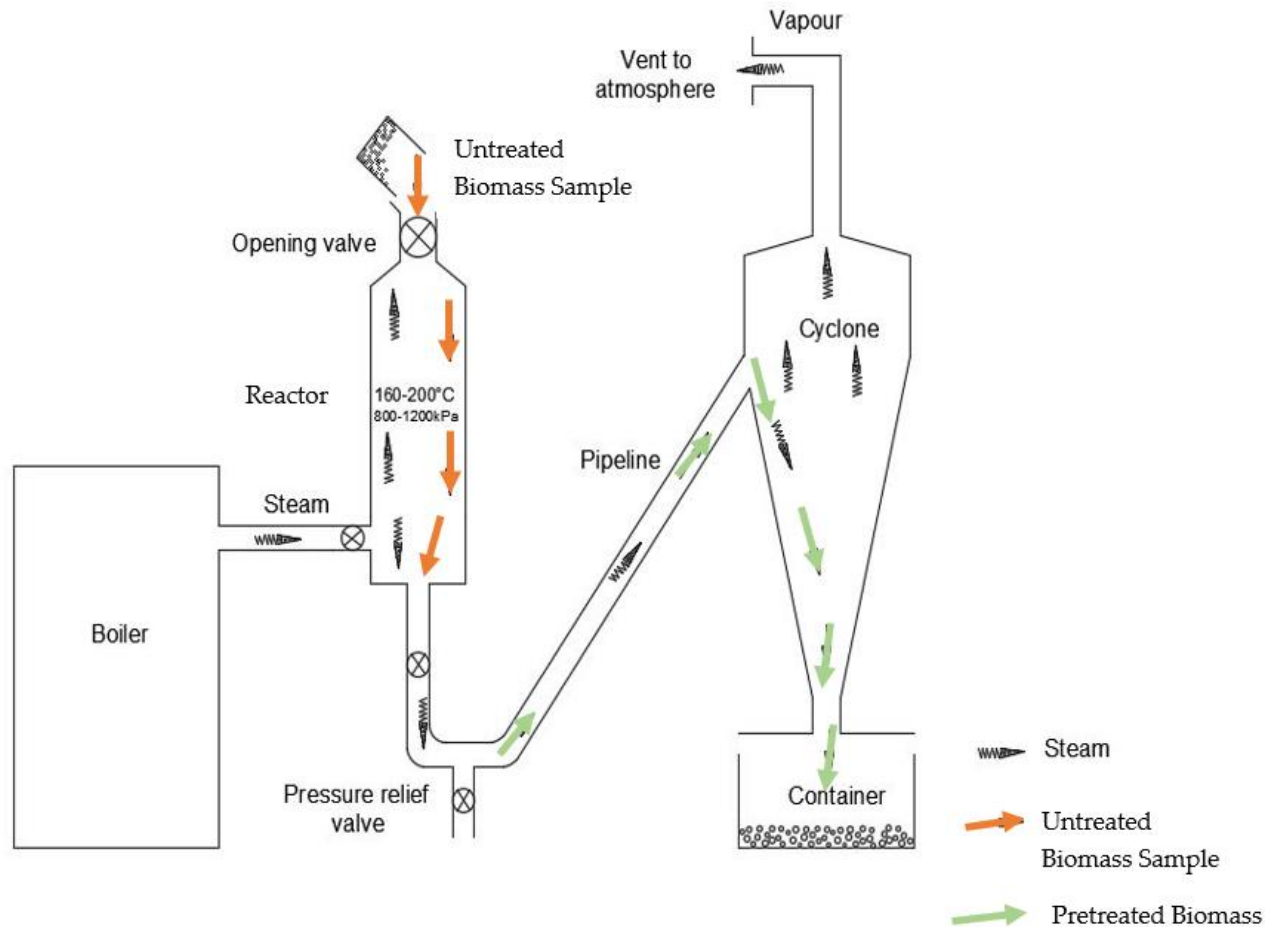


Figure 3.1. Schematic diagram of the steam explosion system used in the experiments.

The samples were steam exploded at three temperatures (160 °C, 180 °C, and 200 °C) and three samples of moisture contents (25%, 35%, and 45% w.b.) for three retention times (5, 7, and 9 min). These conditions were chosen based on the severity of operating conditions, that is, mild, medium, and severe steam explosions. Approximately 1000 g of biomass was loaded through the ball valve located at the hopper of the pressure vessel. The steam was generated inside the boiler, and when the steam reached the required reaction temperature, then the ball valve was manually opened to let the saturated steam be transferred to the reactor chamber (vessel) to treat the sample for a certain period. Various emissions to the atmosphere are considered. All experiments were conducted in three replicates.

3.3.3 Experimental design of steam explosion pretreatment

A factorial design of three factors, temperature (°C), retention time (min), and sample moisture content (%), at three levels was investigated. All experiments were conducted in three replicates. Analysis of variance (ANOVA) was conducted to statistically evaluate differences in the means of all the response variables (dimensional stability, pellet tensile strength, and pellet density)

3.4 Pelletization of the Samples

Pelletization of steam-exploded samples and untreated samples was carried out using a single pellet unit firmly fixed to an Instron tester (Model No. 3366, Instron Corp., Norwood, MA, USA) preset at 4000 N maximum downward force to produce the pellets. The assembly consisted of three parts: (1) a cylinder, (2) a heating tape looped all over the external of the cylinder, and (3) a piston. The body of the cylinder was heated up to about 95 °C by turning on the electric power; during that time, a compressive pressure of approximately 126 MPa at a rate of 50 mm/min was provided to the plunger for a holding time of 1 min, then the pellets were ejected and cooled. Approximately 0.70–0.75 g of the samples, at a moisture content of about 8.5% (w.b.), were introduced into the cylindrical die which was then compressed by the plunger to make pellets. Approximately ten pellets were produced for each pretreatment condition, then stored separately in an airtight plastic container for other experiments.

3.4.1 Pellet unit density and dimensional stability

After producing the pellet, its unit density was determined. The unit density is described as the ratio between the mass of a pellet and its volume. A digital vernier caliper was used to measure the diameter and length of individual pellet in this study, with an accuracy of ± 0.01 mm; on the other hand, a digital weighing balance was used to measure single pellet weight with an accuracy of ± 0.001 g. The dimensional stability, which is described as the volumetric difference in percent

after producing pellets (V_0) and after 14 d of storage (V_{14}), was computed as given in Equation (3.1)

$$\text{Dimensional stability (\%)} = \frac{V_{14} - V_0}{V_0} \times 100 \quad (3.1)$$

3.4.2 Diametral compression test

The tensile strength of the sawdust and oat straw pellets were evaluated by diametral compression test conducted using the Instron tester. Resistance to failure and dust generation because of handling (transportation and storage) is a good sign of the tensile strength of pellets. Kashaninejad and Tabil (2011) performed a similar test using the same equipment to determine the tensile strength of pellets. In this study, a diamond-cutting wheel attached to a Dremel rotary tool (Robert Bosch GmbH, Stuttgart, Germany) and a scalpel were used to cut the pellets diametrically into 2.5 mm specimens. The specimen was placed on its edge on the lower plate of the test rig in the Instron tester, which was padded, and the upper plunger provided a compressed 1000 N load cell at a crosshead speed of 1 mm/min until failure happened. The tensile strength of pellets was calculated using Equation (3.2).

$$\delta_x = \frac{2F}{\pi dl} \quad (3.2)$$

where δ_x is tensile strength (Pa); F is fracture load (N); d is specimen diameter (m); and l is specimen thickness (m).

3.4.3 Moisture absorption

The moisture absorption test of pellets from steam explosion treated and untreated oat straw grinds and white spruce sawdust were carried out after densification to investigate their water uptake. In this study, moisture absorption analysis was conducted in a controlled environment

chamber (Espec SH-641 Benchtop chamber, ESPEC Corp., Osaka, Japan) by estimating the overall moisture uptake of various samples under 90% relative humidity at a temperature of 25 °C. A worst-case scenario was assumed while selecting the conditions for the analysis. Pellet samples were left in the controlled environment for approximately 72 h until the moisture content was constant to evaluate the equilibrium moisture content (EMC). Other researchers used similar conditions for the calculation of EMC of biomass or their pellet form (Lam et al. 2012; Li et al. 2011; Azargohar et al. 2019b).

3.5 Characterization of Raw Material and Pellets

The untreated and steam-treated oat straw and sawdust pellets and grinds were characterized to rate the effects of each treatment.

3.5.1 Moisture content and solid yield

The samples moisture content as received was determined using ASAES358 (2012); about 25 g of samples were oven-dried at a temperature of 103 °C for 24 h. In addition, the grind samples' moisture contents were determined using AACC Standard 44-15A (2005), where 3 g of sample was oven-dried at 130 °C for about 90 min. The moisture content of samples after drying was also determined using the ASAES358 method. These moisture content tests were conducted in three replicates. Solid yield recovery of white spruce sawdust and oat straw were determined on dry solid basis as the percentage of samples recovered after the steam explosion experiment.

3.5.2 Particle size

Determination of the particle size was performed using the ASAE S319 sieving method (Adapa et al. 2011; Iroba et al. 2017; Satpathy et al. 2013; ASABE 2006). A Ro-Tap mechanical sieve shaker (W.S. Taylor Inc., Mentor, OH, USA) was used for this experiment. The selection of

sieve series was based on the variety of particle sizes in the samples. In this work, U.S. sieve numbers 16, 20, 30, 50, 70, and 100 (sieve mesh opening: 1.190, 0.841, 0.595, 0.297, 0.210, and 0.149 mm, respectively) were used. Approximately 100 g of steam-treated and untreated biomass samples were examined in triplicate for particle size distribution. As indicated in the ASAE Standard S319, the sieve shaker was allowed to operate for about 10 min. Equations (3.3) and (3.4) were used to calculate the geometric mean diameter (d_{gw}) and standard deviation of particle diameter (S_{gw}), respectively.

$$d_{gw} = \log^{-1} \left[\frac{\sum_{i=1}^n (W_i \log d_i)}{\sum_{i=1}^n W_i} \right] \quad (3.3)$$

$$S_{gw} = \frac{1}{2} d_{gw} \left[\log^{-1} S_{\log} - (\log^{-1} S_{\log})^{-1} \right] \quad (3.4)$$

where d_{gw} is the geometric mean diameter of particles by mass in mm; n is the number of sieves + 1 pan; d_i is the nominal sieve aperture size of the i th sieve in mm; W_i is the mass on the i th sieve in grams; S_{\log} is the geometric standard deviation of log-normal distribution by mass in the common (base 10) logarithm.

3.5.3 Bulk and particle density

Bulk density of treated and untreated oat straw grinds and white spruce sawdust was measured using a standard 0.5 L cylindrical cup (SWA951, Superior Scale Co. Ltd., Winnipeg, MB, Canada). The samples were introduced to the center of the cylindrical container through a funnel. Material flowing through the funnel was aided by stirring the grind with a steel roller. Excess sample at the top of the cylindrical cup was removed using a steel roller in a zig-zag pattern after

completely filling it. The bulk density of treated and untreated oat straw grinds and white spruce sawdust in kg/m^3 were determined by calculating the mass per unit volume.

Particle density of treated and untreated oat straw grinds and white spruce sawdust were measured using a gas displacement pycnometer (AccuPyc 1340, Micromeritics Instruments Corp., Norcross, GA, USA) at a temperature of 22 ± 0.9 °C. Data were acquired in replicates of 10 for an individual sample.

3.5.4 Elemental analysis

The amounts of carbon, hydrogen, nitrogen, and sulfur (CHNS) present in the samples was determined using the PerkinElmer Elementar CHNS analyzer (Vario EL III, Elementar Americas Inc., Ronkonkoma, NY, USA). The oxygen content in the sample was calculated by difference. Firstly, the equipment was calibrated using sulfanilic acid as a standard. Then, 4–6 mg of the samples were collected in an aluminum foil container and combusted for analysis.

3.5.5 Higher heating value and ash content

A bomb calorimeter (6400 Automatic Isoperibol, Parr Instrument Company, Moline, IL, USA) was used to determine the higher heating value (HHV) of the steam-exploded treated and untreated pellets. Approximately 0.5 g of the samples were burnt in an oxygen-filled metal cylinder submerged in a known volume of water, all held within a thermally insulated chamber. The test was conducted in three replicates for all samples.

3.5.6 Microstructural examination

Scanning electron microscope (SEM Phenom-World, Eindhoven, The Netherlands), together with a stereoscope (Wild M₃Z, Wild Heerbrugg, Gais, Switzerland) having a magnification of 200×, paxcam3 camera (Midwest Information Systems, Villa Park, IL, USA), and Intralux 500

light source, was used to examine the surface morphology of the ground biomass and pellet. A scalpel was used to cut the pellet sample longitudinally and coated with gold (to make sample conductive) to give a gold layer of 10 nm thickness employing a vacuum sputter coater (Q150T ES, Quorum Technologies, Sussex, UK).

3.6 Results and Discussion

3.6.1 Moisture content and solid yield

After steam explosion pretreatment, the moisture content of oat straw samples was between 69.4% and 80.4% (w.b.), while white spruce sawdust samples ranged from 68.5% to 84.4% (w.b.). Samples that were treated at 200 °C had a mean moisture content that was significantly ($p = 0.05$) higher than samples treated at 160 °C and 180 °C. In addition, the average moisture content of samples treated for 9 min was significantly ($p = 0.05$) higher than samples treated for 7 min and 5 min. This is a result of the longer duration the samples remained in the reactor, thereby allowing for more absorption of moisture. Adapa (2011) reported an average moisture content of 76.7% (w.b.) for oat straw when the digester was operated at 180 °C for 4 min. The initial moisture content of the sample is a determining factor in the severity of steam explosion pretreatment, since it affects the ability of heat and chemicals to permeate the biomass (Cullis et al. 2004). The moisture content of steam-exploded samples after drying was between 7.8% and 9.2% (d.b.) for oat straw, while dried sawdust samples ranged from 8.1% to 10.4% (d.b.). Table S3.1 shows the severity factor as determined using the equation given by Chornet and Overend (1987). In the current work, solid yield of white spruce sawdust samples varied between 61% and 85%, while that of oat straw samples varied between 72% and 90%. Lam (2011) reported a solid yield range between 51% and 84% when Douglas fir was steam exploded at 200 °C and 220 °C, respectively. The mean solid yield was significantly ($p = 0.05$) lower for samples treated

at 200 °C, compared to samples treated at 180 °C and 160 °C. In the current study, it was observed that the loss of fine particles during drying amounted to less than 1 g, which corresponds to under 5% of the sample weight. In a similar study, Kobayashi et al. (2009) reported 50% lower losses of solid yield for woody biomass from compressed hot water treatment at 240 °C. Normally, losses are due to inefficiencies in recovery of materials combined with the loss of volatiles during steam explosion (Emmel et al. 2003; Mani et al. 2004).

3.6.2 Particle size analysis

The average particle size of steam-exploded oat straw and white spruce sawdust was significantly smaller than that of the non-treated biomass. This may be because steam explosion pretreatment application disintegrates the lignocellulosic structure of the biomass (Iroba et al. 2017), resulting in decreased shear strength and easy grindability. Geometric mean particle sizes of 0.586 mm and 0.656 mm were recorded for untreated white spruce sawdust and oat straw, respectively. The particle size of steam-exploded biomass particles decreased with increasing severity of the steam explosion pretreatment. The average particle size of oat straw samples after steam explosion was between 0.565 mm and 0.456 mm, while the corresponding values of white spruce sawdust samples were between 0.512 mm and 0.389 mm. Adapa et al. (2011) observed similar results using steam explosion of oat straw. In their work, the average particle size of oat straw was reduced from 0.566 mm to about 0.367 mm when it was treated at 180 °C for 4 min. In addition, Lam (2011) found similar results using steam explosion of Douglas fir (*Pseudotsuga taxifolia*) wood chips treated at 200 °C and 220 °C for 5 min, resulting in a decrease in the mean particle size from 0.42 mm to 0.40 mm (200 °C) and 0.35 mm (220 °C). Increasing reaction severity led to an increase in percentage of smaller particles and fines in each sample. A Shapiro–Wilk test ($p > 0.05$) indicates that the particle size of the grinds from the various

reaction severities were approximately normally distributed. The particle sizes of the ground material should be normally distributed (Shapiro–Wilk test: $p > 0.05$) and should also have close to zero skewness with a lower peak than would be expected for the normal and wider distribution of data. This is reflected in negative Kurtosis values. The particle size distribution of the ground biomass from all screen sizes were skewed along the y-axis, and had a lower peak compared to a normal distribution. Figure 3.2 shows the normal size distribution of the untreated and treated sample particles at different severities. Wider particle size distribution is known to be suitable for densification (Mani et al. 2004). During densification, the finer particles rearrange and occupy the void space between the larger (coarse) particles, which results in the production of denser and more durable pellets (Tabil 1996).

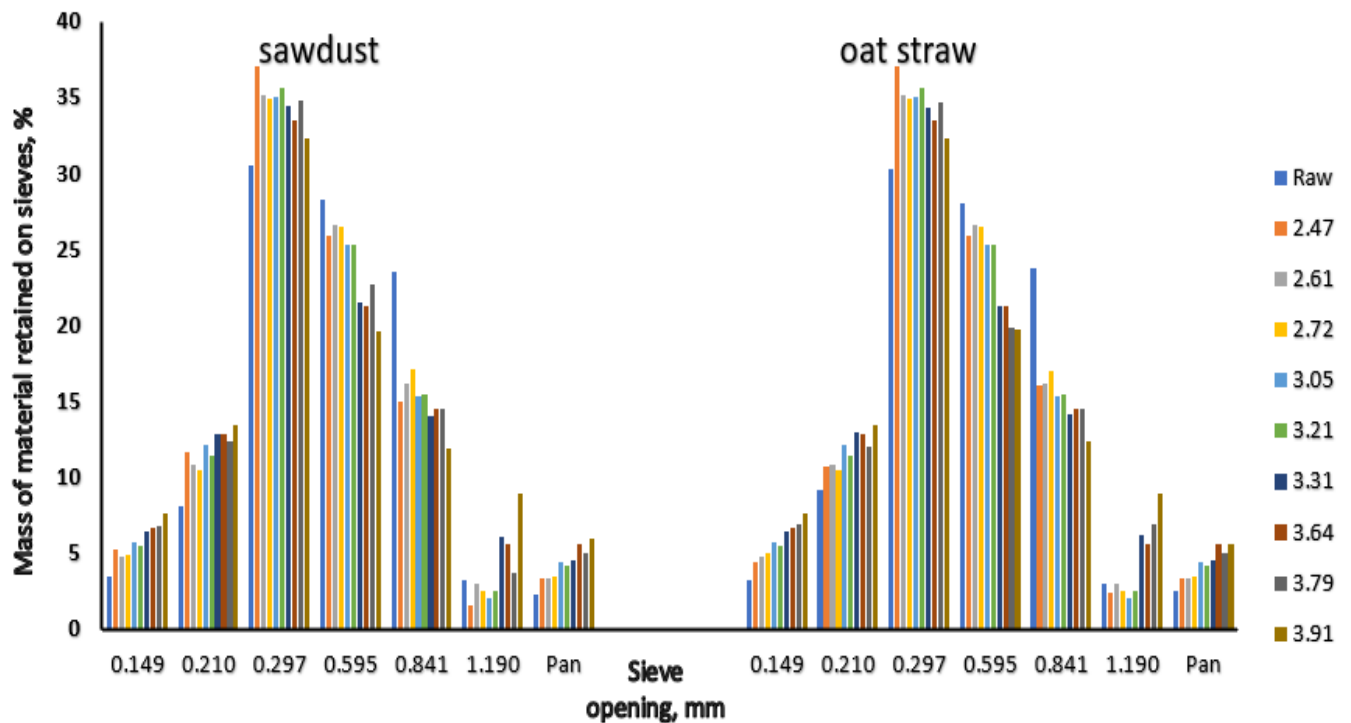


Figure 3.2. Size distribution of the untreated and treated sample particles at different severities.

3.6.3 Properties of pellets from non-treated and steam-exploded samples

The pellet tensile strength, dimensional stability, and unit density of the sawdust and oat straw from the untreated sample fuel pellets were 0.68 MPa and 0.34 MPa, 1011.02 kg/m³ and 1009.67 kg/m³, and 1047.95 kg/m³ and 1031.74 kg/m³, respectively, as shown in Table 3.1. When biomass is heated, lignin softens and melts, which provides thermosetting binder resin properties on the heated material and produces pellets with good dimensional stability and higher density (Tabil 1996). Pretreated samples had higher initial pellet densities compared to untreated ones. Pellet shrinkage was observed for the pretreated pellet samples after 14 d, while the untreated pellet samples overall expanded in the longitudinal directions and diametrically.

The results (Table 3.1) also show that as the treatment severity increased, the pellets treated by steam explosion showed a volumetric expansion after relaxation. Dimensional stability provides data on how stable the formed pellets can be during the transportation process and under storage conditions. Smaller values are associated with increased pellet stability. A positive value (negative value) indicates that there was either diametral or longitudinal expansion (contraction) after 14 d. The results from the current study are in accordance with previous research showing that steam treatment prior to densification improves the compressibility of wood particles and greatly decreases the internal stresses that accumulate during compression. In addition, the tensile strength increment with increasing severity could be attributed to the disruption of the lignocellulosic structure, which is associated with increased degradation of the hemicellulose and lignin, thereby acting as a natural binder through adhesion and thus increasing pellet tensile strength. The bulk density of untreated oat straw and sawdust was significantly higher than the sample treated by steam explosion (Table 3.1). This could be because steam explosion pretreatment application breaks up the arranged and compact lignocellulosic structure of

biomass, leading to lower bulk densities. Similar results have been obtained by other researchers (Adapa 2011; Shaw et al. 2009). The bulk density of treated oat straw ranged from 51.8 kg/m³ to 82.1 kg/m³. The bulk density of treated sawdust ranged from 97.4 kg/m³ to 129.9 kg/m³. The particle density of sawdust and oat straw treated by the steam explosion was significantly higher than the untreated sample. This is because steam explosion disintegrated the biomass into finer components, thereby disorganizing its lignocellulosic structure. The particle densities of steam-exploded oat straw and sawdust ranged from 947 kg/m³ to 1444 kg/m³ and 1172 kg/m³ to 1414 kg/m³, respectively.

Table 3.1. Properties of pellets from non-treated and steam-exploded samples.

| Temp (°C) | RT (min) | M.C. (%) | Sawdust | | | | | Oat Straw | | | | |
|--------------|-------------|-------------|---|-------------|------------------------------|---|---|---|-------------|------------------------------|---|---|
| | | | Pellet Density (kg/m ³) | D.S. (%) | Tensile Strength (MPa) | Bulk Density (kg/m ³) | Particle Density (kg/m ³) | Pellet Density (kg/m ³) | D.S. (%) | Tensile Strength (MPa) | Bulk Density (kg/m ³) | Particle Density (kg/m ³) |
| NT | - | - | 1047.95 | 8.93 | 0.68 | 158.00 | 1051.56 | 1031.74 | 11.07 | 0.35 | 127.87 | 882.23 |
| 160 | 5 | 25 | 1060.71 | 8.07 | 1.05 | 129.90 | 1172.45 | 1154.99 | 10.76 | 0.99 | 82.12 | 947.42 |
| 160 | 5 | 35 | 1090.33 | 7.77 | 0.92 | 123.60 | 1209.32 | 1054.34 | 10.02 | 0.92 | 79.92 | 996.35 |
| 160 | 5 | 45 | 1077.59 | 7.59 | 1.28 | 125.83 | 1278.78 | 1154.81 | 9.38 | 0.97 | 68.24 | 1005.34 |
| 160 | 7 | 25 | 1135.76 | 6.96 | 1.39 | 112.90 | 1280.97 | 1157.88 | 8.65 | 1.02 | 66.23 | 1030.24 |
| 160 | 7 | 35 | 1072.36 | 6.78 | 1.40 | 116.12 | 1281.76 | 1167.44 | 8.43 | 1.25 | 63.42 | 1001.22 |
| 160 | 7 | 45 | 1072.33 | 6.21 | 1.47 | 117.23 | 1300.12 | 1172.28 | 8.39 | 1.30 | 67.87 | 1092.75 |
| 160 | 9 | 25 | 1094.65 | 5.43 | 1.88 | 110.53 | 1352.22 | 1148.31 | 7.58 | 1.55 | 63.32 | 1069.45 |
| 160 | 9 | 35 | 1115.64 | 5.78 | 2.16 | 110.64 | 1367.17 | 1151.46 | 7.39 | 1.43 | 62.71 | 1110.66 |
| 160 | 9 | 45 | 1103.91 | 5.89 | 2.33 | 110.52 | 1370.23 | 1153.56 | 7.32 | 1.46 | 60.28 | 1153.47 |
| 180 | 5 | 25 | 1121.58 | 4.46 | 2.59 | 127.41 | 1356.34 | 1168.82 | 6.59 | 1.68 | 59.19 | 1166.33 |
| 180 | 5 | 35 | 1131.34 | 4.24 | 2.45 | 112.43 | 1373.56 | 1172.89 | 6.42 | 1.56 | 57.97 | 1174.90 |
| 180 | 5 | 45 | 1111.57 | 4.57 | 2.48 | 116.72 | 1377.78 | 1172.41 | 6.63 | 1.62 | 58.52 | 1180.88 |
| 180 | 7 | 25 | 1122.61 | 4.50 | 2.27 | 110.58 | 1381.96 | 1190.31 | 6.75 | 1.65 | 57.93 | 1224.32 |
| 180 | 7 | 35 | 1133.31 | 4.43 | 2.02 | 114.36 | 1389.43 | 1225.07 | 6.62 | 1.64 | 56.84 | 1217.12 |
| 180 | 7 | 45 | 1144.26 | 4.03 | 2.13 | 113.19 | 1391.19 | 1191.72 | 6.43 | 1.73 | 57.91 | 1263.89 |
| 180 | 9 | 25 | 1130.01 | 3.93 | 2.82 | 102.37 | 1400.66 | 1202.59 | 6.25 | 2.03 | 56.21 | 1283.35 |
| 180 | 9 | 35 | 1167.04 | 3.70 | 3.36 | 106.42 | 1402.34 | 1210.21 | 5.23 | 1.98 | 55.82 | 1257.67 |
| 180 | 9 | 45 | 1153.85 | 3.48 | 3.21 | 108.21 | 1407.33 | 1195.25 | 5.24 | 2.12 | 56.25 | 1263.55 |
| 200 | 5 | 25 | 1184.66 | 2.55 | 3.55 | 108.68 | 1406.20 | 1219.43 | 4.55 | 2.11 | 54.16 | 1359.98 |
| 200 | 5 | 35 | 1111.58 | 2.58 | 3.42 | 105.62 | 1402.59 | 1208.22 | 4.44 | 2.09 | 53.18 | 1367.23 |
| 200 | 5 | 45 | 1131.30 | 2.32 | 3.09 | 107.83 | 1405.34 | 1214.58 | 3.62 | 2.33 | 54.17 | 1328.86 |
| 200 | 7 | 25 | 1145.54 | 2.66 | 3.32 | 106.11 | 1408.32 | 1197.96 | 3.51 | 2.63 | 54.46 | 1383.43 |
| 200 | 7 | 35 | 1134.72 | 2.49 | 3.22 | 101.45 | 1406.87 | 1259.83 | 3.05 | 2.54 | 55.56 | 1371.55 |
| 200 | 7 | 45 | 1132.48 | 1.36 | 3.57 | 101.66 | 1407.90 | 1237.87 | 2.43 | 2.64 | 55.29 | 1408.34 |
| 200 | 9 | 25 | 1147.43 | 1.65 | 3.89 | 101.62 | 1411.47 | 1246.93 | 2.95 | 3.13 | 54.83 | 1411.23 |
| 200 | 9 | 35 | 1225.97 | 1.08 | 4.04 | 100.85 | 1415.62 | 1190.15 | 2.04 | 2.98 | 51.82 | 1420.45 |
| 200 | 9 | 45 | 1148.58 | 1.62 | 3.95 | 97.42 | 1414.05 | 1253.21 | 2.45 | 3.34 | 53.12 | 1444.09 |

Temp = temperature; RT = retention time; M.C. = moisture content; N.T. = untreated; D.S. = dimensional stability; n=3.

3.6.4 Elemental composition analysis

Table 3.2 shows that the pretreatment severity affects the elemental composition of biomass samples on a dry matter basis. The carbon, hydrogen, sulfur, and nitrogen content of the untreated oat straw and sawdust was 44.24%, 6.05%, 0.11%, and 0.41%, and 47.49%, 6.66%, 0.02%, and 0.01%, respectively. After the steam pretreatment, the carbon and the nitrogen contents of the samples significantly increased ($p < 0.05$) as the severity increased, while hydrogen content reduced. Some researchers reported similar results from their works on steam-pretreated biomass (Ferro et al; 2004, Iroba 2014; Yan et al. 2009). Angles et al. (2003) also reported that the process of steam explosion causes the lignin to carbonize and even condensed, which results in an increase in the carbon content of the pretreated samples. Generally, during steam explosion, smaller hydrocarbon molecules with low energy density volatilize, which increases the energy density of the residual carbon-rich solids. In this study, it was assumed that the reduction in hydrogen and oxygen is because of the formation of carbon dioxide and water. Hydroxyl groups are connected to the backbones of the biomass structural components. As the steam penetrates the biomass samples, debranch reactions continue and release acetic and uronic acids which hydrolyze hemicelluloses. Furthermore, as the severity of the treatment increases, amorphous cellulose could be partially depolymerized (Glasser et al. 1998; Qing et al. 2010; Selig et al. 2008). The ash content of the oat straw before steam explosion pretreatment is 5.32%, which is much higher than the sawdust (less than 1%). Generally, straw biomass samples have higher ash contents than woody biomass (Lam et al. 2011; Tooyserkani et al. 2013; He et al. 2020). As shown in Table 3.2, the ash content for sawdust and oat straw increased from 0.14% to 0.61% and 5.32% to 6.49%, respectively, as the severity factor increased. The increase in ash content after steam explosion pretreatment is owing to the loss of other components of biomass

as also reported by some researchers (Tooyserkani et al. 2013; Iroba et al. 2014b). Although Han et al. (2010) reported a reduction in ash content from 1.6–4%, this may be due to the removal of ash during the release of steam.

Table 3.2. Elemental composition and final moisture content of steam-exploded and non-treated biomass grinds (dry matter basis).

| C | R | M.C. (%) | C (%) | H (%) | N (%) | S (%) | O (%) | Ash (%) | H/C | O/C |
|-----------|------|-------------|--------------|-------------|-------------|-------------|-------|---------|------|------|
| Sawdust | NT | 8.69 ± 0.16 | 47.47 ± 0.46 | 6.66 ± 0.12 | 0.11 ± 0.02 | 0.21 ± 0.01 | 45.41 | 0.14 | 0.14 | 0.96 |
| | 2.47 | 8.62 ± 0.12 | 47.96 ± 0.23 | 6.49 ± 0.07 | 0.15 ± 0.01 | 0.20 ± 0.01 | 44.91 | 0.29 | 0.13 | 0.94 |
| | 2.61 | 8.60 ± 0.13 | 47.98 ± 0.18 | 6.51 ± 0.11 | 0.21 ± 0.02 | 0.23 ± 0.01 | 44.78 | 0.29 | 0.13 | 0.93 |
| | 2.72 | 8.55 ± 0.26 | 48.24 ± 0.21 | 6.48 ± 0.06 | 0.26 ± 0.10 | 0.18 ± 0.02 | 44.54 | 0.30 | 0.13 | 0.92 |
| | 3.05 | 8.51 ± 0.08 | 48.45 ± 0.11 | 6.45 ± 0.03 | 0.30 ± 0.01 | 0.27 ± 0.03 | 44.21 | 0.32 | 0.13 | 0.91 |
| | 3.20 | 8.49 ± 0.14 | 48.53 ± 0.13 | 6.43 ± 0.10 | 0.33 ± 0.01 | 0.22 ± 0.03 | 44.11 | 0.38 | 0.13 | 0.91 |
| | 3.31 | 8.41 ± 0.20 | 48.61 ± 0.08 | 6.39 ± 0.06 | 0.32 ± 0.01 | 0.20 ± 0.01 | 44.06 | 0.42 | 0.13 | 0.90 |
| | 3.64 | 8.33 ± 0.26 | 49.54 ± 0.15 | 6.40 ± 0.12 | 0.35 ± 0.03 | 0.23 ± 0.02 | 42.94 | 0.54 | 0.12 | 0.87 |
| | 3.79 | 8.27 ± 0.17 | 49.67 ± 0.10 | 6.38 ± 0.05 | 0.38 ± 0.01 | 0.22 ± 0.01 | 42.77 | 0.58 | 0.12 | 0.86 |
| | 3.90 | 8.24 ± 0.22 | 50.10 ± 0.09 | 6.35 ± 0.08 | 0.42 ± 0.04 | 0.24 ± 0.04 | 42.28 | 0.61 | 0.12 | 0.84 |
| Oat straw | NT | 8.41 ± 0.18 | 44.24 ± 0.22 | 6.05 ± 0.11 | 0.41 ± 0.02 | 0.11 ± 0.02 | 43.87 | 5.32 | 0.14 | 0.99 |
| | 2.47 | 8.39 ± 0.20 | 44.36 ± 0.26 | 6.04 ± 0.08 | 0.39 ± 0.01 | 0.26 ± 0.04 | 43.59 | 5.36 | 0.13 | 0.98 |
| | 2.61 | 8.20 ± 0.31 | 44.39 ± 0.09 | 6.01 ± 0.03 | 0.38 ± 0.03 | 0.18 ± 0.02 | 43.37 | 5.67 | 0.13 | 0.97 |
| | 2.72 | 8.19 ± 0.22 | 44.48 ± 0.17 | 5.99 ± 0.01 | 0.37 ± 0.02 | 0.14 ± 0.01 | 43.16 | 5.86 | 0.13 | 0.97 |
| | 3.05 | 8.14 ± 0.23 | 44.87 ± 0.28 | 5.98 ± 0.12 | 0.39 ± 0.02 | 0.15 ± 0.02 | 42.64 | 5.97 | 0.13 | 0.95 |
| | 3.20 | 8.12 ± 0.05 | 44.89 ± 0.12 | 5.92 ± 0.09 | 0.35 ± 0.05 | 0.11 ± 0.05 | 42.65 | 6.08 | 0.13 | 0.95 |
| | 3.31 | 8.01 ± 0.06 | 45.05 ± 0.03 | 5.89 ± 0.02 | 0.33 ± 0.01 | 0.10 ± 0.02 | 42.41 | 6.22 | 0.13 | 0.94 |
| | 3.64 | 7.99 ± 0.17 | 45.57 ± 0.11 | 5.87 ± 0.10 | 0.36 ± 0.02 | 0.11 ± 0.01 | 41.80 | 6.29 | 0.12 | 0.92 |
| | 3.79 | 7.98 ± 0.32 | 45.66 ± 0.07 | 5.88 ± 0.04 | 0.37 ± 0.01 | 0.11 ± 0.01 | 41.61 | 6.37 | 0.12 | 0.91 |
| | 3.90 | 7.98 ± 0.06 | 45.65 ± 0.08 | 5.84 ± 0.01 | 0.41 ± 0.01 | 0.11 ± 0.01 | 41.50 | 6.49 | 0.12 | 0.90 |

R = severity factor; M.C. = moisture content (d.b.); C = carbon; H = hydrogen; N = nitrogen; S = sulfur; O = oxygen; H/C = hydrogen to carbon ratio; O/C = oxygen to carbon ratio; and NT = non-treated.; n=3.

3.6.5 Higher heating values (HHV) of pretreated and untreated biomass

The determination of HHV was essential due to the expected use of biomass treated by steam explosion for home heating or to produce electricity. The HHV of the untreated sawdust and oat straw was 18.02 MJ/kg and 16.84 MJ/kg, respectively. Severity of the treatment condition influences the HHV, as shown in Figure 3.3. The HHV of treated biomass increased up to a maximum of about 9.5% and 7% compared to the untreated sawdust and oat straw, respectively. Higher moisture content negatively affected the HHV of the solid pellet, while increased

temperature positively affected the HHV. Moisture presence in fuel caused smoke and promoted partial combustion. Bhattacharya et al. (2002) studied the effect of moisture content on efficiencies of biomass-fired cookstoves and reported a reduction in efficiency in all stoves with an increase in fuel moisture content from 10% to 25%. In a similar manner, Yibo et al. (2014) reported that the burning rate of biomass in a semi-gasified cookstove decreased by 33.3% from 30.0 g/min at 5.9% moisture content to 20.0 g/min at 22.1% moisture content, and the cooking power was reduced by 36.1% from 1910 W at 5.9% moisture content to 1220 W at 22.1% moisture content. Lam et al. (2012) and Kumar et al. (2010) observed an increase in HHV, ash, and lignin content when whitewood biomass was steam pretreated. Adapa (2011) reported that the HHV of oat straw increased from 16.40 MJ/kg to 17.80 MJ/kg after steam explosion pretreatment at 180 °C, using a steam pressure of 900 kPa for 4 min. Any chemical, structural, or physical changes in the composition of lignocellulosic biomass (cellulose, hemicellulose, and lignin) could potentially lead to changes in the HHV of the biomass. As stated earlier, during steam explosion, small hydrocarbon molecules with low energy density volatilize, thereby leaving behind the remaining components that increase energy density. In addition, carbonization of the biomass sample during steam explosion pretreatment resulted in higher energy content. Steam explosion affects the HHV of the formed pellets by affecting the raw biomass structural compositions, and this is achieved by autohydrolysis, which removes the extractives with low volatility resulting in increased HHV (Lam et al. 2015; Tang et al. 2018).

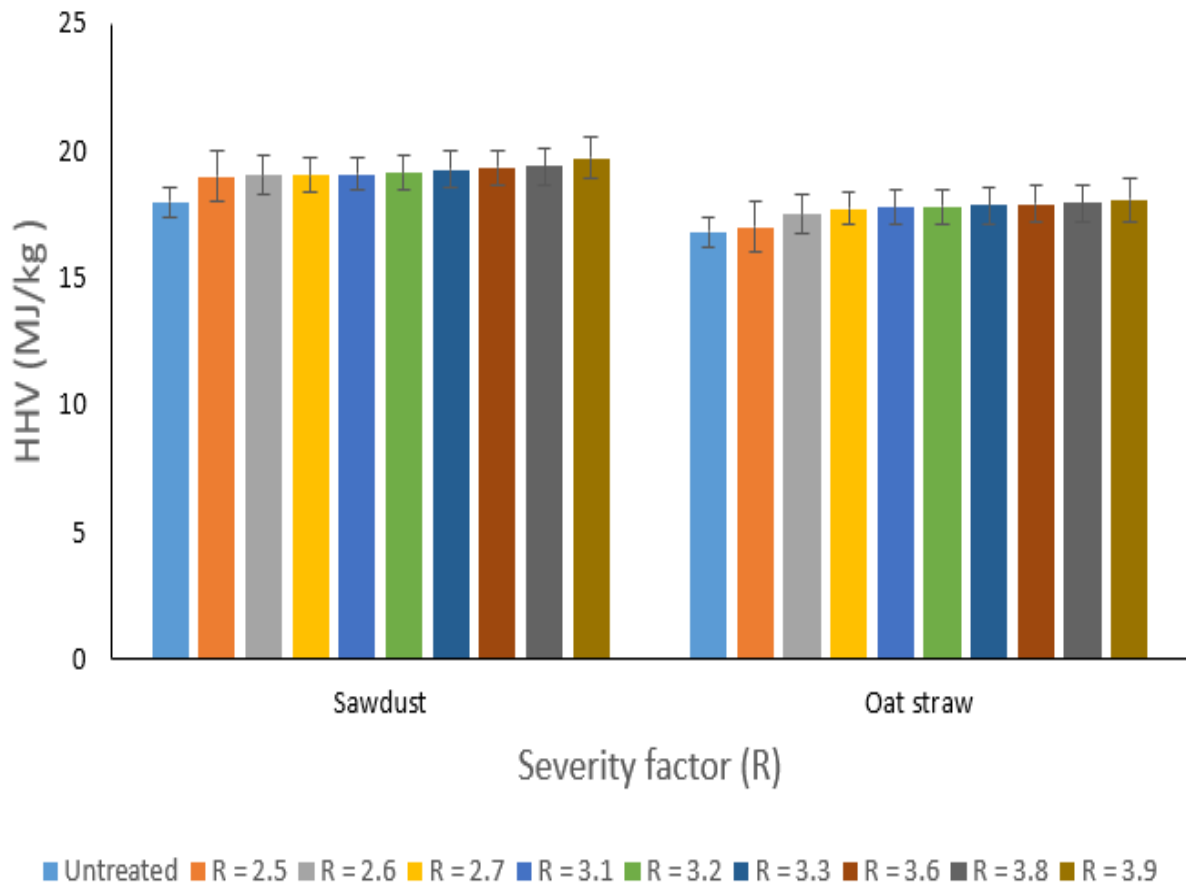


Figure 3.3. Higher heating value of steam-exploded and untreated biomass under different severities.

3.6.6 Moisture absorption

Generally, the absorption of moisture by raw biomass samples occurs because of the presence of the OH group. During the steam explosion pretreatment, the lignocellulosic structure of the straw and sawdust are disintegrated by the sudden pressure change (from a very high value to 0 kPa); the lignin and hemicellulose content are hydrolyzed by this process, a portion of which are washed and drained with wastewater. This process tends to destroy the OH group, thereby causing the biomass sample to lose the ability to form hydrogen bonds (Jin and Chen 2006; Adapa et al. 2010). Several research groups optimized the steam explosion condition and

concluded that less severe conditions resulted in the recovery of hemicelluloses. Wu et al. (1999) used optimization of steam explosion to enhance hemicellulose recovery and enzymatic hydrolysis of cellulose in softwoods. In addition, Qing et al. (2010) stated that mild conditions of steam explosion caused less sugar degradation when corn stover was pretreated. Figure 3.4 represents the graph of moisture content against time when the pellets were kept in the humidifier. After 1 h of subjecting the pellets to the humidifier condition, the untreated samples exhibited the least hydrophobic behavior, with a moisture uptake of 6.24% and 5.60% for oat straw and sawdust, respectively. The highest hydrophobic characteristic was observed in sawdust pellet samples steam exploded at 200 °C with moisture uptake of 2.89%. This could explain the fact that hemicelluloses were responsible for moisture absorption and biological degradation, therefore a decreased hemicellulose content reduced the total moisture absorption of the biomass. Lam (2011) found similar results when Douglas fir (*Pseudotsuga taxifolia*) wood chips were steam exploded. Hence, steam-exploded pellets are less vulnerable to biological deterioration and distortion after moisture absorption. This study shows that steam-exploded pellet samples became more hydrophobic as the treatment severity condition increased.

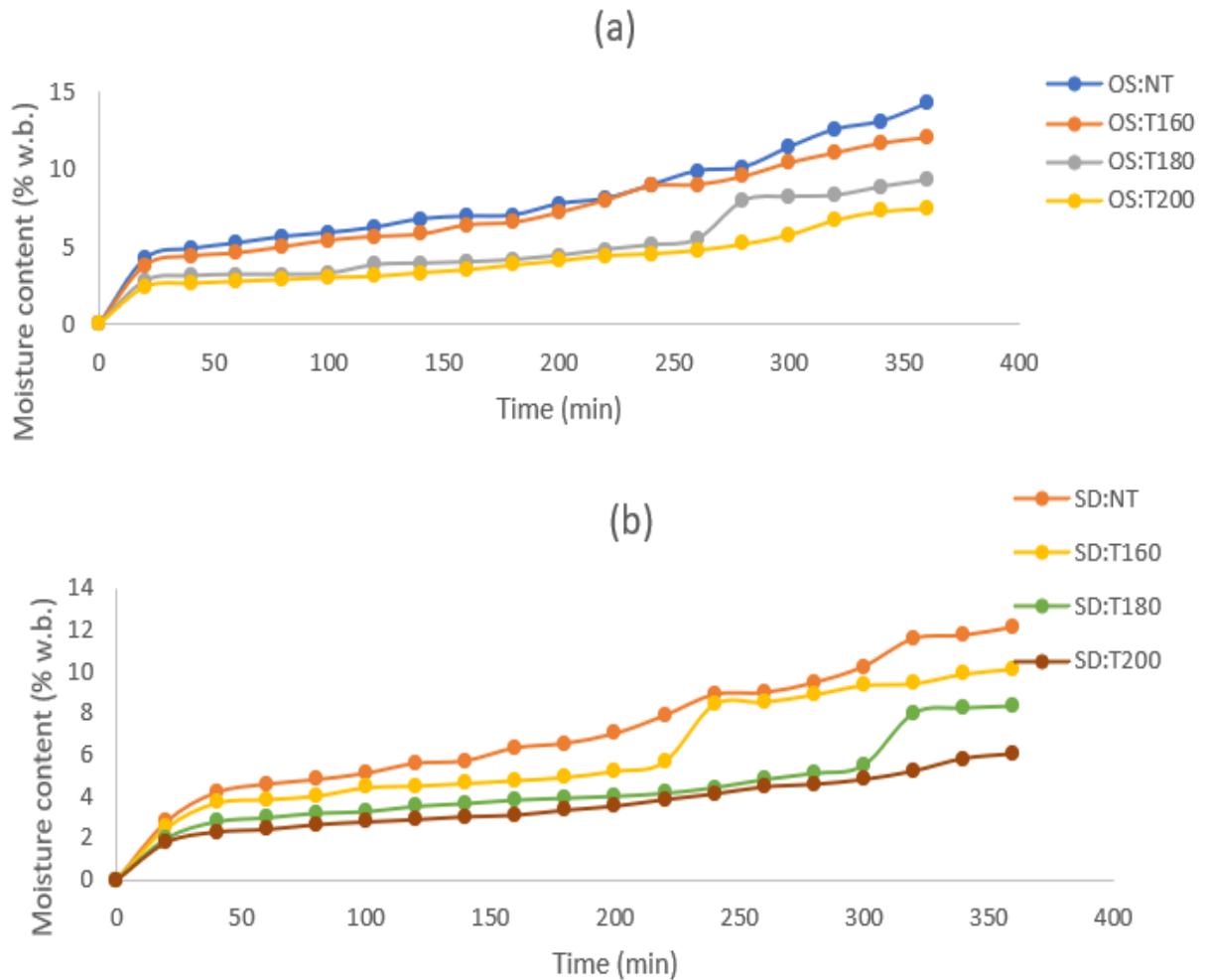


Figure 3.4. Moisture absorption of steam-treated and non-treated oat straw (a) and sawdust (b) pellets. OS: NT (oat straw: untreated); OS: T160 (oat straw: steam exploded at 160 °C); OS: T180 (oat straw: steam exploded at 180 °C); OS: T200 (oat straw: steam exploded at 200 °C); SD: NT (sawdust: untreated); SD: T160 (sawdust: steam exploded at 160 °C); SD: T180 (sawdust: steam exploded at 180 °C); SD: T200 (sawdust: steam exploded at 200 °C).

3.6.7 Microstructural analysis

The microscale impact of the steam explosion pretreatment of sawdust and oat straw grinds can be explained further by microstructural analysis, giving insight on the morphology of the pellet samples. Figure 3.5a–h shows the SEM micrographs of the longitudinal cross-section of the untreated and steam-exploded biomass pellets. Figure 3.5a, e shows the pellet sample from the

untreated oat straw and sawdust, respectively, with pore spaces, and the rough surface with more bonded particles that are loosely compacted are evident. The SEM images of the pellets from steam-exploded oat straw and sawdust (Figure 3.5b–d, f–h) reveal more tightly bonded particles and a cemented surface with fewer pores when compared with the untreated oat straw and sawdust. A lengthy defibrillation of fibers was noticed after the steam explosion pretreatment, mainly because of the mechanical effect from adiabatic expansion of absorbed water during the process. This shows that the biodeterioration of the lignocellulosic components of the biomass during steam explosion liberated more of the essential binders in the sample, resulting in high pellet tensile strength observed in the steam-exploded oat straw and sawdust pellets relative to the untreated oat straw and sawdust pellets. Similar observations were reported by some researchers (Gastaldi et al. 1998; Kristensen et al. 2008).

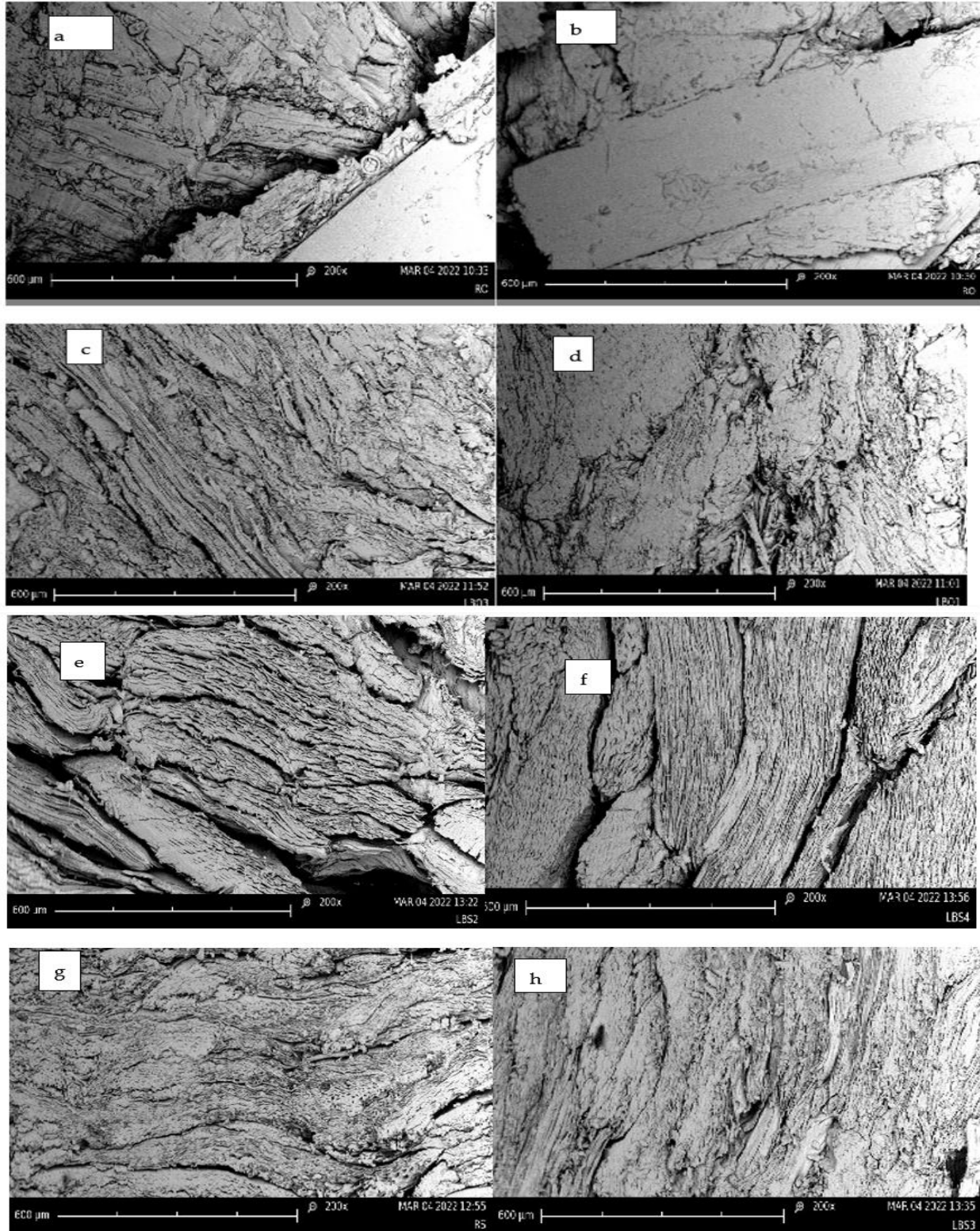


Figure 3.5. Scanning electron microscope images of the longitudinal cross-section of the untreated and steam-treated oat straw and sawdust pellets: (a) OS: NT (oat straw: untreated); (b) OS: T160 (oat straw: steam exploded at 160 °C); (c) OS: T180 (oat straw: steam exploded at 180 °C); (d) OS: T200 (oat straw: steam exploded at 200 °C); (e) SD: NT (sawdust: untreated); (f) SD: T160 (sawdust: steam exploded at 160 °C); (g) SD: T180 (sawdust: steam exploded at 180 °C); (h) SD: T200 (sawdust: steam exploded at 200 °C).

3.6.8 Effects of independent variables and their interactions on pellet quality

The effect of the steam explosion temperature, time, and moisture content after factorial regression are presented in Table 3.3. The analysis of variance (ANOVA) results showed that the p -values for all the response variables (dimensional stability, pellet tensile strength, and pellet density) studied showed a significant regression value ($p < 0.05$), excluding the pellet density of oat straw. The relationship among the three factors temperature, time, and moisture content of sample (T, t, MC) did not show significant effect on the response variables of oat straw pellets. Sample moisture content also affected the pellet density sawdust significantly. Generally, as sample moisture content increased, pellet density decreased. The compaction of hay across a wide range of moisture contents (28–44% w.b.) was studied by Gustafson and Kjelgaard (1963), where they reported a decrease in density of the pellets as sample moisture contents increased. Furthermore, Rehkugler and Buchele (1969) noticed that the relaxed density of pellets decreased with respect to sample moisture content ranging between 6% and 25% (w.b.). For both biomass samples, steam explosion temperature and retention time had a significant effect ($p < 0.05$) on all the response variables investigated. Among the independent variables, the sample moisture content indicated no significant effect on the response variables of the oat straw pellets. This may be because of the fibrous nature of oat straw.

Table 3.3. Analysis of variance (ANOVA) table for factors affecting dimensional stability, pellet density, and tensile strength of biomass pellets.

| Source | DF | Sawdust | | | | Oat Straw | | | | |
|--------------------------------|----|----------|----------|---------|---------|-----------|----------|----------|---------|---------|
| | | SS | MS | F-Value | p-Value | DF | SS | MS | F-Value | p-Value |
| Dimensional stability | | | | | | | | | | |
| Temperature(<i>T</i>) | 1 | 98.80 | 98.80 | 708.55 | 0.00 | 1 | 132.74 | 132.74 | 584.37 | 0.00 |
| Time (t) | 1 | 7.46 | 7.46 | 53.52 | 0.00 | 1 | 14.15 | 14.15 | 62.30 | 0.00 |
| Moisture Content (<i>MC</i>) | 1 | 0.55 | 0.55 | 3.93 | 0.06 | 1 | 1.81 | 1.81 | 7.95 | 0.01 |
| <i>Txt</i> | 1 | 0.87 | 0.87 | 6.24 | 0.02 | 1 | 0.61 | 0.61 | 2.67 | 0.12 |
| <i>TxMC</i> | 1 | 0.05 | 0.05 | 0.37 | 0.55 | 1 | 0.03 | 0.03 | 0.14 | 0.72 |
| <i>TxMC</i> | 1 | 0.03 | 0.03 | 0.20 | 0.66 | 1 | 0.02 | 0.02 | 0.09 | 0.77 |
| <i>TxtxMC</i> | 1 | 0.07 | 0.07 | 0.49 | 0.49 | 1 | 0.06 | 0.06 | 0.26 | 0.62 |
| Error | 19 | 2.65 | 0.14 | | | 19 | 4.32 | 0.23 | | |
| Total | 26 | 110.47 | | | | 26 | 153.73 | | | |
| Pellet Density | | | | | | | | | | |
| Temperature(<i>T</i>) | 1 | 16138.90 | 16138.90 | 25.24 | 0.00 | 1 | 28251.40 | 28251.40 | 36.51 | 0.00 |
| Time (t) | 1 | 3943.30 | 3943.30 | 6.17 | 0.02 | 1 | 2969.10 | 2969.10 | 3.84 | 0.07 |
| Moisture Content (<i>MC</i>) | 1 | 250.00 | 250.00 | 0.39 | 0.44 | 1 | 189.90 | 189.90 | 0.25 | 0.63 |
| <i>Txt</i> | 1 | 6.60 | 6.60 | 0.01 | 0.92 | 1 | 141.00 | 141.00 | 0.18 | 0.67 |
| <i>TxMC</i> | 1 | 65.20 | 65.20 | 0.1 | 0.75 | 1 | 39.90 | 39.90 | 0.05 | 0.82 |
| <i>TxMC</i> | 1 | 543.20 | 543.20 | 0.85 | 0.37 | 1 | 2.60 | 2.60 | 0.00 | 0.95 |
| <i>TxtxMC</i> | 1 | 482.50 | 482.50 | 0.75 | 0.40 | 1 | 4.10 | 4.10 | 0.01 | 0.94 |
| Error | 19 | 12150.60 | 639.50 | | | 19 | 14701.40 | 773.80 | | |
| Total | 26 | 33580.30 | | | | 26 | 46299.40 | | | |
| Tensile strength | | | | | | | | | | |
| Temperature(<i>T</i>) | 1 | 18.34 | 18.34 | 228.95 | 0.00 | 1 | 9.25 | 9.25 | 396.47 | 0.00 |
| Time (t) | 1 | 2.58 | 2.58 | 32.16 | 0.00 | 1 | 1.84 | 1.84 | 78.77 | 0.00 |
| Moisture Content (<i>MC</i>) | 1 | 0.03 | 0.03 | 0.39 | 0.44 | 1 | 0.03 | 0.03 | 1.24 | 0.28 |
| <i>Txt</i> | 1 | 0.14 | 0.14 | 1.76 | 0.20 | 1 | 0.15 | 0.15 | 6.61 | 0.02 |
| <i>TxMC</i> | 1 | 0.07 | 0.07 | 0.86 | 0.37 | 1 | 0.01 | 0.01 | 0.26 | 0.62 |
| <i>TxMC</i> | 1 | 0.13 | 0.13 | 1.60 | 0.22 | 1 | 0.00 | 0.00 | 0.02 | 0.90 |
| <i>TxtxMC</i> | 1 | 0.01 | 0.01 | 0.14 | 0.71 | 1 | 0.00 | 0.00 | 0.02 | 0.89 |
| Error | 19 | 1.52 | 0.08 | | | 19 | 0.44 | 0.02 | | |
| Total | 26 | 22.82 | | | | 26 | 11.71 | | | |

3.7 Conclusions

This work investigated the effects of temperature, time, and sampled moisture content on the mechanical properties of biomass (oat straw and spruce sawdust) pellets. These variables showed a significant effect on all the response variables studied for sawdust but not for oat straw. This study indicated that the most significant ($p < 0.05$) factors in the steam explosion of the biomass

for pellet production were temperature and time. The p -values associated with the regression models for the dimensional stability, pellet unit density, and tensile strength of the steam exploded pellets were significant ($p < 0.05$). Investigation of the steam explosion technology shows that some factors influence the productivity of the process, including retention time, initial sample moisture content, and pressure. Steam explosion breaks up the lignocellulosic structure of biomass, leading to lower bulk densities of pellets. After steam explosion pretreatment the carbon and the nitrogen content of the steam-exploded sample increased significantly ($p < 0.01$), concomitant with temperature and retention time, while biomass hydrogen and oxygen content decreased. Hydroxyl groups are linked to biomass structural components, and their volatilization during steam explosion leads to an increase in the energy density of the residual carbon-rich solids. In addition, the carbonization and autohydrolysis process in the steam explosion removes the extractives with low volatility, resulting in increased HHV. Higher moisture content negatively affects the HHV of the solid pellet, while increased steam explosion temperature positively affects the HHV. During the steam explosion pretreatment, the lignocellulosic structure of the straw and sawdust are disintegrated by the sudden pressure change and the lignin and hemicellulose are hydrolyzed by this process, a portion of which are washed and drained with wastewater. This process tends to destroy the OH group, thereby causing the biomass sample to lose the ability to form a hydrogen bond and increasing the hydrophobic characteristic of the sample. This study showed that steam-exploded pellet samples became more hydrophobic as the treatment severity condition increased. Extensive defibrillation of fibers was observed after the steam explosion pretreatment, mainly because of the mechanical effect from the adiabatic expansion of absorbed water during the process. SEM images of the pellets from steam-exploded pellets reveal more tightly bonded particles and cemented surfaces with fewer

pores when compared with the untreated pellets. In this study, steam explosion pretreatment resulted in positive effects on the quality of pellets.

Chapter 4

Effect of Torrefaction on the Physiochemical Properties of White Spruce Sawdust for Biofuel Production

- A similar version of this chapter has been published in the journal *Fuels*: Onyenwoke, C., L.G. Tabil, E. Mupondwa, D. Cree, and P. Adapa. 2023. Effect of torrefaction on the physiochemical properties of white spruce sawdust for biofuel production. *Fuels* 4: 111–131. <https://doi.org/10.3390/fuels4010008>.

Contribution of the Ph.D. Candidate

The experimental work, data collection and analysis, original drafting, review and editing of the manuscript was done by the Ph.D. candidate, while Drs. L.G. Tabil, E. Mupondwa, D. Cree, and P. Adapa, provided resources and editorial input. Dr. L.G. Tabil performed the supervisory role, while Drs. L.G. Tabil, E. Mupondwa and D. Cree also carried out the project administrative roles.

Contribution of the Paper to the Overall Study

This paper addressed the second and part of the third objectives of this study: to investigate torrefaction pretreatment of sawdust and oat straw to improve their quality as biofuel pellets and to investigate the combined effect of pretreatment and binders on the quality of pellet formed. This study evaluates the physiochemical characteristics of the pellets formed from torrefied sawdust such as measuring tensile strength, moisture uptake rate, bulk density, pellet density and scanning electron microscopy (SEM) analysis. Also, the chemical components and thermochemical analysis of the pellets were measured. These include ultimate analysis,

proximate analysis, and determination of high heating value. Furthermore, the results of this study demonstrated that torrefaction pretreatment of sawdust led to an improvement in the higher heating value (HHV), hydrophobicity, bulk, particle density, pellet dimensional stability, and pellet density. However, the pellet tensile strength decreased as the torrefaction severity increased. Pellet tensile strength is relevant during transportation and storage. Therefore, to enhance the tensile strength of the pellets, the introduction of a binder was necessary. Torrefaction liquid and sawdust were used as additives at different proportions during pelletization. The results of this research can be useful to bioenergy plants.

4.1 Abstract

Torrefaction pretreatment is a mild form of pyrolysis that has the potential to produce a high-quality raw material for making biofuel that serves as a replacement for coal in the bioenergy industry. Microwave-assisted torrefaction was conducted on white spruce sawdust (WSS) at temperatures of 200 °C, 250 °C, and 300 °C and retention times of 5 min, 7 min, and 9 min in an inert environment. The torrefaction process produces a solid carbon, commonly known as biochar, and condensable (torrefaction liquid (TL)) and non-condensable gases. In this study, torrefaction characteristics were investigated to observe its effects on the thermal and physiochemical properties of the pellets produced. During the torrefaction process, a significant mass loss associated with the decomposition of hemicellulose was observed. The hemicellulose content drastically reduced to approximately 1.8% and the cellulose content was reduced by approximately 10%, while the apparent lignin content was approximately 35% as the severity increased. This led to an improvement in the higher heating value (HHV), hydrophobicity, bulk, particle density, pellet dimensional stability, and pellet density. However, the pellet tensile

strength decreased as the torrefaction severity increased. Pellet tensile strength is a critical indicator of biomass pellets that expresses the force required to crush or damage a pellet. Therefore, to enhance the tensile strength of the pellets, the introduction of a binder was necessary. Torrefaction liquid and sawdust were used as additives at different proportions during pelletization. The addition of binders (torrefaction liquid and sawdust) to the pellet formulation increased the tensile strength of the torrefied WSS by approximately 50%. The OH groups in the biomass break down to a limited degree due to dehydration. This hinders the formation of H bonds, thereby increasing the chances that the pretreated biomass will become hydrophobic. The SEM graphs showed that the torrefied WSS pellets demonstrated more firmly glued surfaces with fewer pores spaces when set side by side with the raw pellets. The thermogravimetric analysis conducted showed that the torrefaction of WSS slightly reduced its thermal stability.

4.2 Introduction

A genuine solicitude is drawn regarding the increased risk of environmental threats linked with fossil fuel utilization (Dai et al. 2020; Lehmann 2007). With the constant unsteadiness of energy prices and the rapid depletion of conventional energy resources, it is urgent to source sustainable, renewable, and eco-friendly forms of energy (Czernik and Bridgwater 2004). Biomass stands out in the substitute energy competition because it is a carbon-neutral bioenergy source, has a low cost, and is abundantly available (Zhang et al. 2021; Cai et al. 2017). However, some difficulties linked with the utilization of biomass in its unprocessed state (such as a low energy density, low bulk density, a heterogeneous nature, low efficiency, ash accumulation, and a high moisture content) compromise its potential development in the bioenergy industry (Arias et al. 2008; Strandberg et al. 2015). In recent years, the pretreatment of biomass has demonstrated some

improvement in the characteristics of the treated biomass. Torrefaction is a biomass heat operation associated with roasting a biomass sample in a controlled environment at torrefaction temperatures, which range from 200 to 300 °C. Torrefaction is commonly known as mild pyrolysis, and it has been investigated for the pretreatment of biomass to make the biomass more hydrophobic, reduce its moisture content, and increase its calorific value, resulting in an increase in the volumetric energy density of the biomass. In addition, torrefaction with pelletization can reduce the transportation costs of biomass and intensify its heating value and hydrophobicity (Li et al. 2012a; Dudyński et al. 2015). These changes make biomass more combustible, storable, and attractive (Cai et al. 2017; Bach et al. 2017).

Approximately 350 million hectares of land in Canada are forest, making Canada the third in the world with respect to forest area (Statista Research Department 2022). The forestry industry contributes majorly to the country's economy. In 2020, the wood-product-manufacturing sector had a nominal GDP of approximately CAD \$12.6 billion, making it the most lucrative of the forestry industries (Statista Research Department 2022). Canada's main trading partner in forest resources is the United States, with Canadian exports valued at approximately CAD \$25 billion in the year 2020. The most-sold Canadian forest products are millwork, sawmill, and lumber, with export rates valued slightly above CAD \$22 billion worldwide (Statista Research Department 2022). The utilization of forestry residues to generate energy has traditionally existed in Canada, especially in the forestry industry (Onyenwoke et al. 2022). The feedstock used by these industries for generating electricity and other forms of energy are forest residues such as sawdust, bark, and branches. Some varieties of trees found in Saskatchewan (SK), Canada, include trembling aspen, white spruce, jack pine, and black spruce. Saskatchewan has four large sawmills and several smaller sawmills; approximately 1.3 million m³ (545 million

board feet) of spruce-pine-fir (SPF) lumber is produced annually from these mills (Saskatchewan Forest Products Buyers' Guide 2019). In Canada, white spruce (*Picea glauca*) is found mostly in the boreal forest (northern coniferous forest). In Saskatchewan, the total annual allowable harvest is 28% of the coniferous forest and 16% of the total forest. Consequently, white spruce accounts for a substantial part of these harvests (Saskatchewan Forest Products Buyers' Guide 2019). Lignocellulosic biomass is readily available and can settle the issues arising from the use of biomass between food and energy (Rabemanolontsoa and Saka 2016). The utilization of forest residues, especially sawdust, for biofuel production decreases the dependency on food-resource biomass while also adding value to forest residues (Adapa et al. 2011).

Densification using binders such as starch, lignin, sawdust, and moisture has been studied by different researchers. Table 4.1 summarizes the effect of different binders on the pelletization of biomass as it has been reported by other researchers. Frequently used binders include starch and lignin; however, they are expensive and have competing uses as food ingredients (Cai et al. 2017). While it intends to enhance the thermal, mechanical, and physiochemical properties of torrefied sawdust pellets, this research also explores the utilization of cheap and non-food-based binders. In this study, white spruce sawdust and torrefaction liquid (TL) were introduced as binders during the pelletization process after the microwave-assisted torrefaction of WSS. The torrefaction gases (a condensable liquid) contain a large amount of energy. Therefore, introducing a closed recycling system has an economically viable potential for the torrefaction system. It is significant that this energy is utilized productively in the system (torrefaction and densification) to approach and settle the process stability, system reliability, and safety problems that currently avert commercial-scale torrefaction. This study also emphasizes the consequences

of torrefaction and binders on the thermal, mechanical, and physiochemical characteristics of pellets formed from torrefied WSS.

Table 4.1. Effect of binders on the pelletization of biomass, as determined by other researchers.

| Biomass | Binder | Key Findings | References |
|--------------------------------|---|---|----------------------|
| Sawdust (Norway spruce) | Four Starch grades (wheat, oxidized corn, potato, and oxidized potato). | <ul style="list-style-type: none"> . The durability of the pellets formed increased and did not decrease after 7 months of storage; . The energy consumption during the pelletization process decreased; . Oxidized corn additive was reported to have the best result in both cases mentioned above: it reduced the energy consumption during pelletization by 14%. . Proline and lignin produced high-quality pellets when set side by side with other additives; . Increasing the quantity of proline and | Ståhl et al. 2012 |
| Spruce sawdust | Lignin, lignosulfonate, proline, corn starch, and torrefied oat hull. | <ul style="list-style-type: none"> reducing the quantity of lignin in the pellet formulation enhanced the density and mechanical strength of the pellets; . The addition of torrefied oat hull to the pellet formulation improved the heating value and the moisture resistance slightly but reduced the density and mechanical strength of the pellets. . The densification of the torrefied sample into strong pellets was difficult compared with the raw sample; | Abedi et al. 2018 |
| Softwood residues | Moisture | <ul style="list-style-type: none"> . The addition of moisture into the torrefied samples enhanced the compression process and produced better pellets. | Peng et al. 2013 |
| Pine sawdust | Sawdust | <ul style="list-style-type: none"> . Sawdust is a low-cost and effective binder when compared with lignin and starch. | Peng et al. 2015 |
| Wood sawdust | Steam-treated spruce sawdust | <ul style="list-style-type: none"> . The addition of the binder during the pelletization process increased the pellet tensile strength by 50%. | Alizadeh et al. 2022 |

4.3 Materials and Methods

The torrefaction and pelletization process for white spruce sawdust (WSS) is shown in Figure 4.1. These processes and their effects are exhaustively discussed in depth in the consequent sub-heading. The torrefaction pretreatment process of WSS saves cost and energy since the feedstock in this study requires little or no preprocessing due to its nature (a fine particle).

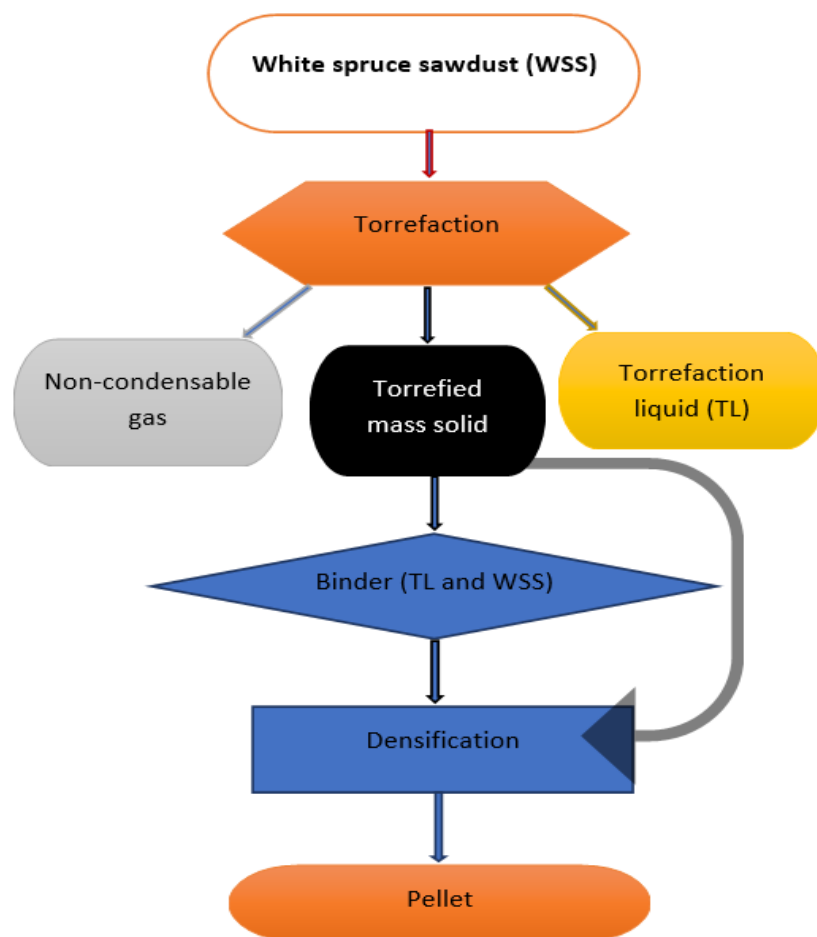


Figure 4.1. Torrefaction and pelletization process for white spruce sawdust.

4.3.1 Feedstock collection and preparation

White spruce sawdust (WSS) was received from the sawmill of NorSask Forest Products Inc. (Meadow Lake, SK, Canada) in the middle of 2020. The moisture content of the as-received sawdust was approximately 42% wet basis (w.b.). The samples were screened with a 0.8 mm sieve size to remove irregular particles. They were then preserved in a fastened plastic container in a conditioned space at a temperature of approximately 4 °C to be used for experimentation. The torrefaction liquid (TL) used as an additive during pelletization was a product of this study.

4.3.2 Torrefaction experiment

The torrefaction pretreatment of the WSS was carried out to improve its physicochemical characteristics. The setup of the lab-scale microwave oven and the pretreatment operation were similarly narrated by Valdez et al. (2021). Figure 4.2 shows a schematic diagram of the microwave torrefaction setup. Different temperatures of 200 °C (light torrefaction), 250 °C (moderate torrefaction), and 300 °C (severe torrefaction) and residence times of 5, 7, and 9 min were considered. Initially, a power level of 650 W was set before the microwave oven was turned on. The power level had to be controlled at different torrefaction residence times (decreased or increased) to attain a constant temperature (200, 250, or 300 °C) for the total residence time. A power level ranging from 400 to 750 W was recorded throughout the torrefaction process. The masses of torrefied solid (biochar) and liquid produced were recorded after each experiment, the experiments were performed three times, and the results were averaged. Equation (4.1) by Overend et al. (1987) was used to estimate the degree of torrefaction, which is a function of the residence time and temperature [22] that is also known as the severity factor.

$$SF = \log \int \exp \left[\frac{T - 100}{14.5} \right] dt \quad (4.1)$$

where, SF indicates the severity factor, T represents the torrefaction temperature ($^{\circ}\text{C}$), and t represents the residence time (min).

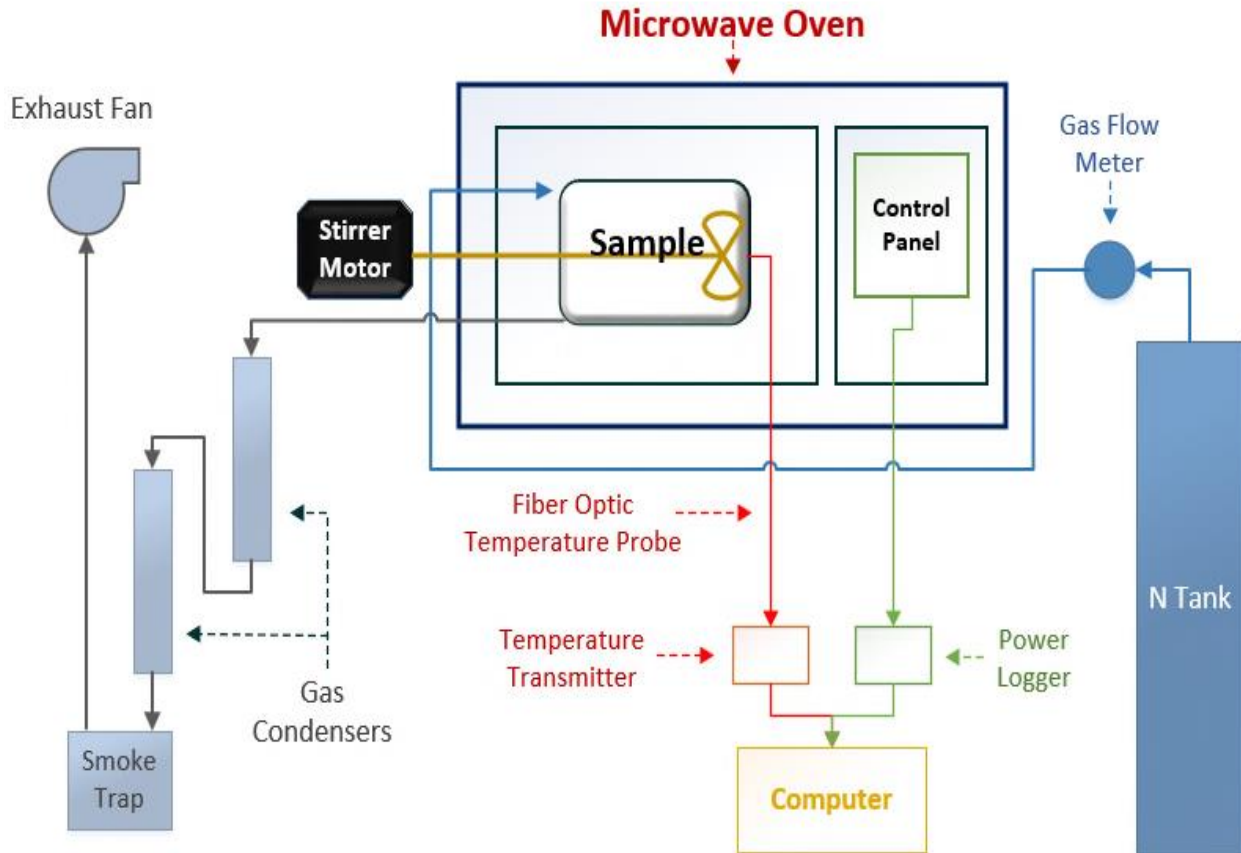


Figure 4.2. Schematic diagram of the microwave-assisted torrefaction pretreatment setup. Extracted from (Valdez et al. 2021).

4.3.3 Experimental design of torrefaction pretreatment

A factorial experimental design of two independent variables, the temperature of operation ($^{\circ}\text{C}$) and the residence time of the process (min) at three` levels for each factor (temperature at 200, 250, and 300 $^{\circ}\text{C}$ and retention time at 5, 7, or 9 min), were studied. The experiments were performed three times, and the results were averaged. An analysis of variance (ANOVA) was

carried out to evaluate the effect of the pretreatment operation on the response variables (dimensional stability, pellet tensile strength, and pellet density).

4.3.4 Densification

The torrefied WSS was pelletized to investigate the influence of residence time and torrefaction temperature on the pellets formed. The effect of binders on the pelletization was also studied using different formulations (torrefaction liquid (TL) and sawdust) and torrefied samples. Table 4.2 shows the different formulations that were pelletized to analyze the effect of the binders on the physiochemical characteristics of the respective pellets.

The pelletization of torrefied spruce sawdust samples and untreated spruce sawdust samples were executed using a similar setup and procedure as narrated by Onyenwoke et al. (2022).

Table 4.2. Formulations for pelletizing with the torrefaction liquid and sawdust.

| Composition | Torrefied WSS (%) | Torrefaction Liquid (%) | Sawdust (%) |
|--------------------|--------------------------|--------------------------------|--------------------|
| NB | 100 | - | - |
| TFO | 75 | 25 | - |
| TFS | 75 | - | 25 |
| TTOS | 33.3 | 33.3 | 33.3 |
| TWOS | 75 | 12.5 | 12.5 |

NB: Without binder; TFO: 75% torrefied WSS with 25% TL; TFS: 75% torrefied WSS with 25% sawdust; TTOS: 33.3% torrefied WSS with 33.3% sawdust and TL; TWOS: 75% torrefied WSS with 12.5% sawdust and TL each.

4.3.5 Characterization of torrefied white spruce sawdust

The characterization of the untreated and treated white spruce sawdust was carried out to estimate the influence of the residence time and torrefaction temperature on the WSS.

4.3.5.1 Moisture content and torrefaction yield

The amount of moisture present in the white spruce sawdust when it was collected was evaluated using ASAES358 (2012), while the quantity of moisture available after the torrefaction process was calculated by following the AACC Standard 44-15A (2005). The experiments were performed three times, and the results were averaged. Equation (4.2) was used to evaluate the mass yield of the pretreatment process. After each pretreatment, the biochar and the TL were weighed and kept in airtight Ziploc bags. The gas yield was determined by summing the solid yield and the TL yield and then subtracting them from the initial sample introduced.

$$M_Y = \frac{M_T}{M_I} \times 100 \quad (4.2)$$

where M_Y represents the percent mass yield of the torrefaction operation (%), M_T represents the mass of the torrefied sample (g), and M_I represents the mass of the untreated WSS sample (g). The quantity of weight loss and the amount of energy output were determined using Equations (4.3) and (4.4), respectively.

$$W_L = \frac{M_1 - M_2}{M_1} \times 100 \quad (4.3)$$

$$E_Y = \frac{H_2 M_2}{H_1 M_1} \times 100 \quad (4.4)$$

where W_L represents the weight loss; E_Y represents the energy yield; M_1 represents the mass of the untreated WSS; M_2 represents the mass of torrefied WSS; H_2 represents the higher heating value (HHV) of the WSS after torrefaction, and H_1 represents the HHV of the untreated WSS sample.

4.3.5.2 Particle size analysis

The ASABE (2006) procedure was used to examine the particle size of the WSS samples. Some researchers have reported a similar experiment in which the same setup and procedure were considered in evaluating the particle size of torrefied and untreated biomass (ASABE 2006; Iroba et al. 2017; Satpathy et al. 2013). Approximately 100 g of biochar and controlled samples were introduced into stacked sieves of various size openings, and the mechanical sieve shaker was left to run for approximately 10 min. The experiment was performed in three replicates and the results were averaged. Equations (4.5) and (4.6) were used to determine the geometric mean diameter (d_{gw}) and the standard deviation of the particle diameter (S_{gw}) of the WSS samples, respectively.

$$d_{gw} = \log^{-1} \left[\frac{\sum_{i=1}^n (W_i \log d_i)}{\sum_{i=1}^n W_i} \right] \quad (4.5)$$

$$S_{gw} = \frac{1}{2} d_{gw} \left[\log^{-1} S_{log} - (\log^{-1} S_{log})^{-1} \right] \quad (4.6)$$

where d_{gw} denotes the geometric mean diameter (mm); n denotes the number of sieves + 1 pan; W_i denotes the mass on the i th sieve (g); d_i represents the nominal sieve opening size of the i th sieve (mm); S_{log} represents the geometric standard deviation of log-normal distribution by mass in the common (base 10) logarithm.

4.3.5.3 Bulk and particle density

The bulk density of the unprocessed and torrefied WSS was determined due to its importance in logistics, storage, and densification. The setup and procedure for determining the bulk and particle density were similar to what was described by Onyenwoke et al. (2022). The bulk density of the WSS samples in kg/m^3 was estimated by the weight of the sample per its unit

volume. The experiment for bulk density was performed in three replicates and averaged, while particle density data were obtained in replicates of 10 and the results were averaged.

4.3.5.4 Elemental and proximate analysis

An Elementar analyzer (Vario EL III, Elementar Americas Inc., Ronkonkoma, NY, USA) was utilized to evaluate the amount of carbon (C), hydrogen (H), nitrogen (N), and sulfur (S) present in the raw sample and the biochar samples, while the quantity of oxygen available in the sample was estimated by difference. The setup of the equipment and procedures was similarly described by Onyenwoke et al. (2022).

The proximate analysis of the unprocessed WSS sample and biochar was evaluated by the ASTM standard procedures. The quantity of ash in the untreated WSS and the WSS biochar, moisture content (MC), and volatile matter (VM) were evaluated by the ASTM 3173-87 (2010), ASTM D3175-89 (1996), and ASTM 3174-04 (1997) standard procedures, respectively. Additionally, the amount of fixed carbon (FC) present in the samples was determined by differences (Bai et al. 2018), as provided in Equation (4.7).

Fixed Carbon

$$= 100 - (\text{ash content} + \text{moisture content} + \text{volatile mater}) \quad (4.7)$$

4.3.5.5 Higher heating value (HHV)

The HHV evaluation of both the biochar and the untreated WSS was carried out with the aid of a bomb calorimeter (6400 Automatic Isoperibol, Parr Instrument Company, Moline, IL, USA). The setup and the procedure for determining the HHV were similar to what was described by Onyenwoke et al. (2022). The test was executed three times and the results were averaged.

4.3.5.6 Chemical composition of torrefied WSS

The chemical content available of the torrefied WSS and the untreated WSS was estimated using the ANKOM methods for neutral detergent fiber (NDF), acid detergent fiber (ADF), and acid detergent lignin (ADL). ANKOM methods 5, 6, and 8 were used to determine the ADF, NDF, and ADL, respectively (ANKOM 2021a; ANKOM 2021b; ANKOM 2021c). A similar setup and procedure were used by Alizadeh et al. (2022) to determine the chemical composition of sawdust.

4.3.6 Pellet quality analysis

Analyzing the pellet quality was essential because of handling, transportation, and storage. It is also important in terms of designing a torrefaction system for a new pellet mill or scaling up an existing pellet mill.

4.3.6.1 Pellet unit density and dimensional stability

Pellet density and dimensional stability are two basic characteristics that enhance pellet quality. After the production of a pellet, its weight and volume were measured immediately, and it was kept in airtight Ziploc bags for further experiments. The pellet density is the ratio between the mass of the pellet and its volume. Approximately 10 pellet samples were used for this experiment. The pellets were carefully examined and stored for 14 d under 30–40% relative humidity and a temperature of 20–25 °C. The change in volume after 14 d (V_{14}) was then observed and recorded. The pellet dimensional stability is expressed as the ratio of the change in volume after storage for 14 d (V_{14}) and the initial volume of the pellet after production (V_0), which is evaluated using Equation (4.8).

$$\text{Dimensional stability (\%)} = \frac{V_{14} - V_0}{V_0} \times 100 \quad (4.8)$$

4.3.6.2 Pellet tensile strength

A diametral compression experiment was performed with the aid of the Instron tester to determine the tensile strength of the sawdust pellet. Kashaninejad and Tabil (2011) conducted the same test using a similar instrument to evaluate the pellet tensile strength. The experiment was replicated ten times and the results were averaged. Equation (4.9) was used to calculate the tensile strength of pellets.

$$\delta_x = \frac{2F}{\pi dl} \quad (4.9)$$

where δ_x represents the tensile strength in Pa; F is the applied load (N); d is the pellet diameter (m); l is the pellet sample thickness (m).

4.3.6.3 Moisture uptake test

A humidifier chamber (Espec SH-641 Benchtop chamber, ESPEC Corp., Osaka, Japan) operating under 90% humidity and a temperature of 25 °C was considered when carrying out the moisture uptake experiment in this study. This was necessary because pellets tend to absorb moisture and distort during transportation through the harsh weather conditions in Canada. The influence of the binders introduced during the pelletization of the WSS biochar samples was investigated through the moisture absorption experiment. The setup and the procedure for this experiment were similar to what was described by other researchers (Azargohar et al. 2019a; Lam et al. 2012; Li et al. 2011).

4.3.6.4 Microstructural examination

The surface and structural transformation of the biochar pellet were examined with the help of a scanning electron microscope (SEM) (Phenom-World, Eindhoven, the Netherlands) combined with a stereoscope. The biochar pellet samples were first prepared and coated with gold, following a procedure similar to what was described by Onyenwoke et al. (2022).

4.3.6.5. Thermogravimetric analysis (TGA)

The TGA of the untreated and torrefied WSS was determined with the aid of Pyris Diamond TG/DTA equipment (Perkin-Elmer, Waltham, MA, USA). The setup and procedure for determining the TGA were similar to what was described by Alizadeh et al. (2022).

4.4 Results and Discussion

4.4.1 Torrefaction severity factor

The severity factor (SF) represents both the residence time and the temperature (Overend et al. 1987). Table 4.3 represents the SF results acquired from this study: the temperature and time increased as the SF increased. The basic reaction that occurs during torrefaction pretreatment includes the depolymerization, carbonization, and devolatilization of hemicellulose, cellulose, and lignin (Tumuluru et al. 2011). Based on the torrefaction temperature, the traditional torrefaction pretreatment processes can be categorized into light (200–235 °C), mild (235–275 °C), and severe (275–300 °C) torrefaction (Valdez et al. 2021). Among the three main components of biomass, the most affected component is hemicellulose due to its thermal decomposition temperature, which ranges between 215 and 315 °C. Light torrefaction thermally degraded the hemicellulose in biomass to a certain extent, while lignin and cellulose were hardly

or slightly affected owing to their higher thermal decomposition temperatures (Rousset et al. 2011). During mild torrefaction, hemicellulose was significantly reduced, and the cellulose was slightly consumed. The severe torrefaction process significantly affected the cellulose and hemicellulose components of the biomass as well as the energy density, solid, and energy value.

Table 4.3. Severity factor of microwave-assisted torrefaction of white spruce sawdust.

| Temperature (°C) | Time (min) | Severity Factor |
|-------------------------|-------------------|------------------------|
| 200 | 5 | 3.64 |
| 200 | 7 | 3.79 |
| 200 | 9 | 3.91 |
| 250 | 5 | 5.11 |
| 250 | 7 | 5.26 |
| 250 | 9 | 5.37 |
| 300 | 5 | 6.59 |
| 300 | 7 | 6.73 |
| 300 | 9 | 6.84 |

4.4.2 Characterization of untreated and torrefied white spruce sawdust

The characterization of the untreated and torrefied WSS was carried out to investigate the effect of torrefaction on the physiochemical properties of the WSS. The results from the samples analysis are subsequently presented and discussed.

4.4.2.1 Moisture content and torrefaction yield

The amount of moisture present in the white spruce sawdust samples after the torrefaction pretreatment ranged between approximately 1% and 4%. The average moisture content of

torrefied WSS at 200 °C was significantly ($\alpha = 0.05$) higher when set side by side with the average moisture content of torrefied WSS at 250 °C and 300 °C. The average moisture content of WSS torrefied for 5 min was significantly ($\alpha = 0.05$) higher compared to the average moisture content of WSS torrefied for 7 min and 9 min. Normally, the quantity of moisture recorded for the raw biomass ranged from 10 to 50%. Torrefaction is a moisture-reduction process that reduces the amount of water present in the torrefied sample to approximately 1–3% on a weight basis (Tumuluru et al. 2011). The high presence of moisture in the raw biomass reduces the torrefaction efficiency and increases the cost of producing fuel (Evergreen Renewables 2022). The initial moisture content of the WSS sample used in this study was approximately 25%, which reduced drastically to approximately 1–3% after the severe torrefaction pretreatment. Cai et al. (2017) reported a moisture content as low as 0.07% after torrefaction under 280–300 °C. This could be due to the initial quantity of water present in the raw biomass sample and the severity of the torrefaction. Basically, as the SF increases, the moisture content reduces, thereby reducing the risk of biological deterioration and storage and transportation costs (Tumuluru et al. 2011).

Supplementary Material Figure S4.1 shows the product yields of torrefaction, which constitute a brown to solid-black mass known as biochar, condensable gas (torrefaction liquid), and non-condensable gas. The biochar yield decreased as the SF increased, but the liquid and gas yield increased with its increasing severity. When raw biomass is subjected to torrefaction, its dry matter reduces because of thermal breakdown. The biochar yield is a critical measure of how biomass resists thermal breakdown (Chen et al. 2015b). During the torrefaction operation, weight loss is a crucial factor for evaluating the process advancement. Figure 4.3 shows the weight loss and energy yield of the WSS at different SFs. As the severity increased, a higher weight loss was

observed. The most significant weight loss was noted at a severity factor of 6.84, having a weight loss of about 25%. A similar result was reported (Cai et al. 2017) when sawdust was torrefied at a temperature of 280–300 °C, which yielded a weight loss of 27.72%. However, the weight loss is attributed to the thermal breakdown of the three major contents: cellulose, hemicellulose, and lignin (Chew and Doshi 2011). The energy yield decreased as the SF intensified. This concurs with the result reported by Cai et al. (2017) in which 22.37% was the amount of energy lost as the torrefaction became severe. Although a mass loss was observed, the biomass characteristics improved (Chew and Doshi 2011; Yan et al. 2010; Van der Stelt et al. 2011; Li et al. 2012b).

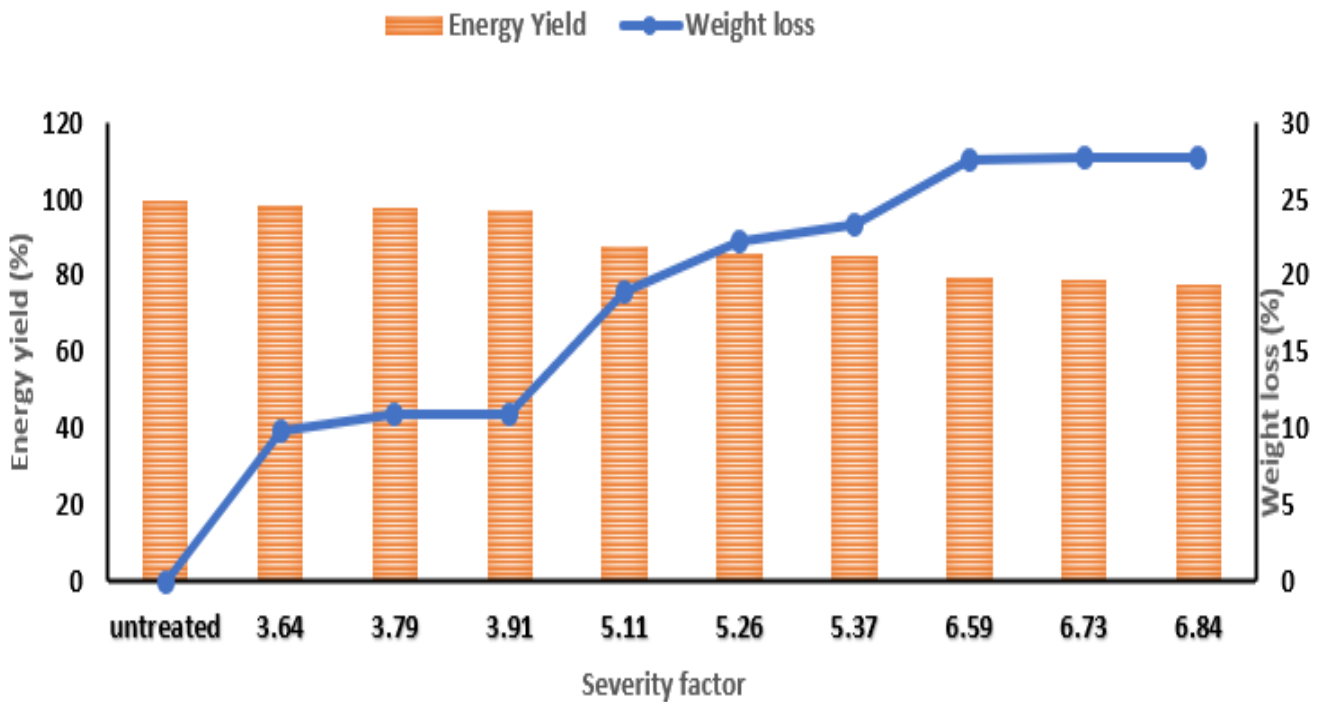


Figure 4.3. Weight loss and energy yield of WSS at different severity factors.

4.4.2.2 Particle size analysis

The size and shape of particles determine the available surface area needed for the particles to join firmly during pellet formation. This is a critical factor for not only the mechanical properties

of pellets (Rudolfsson et al. 2015; Serrano et al. 2011) but also the energy utilized during pelletization (Ghiasis 2020; Huang et al. 2017). Figure 4.4 represents the normal size distribution of torrefied WSS at different severities. A narrower particle size distribution was observed, making it suitable for densification (Mani et al. 2004). This is possible when tiny particles rearrange and fill the gaps between larger particles (Tabil 1996). The increasing severity condition resulted in an increase in the proportion of fine particles in the sample. A Shapiro–Wilk test ($p > 0.05$) demonstrated that the biochar particle sizes from the different severity conditions were approximately normally distributed. A similar trend was reported (Phanphanich and Mani 2011) when the same experiment was carried out on pine chips and logging residues. A geometric mean particle size of 0.36 mm was recorded for the untreated WSS. Table 4.4 shows the geometric mean diameter (d_{gw}) and geometric standard deviation (S_{gw}) of the untreated WSS and WSS biochar particles. The d_{gw} ranged from 0.32 mm to 0.35 mm, while the S_{gw} ranged from 0.14 mm to 0.22 mm. Chen et al. (2011) observed similar results using the torrefaction of bamboo, pine chips, and logging residues. Alizadeh et al. (2022) also reported a 0.32 mm to 0.38 mm range for the d_{gw} of torrefied sawdust and an S_{gw} range from 0.14 mm to 0.21 mm.

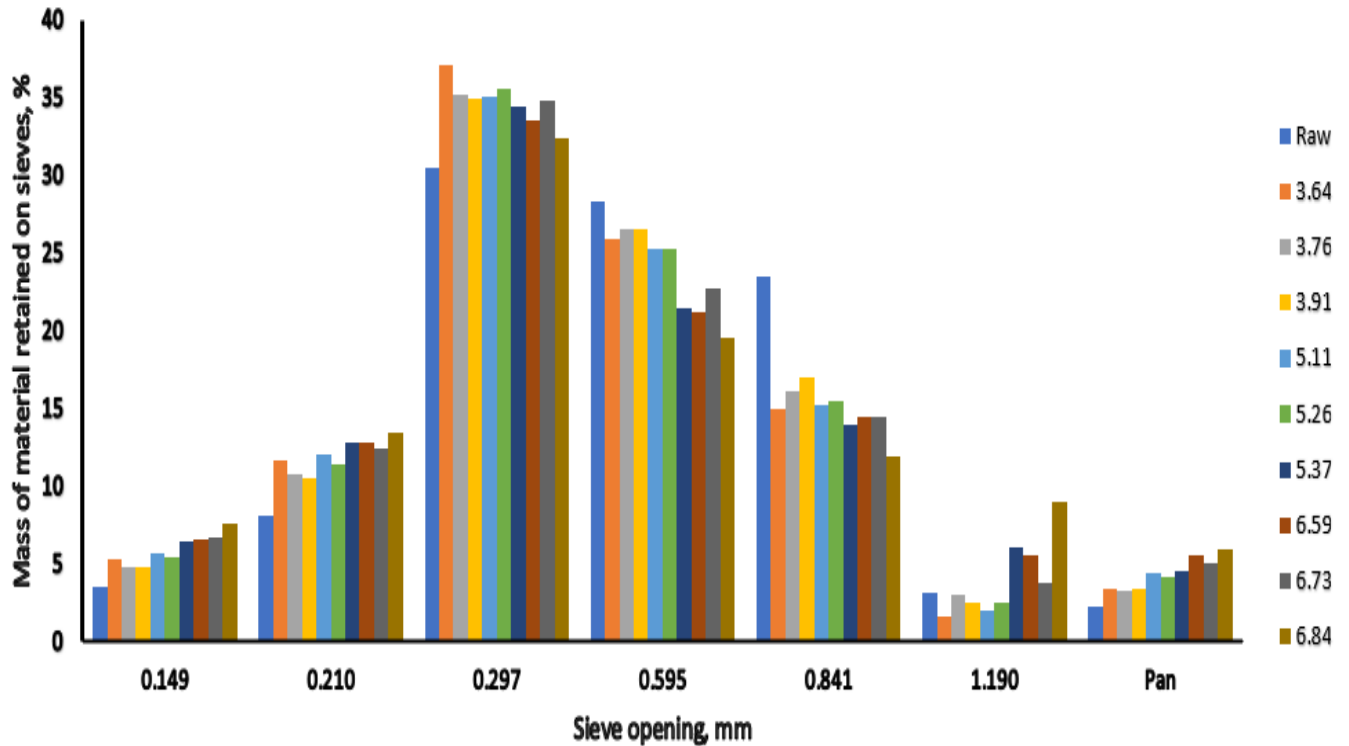


Figure 4.4. Particle size distribution of torrefied WSS at different severities.

4.4.2.3 Bulk and particle density

Table 4.4 depicts the bulk and particle density of the unprocessed WSS and torrefied WSS at different temperatures. The bulk density of the raw WSS was notably higher than the treated samples. This could be because the torrefaction pretreatment application tends to disintegrate the organized and compacted lignocellulosic structure (cellulose, hemicellulose, and lignin) of biomass, resulting in a lowered bulk density. This agrees with results reported by other researchers (Peng et al. 2014, Chen and Kuo 2011). The bulk density of the biochar WSS ranged from 110.24 kg/m³ to 167.24 kg/m³. The particle density of the biochar WSS was greater than the untreated samples. The higher particle density of the torrefied WSS could be because of the shrinkage of biomass samples that occurs during the torrefaction process (Mani et al. 2006a).

The particle densities of the treated WSS ranged from 1250.68 kg/m³ to 1454.84 kg/m³. Peng et al. (2014) obtained similar results when spruce sawdust was torrefied at 240–340 °C.

Table 4.4. Particle size, bulk, and particle density of raw and torrefied WSS at different temperatures.

| Temperature (°C) | Time (min) | d _{gw} (mm) | S _{gw} (mm) | Bulk Density (kg/m ³) | Particle Density (kg/m ³) |
|------------------|------------|----------------------|----------------------|-----------------------------------|---------------------------------------|
| Untreated | - | 0.36 | 0.28 | 199.75 | 1054.56 |
| 200 | 5 | 0.34 | 0.22 | 167.24 | 1250.68 |
| 200 | 7 | 0.34 | 0.22 | 160.32 | 1277.79 |
| 200 | 9 | 0.35 | 0.17 | 158.56 | 1293.90 |
| 250 | 5 | 0.34 | 0.18 | 155.67 | 1345.11 |
| 250 | 7 | 0.33 | 0.16 | 150.00 | 1357.26 |
| 250 | 9 | 0.33 | 0.14 | 147.87 | 1365.37 |
| 300 | 5 | 0.34 | 0.17 | 120.44 | 1406.59 |
| 300 | 7 | 0.32 | 0.16 | 116.23 | 1426.73 |
| 300 | 9 | 0.32 | 0.14 | 110.24 | 1454.84 |

NB: d_{gw} denotes the geometric mean diameter of WSS; S_{gw} denotes the geometric standard deviation of WSS.

4.4.2.4 Elemental and proximate analysis

Table S4.1 depicts that SF affected the elemental and proximate contents of WSS on a dry-matter basis. The carbon (C), hydrogen (H), sulfur (S), and nitrogen (N) contents of the raw WSS were 47.47%, 6.66%, 0.21%, and 0.11%, respectively, while the amount of oxygen (O) present in the raw WSS was determined by difference to be 45.41%. After the pretreatment operation, the hydrogen and oxygen contents of the samples decreased, while the quantities of carbon and nitrogen present in the biochar significantly increased ($p < 0.05$) as the SF increased. Zanzi et al. (2022) reported a similar trend when miscanthus was torrefied. Additionally, in their respective studies on torrefaction of sawdust, barley straw, and oat hull, Alizadeh et al. (2022), Sarker et al. (2020), and Valdez et al. (2021) made similar observations in which an increase in severity led to increases in C and decreases in H and O. Generally, during the torrefaction pretreatment, the

energy density of the biochar (residual carbon-rich solids) increases due to the volatilization of the smaller hydrocarbon compounds, which have less energy density. In this study, the decrease in H and O was assumed to be due to the formation of carbon dioxide and water during the process. The changes occurring elementally in the biomass are usually demonstrated by the Van Krevelen diagram. The theory of the Van Krevelen diagram of the untreated WSS and WSS biochar is illustrated in Figure 4.5. The atomic H/C and the atomic O/C proportions follow an almost linear relationship. The O/C and H/C proportions also reduced as the severity increased, which resulted in a lower production of smoke and water vapor during burning. The energy loss was also reduced during combustion because of the reduction in the proportions of O/C and H/C in the WSS biochar.

The characterization of solid biomass fuels showed that FC and VM were the two significant measures. Fuels with a higher VM normally have a lower FC, resulting in a lower calorific value and a higher reactivity. Subsequently, when the amount of FC in the solid fuel is very large, the VM value is usually very low, leading to an improvement in the energy value. In this study, it was observed that as the SF intensified, the VM reduced, while the FC increased. The drastic reduction in VM may be due to partial carbonization and dehydration during the torrefaction process. Similar trends were observed by other researchers (Alizadeh et al. 2022; Sarker et al. 2020; Bridgeman et al. 2008; Lu et al. 2012; Wannapeera et al. 2011).

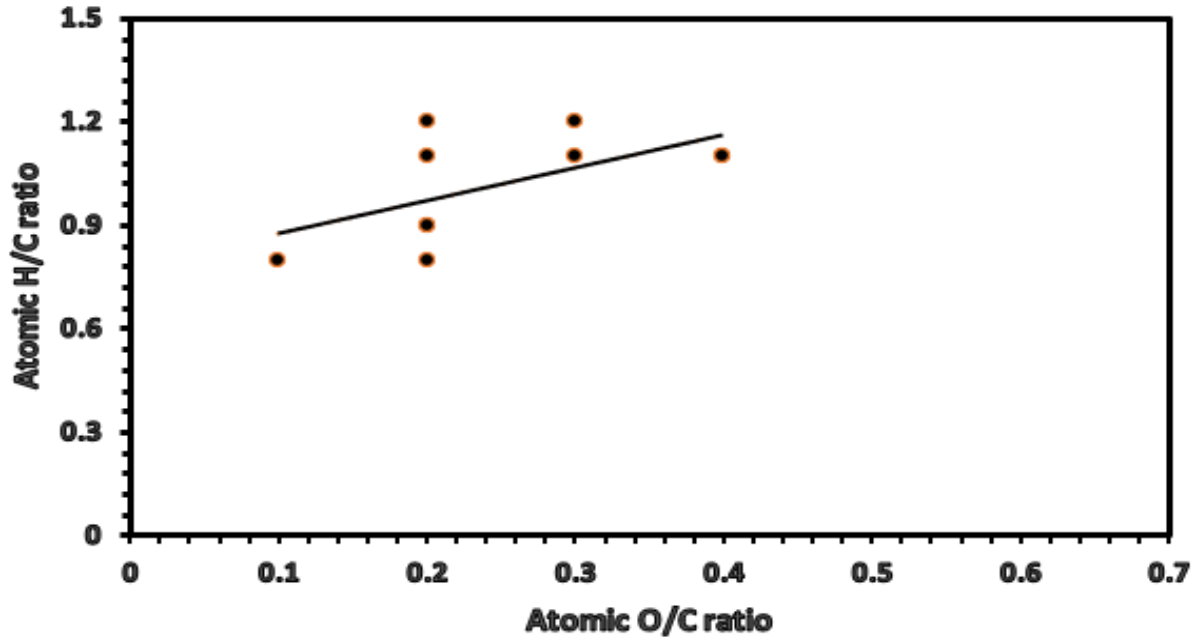


Figure 4.5. Van Krevelen diagram of untreated and WSS biochar.

4.4.2.5 Energy value and chemical composition of untreated and WSS biochar

The energy value of biochar, simply known as the calorific or higher heating value (HHV) of biochar, is the total amount of energy produced as heat when the biochar is subjected to complete combustion. The biomass loses more hydrogen and oxygen content compared to the carbon content during the pretreatment operation, which improves the HHV of the resulting product. Recent studies (Onyenwoke et al. 2022; Alizadeh et al. 2022; Sarker et al. 2020) have shown that an increase in FC leads to an enhancement of the calorific value. Figure 4.6 represents the higher heating value and chemical content of the untreated and treated WSS. The HHV improved as the SF intensified. The HHV of the untreated WSS was 18.21 MJ/kg, which increased by approximately 25% as the torrefaction severity increased. Chen et al. (2015b) reported that the HHV of a torrefied sample improved with the severity condition of torrefaction. Sarker et al. (2020) recorded an HHV between 22 and 25 MJ/kg when selected agricultural biomasses were torrefied.

The major composition of lignocellulosic biomass is cellulose, hemicellulose, lignin, and minor traces of extractives. These structures have different thermal stability, and the level of their chemical alteration is subject to the type of feedstock and torrefaction conditions used. Tumuluru et al. (2011) reported three temperature zones that were observed during the torrefaction operation: (1) non-reactive (50–150 °C); (2) reactive (150–200 °C); and (3) destructive (200–300 °C). The different occurrences in these zones were the dehydration, devolatilization, and carbonization of hemicellulose and, finally, the depolymerization and softening of lignin and the depolymerization devolatilization of cellulose. These reactions significantly improved the physiochemical properties and energy content of the biomass (Tumuluru et al. 2011; Tumuluru et al. 2021).

The lignocellulosic component analysis of the WSS at different SFs is shown in Figure 4.6. Through the torrefaction operation, a huge mass loss was observed. This mass loss was linked with the breakdown of hemicellulose. Cellulose also undergoes decomposition, similar to hemicellulose but to a lesser extent. Lignin is difficult to decompose during torrefaction because of its higher thermal decomposition temperature. The hemicellulose content drastically reduced to approximately 1.8% and the cellulose content reduced by approximately 10%, while the lignin gained approximately 35% as the severity increased. Results from other researchers agree with these findings (Phusunti et al. 2018; Oyebode and Ogunsuyi 2021; Baicar et al. 2018).

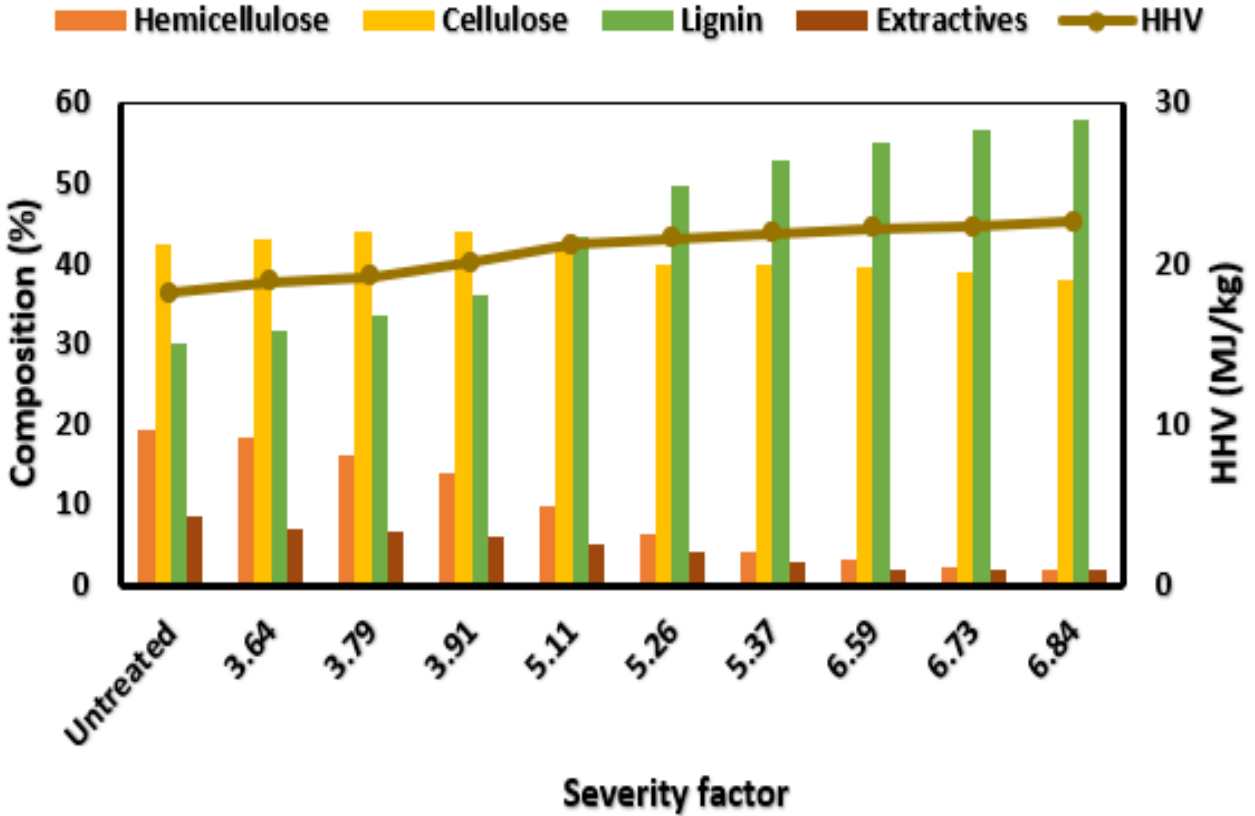


Figure 4.6. HHV and chemical composition of untreated and treated WSS.

4.4.3 Pellet properties

Peng et al. (2013) reported that the pelletization of torrefied samples was difficult when compared with untreated samples. However, the addition of binders not only makes pelletization much easier but also improved the pellet properties (Peng et al. 2015; Alizadeh et al. 2022). The effect of additives during pelletization was investigated and is described in the following subsections.

4.4.3.1 Pellet unit density and dimensional stability (DS)

Figure 4.7 represents the pellet unit density and the weight loss at different SFs. The pellet density was observed to have decreased after the torrefaction pretreatment process. The pellet unit density reduced to approximately 915.42 kg/m³ from 1048.56 kg/m³ at a weight loss of 18%,

while the mass loss through the torrefaction process of the WSS led to lower pellet densities of the torrefied WSS (Poudel et al. 2018). In their study, Peng et al. (2014) reported that at a 30% weight loss, the pellet density decreased to 930.01 kg/m³ from 1190.02 kg/m³ and from 1230.11 kg/m³ to 970.02 kg/m³ for commercial white and brown wood pellets, respectively. Alizadeh et al. (2022) also reported a reduction in the pellet unit value from 1044.61 kg/m³ to 955.96 kg/m³ for a higher SF.

Table S4.2 depicts the pellet properties of the torrefied WSS, indicating that the DS of the pellets improved as the torrefaction severity increased. The DS provides information about the firmness of the produced pellets through handling, transportation, and storage. When lesser values are obtained during this test, it shows that the pellet stability is good, while large values indicate that the pellet is unstable. When positive and negative values are obtained, it indicates that expansion and contraction were observed in the pellet, respectively. Results from other researchers agree with these findings (Onyenwoke et al. 2022; Alizadeh et al. 2022. Valdez et al. 2021).



Figure 4.7. Pellet unit density and weight loss of raw WSS and WSS biochar for different severity factors.

4.4.3.2 Pellet tensile strength

The tensile strength and particle density are two major characteristics that define the quality of biomass pellets (Wang et al. 2020). Pellet tensile strength is an important property of biomass pellets that expresses the force required to crush or damage a pellet. Generally, the results obtained in this study showed a decrease in value as the torrefaction severity increased. Hemicellulose and cellulose polymer degradation was believed to be one reason for the tensile strength loss of biomass pellets under severe torrefaction treatment (PFI 2011). Shang et al.

(2012) reported that the pellet tensile strength reduced by 90% after severe torrefaction. Figure 4.8 shows the pellet tensile strength after torrefaction. The tensile strength of the raw WSS pellet was 1.68 MPa, which slightly increased to 1.82 MPa after light torrefaction, reduced to 1.23 MPa after mild torrefaction, and decreased further to 0.78 MPa when the torrefaction became severe. This was possibly due to the variance in the thermal stability of the WSS structures and the degree of their chemical changes in different stages of torrefaction.

To improve the strength of the torrefied sawdust pellets in this study, the utilization of cheap and non-food-based binders was explored. White spruce sawdust and torrefaction liquid (TL) were introduced to the pelletization step in different formulations after the microwave-assisted torrefaction of the WSS. Figure 4.8 represents the tensile strength of the pellets with and without binders. It was observed that the additives improved the binding mechanism between torrefied particles during densification, which improved the tensile strength of the pellet. This agrees with other researchers (Bai et al. 2018; Kanwal et al. 2019; Sarker et al. 2021) who conducted similar works using different binders in pellet making.

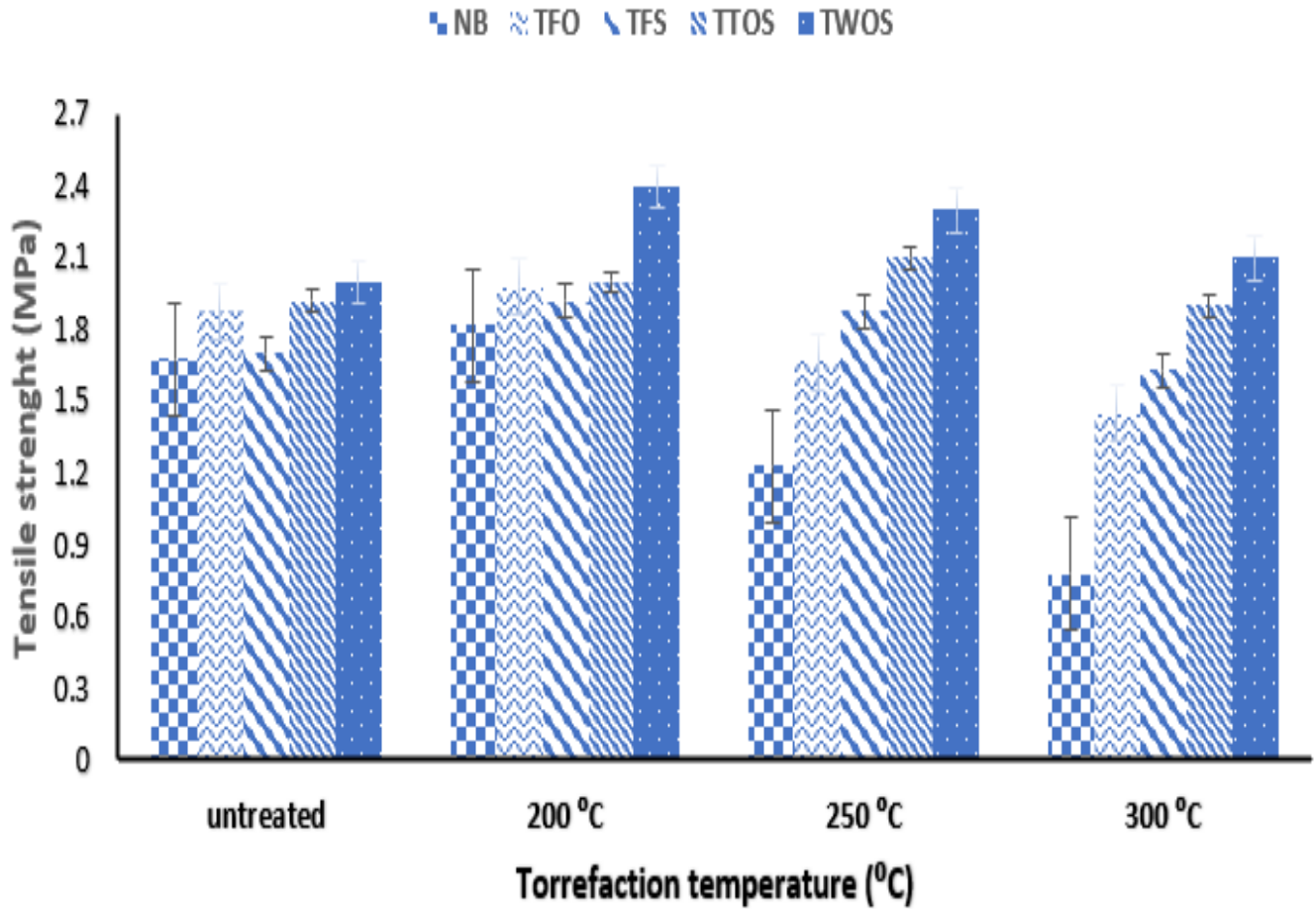


Figure 4.8. Tensile strength of pellets from WSS untreated and torrefied with or without binders. NB: Without binder; TFO: 75% torrefied WSS with 25% TL; TFS: 75% torrefied WSS with 25% sawdust; TTOS: 33.3% torrefied WSS with 33.3% sawdust and TL; TWOS: 75% torrefied WSS with 12.5% sawdust and TL each.

4.4.3.3 Moisture uptake test

Due to the different weather circumstances in Canada for the storage and transportation of biomass pellets, it was necessary to conduct a moisture uptake test. The pellets might be exposed to harsh weather conditions which could affect their quality. Figure 4.9 demonstrates the absorption results for pellets from different torrefaction temperatures. During the experiment, the untreated pellets were observed to have enlarged and distorted within a short time in the humidifier chamber. Although the torrefied WSS pellets maintained a good form of firmness,

they also swelled and disintegrated after some hours in the chamber. The moisture absorption of torrefied sample pellets compared to untreated pellets was previously studied by other researchers (Rudolfsson et al. 2017, Ghiasi et al. 2014; Zhang et al. 2020). During the torrefaction operation, the OH groups in the biomass are destroyed to a limited degree by dehydration, hindering the formation of H bonds and thereby encouraging the pretreated biomass to become hydrophobic (Tumurulu et al. 2021). In general, the WSS biochar pellets treated under severe torrefaction had a low moisture uptake capacity, which may be due to the more intensive breakdown of polymers and the decrease in H bonding sites (Wang et al. 2020). For example, the WSS torrefied at 200 °C in which approximately 6% of the hemicellulose was degraded had a moisture uptake of up to 10%, while the WSS torrefied at 300 °C was able to take up less than 5% moisture due to the degradation of almost 60% of the hemicellulose. The effect of the binders on the water absorption rate of the pellets was also examined; these results are displayed in Figure S4.2. The introduction of TL and sawdust to the biochar pellet formulation ameliorated the surface structure by enhancing the adhesive mechanism of the biochar particles. This lessened the microporous surface tension, which decreased the uptake rate of the available water (Onyenwoke et al. 2022; Kambo and Dutta 2014).

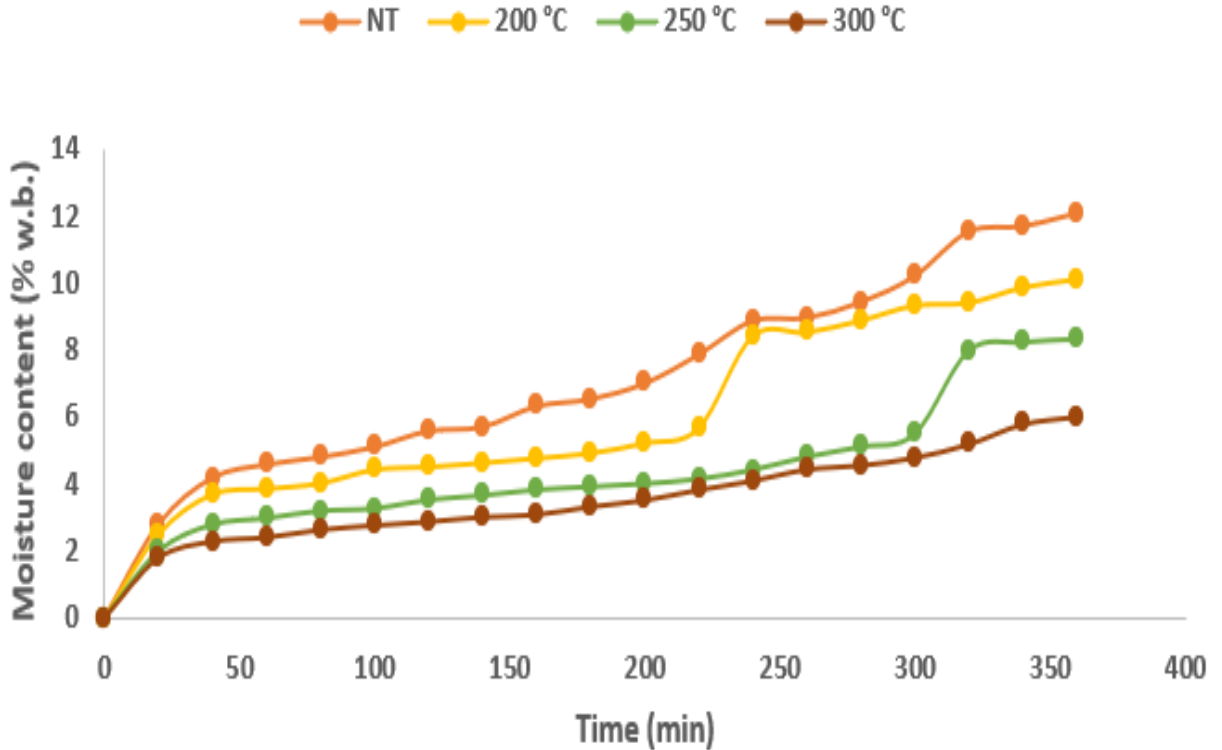


Figure 4.9. Moisture absorption of torrefied and non-torrefied WSS pellets.

4.4.3.4 SEM analysis

The binding mechanism of the pelleted biochar particles was further analyzed with the help of scanning electron microscopy (SEM), which aided in the observation of the internal stratification of the pellets formed. The microscale effect of the pretreatment process on the WSS was justified by a microstructural examination, which provided a perception of the structural pattern of the biochar pellet samples. Figure 4.10a–d shows the SEM micrograph images of the WSS pellets. Figure 4.10a presents the micrographs of the untreated WSS pellet, while Figure 4.10b, c and d shows the micrograph of the torrefied pellets at temperatures of 200 °C, 250 °C, and 300 °C, respectively. It was observed that the torrefied WSS pellets showed more firmly glued surfaces with fewer pore spaces when set side by side with the raw pellets. The WSS biochar pellets also showed less porosity when compared with raw pellets. This may be due to

the breakdown of and depolymerization of the hemicellulose and cellulose matrix through the torrefaction of the WSS. The structural breakdown exposes more surface area, which is suitable for pelletization. Similar observations were reported by other researchers (Gastaldi et al. 1998; Kristensen et al. 2008). The micrographs of the untreated WSS pellet and the torrefied pellets at a higher magnification are presented in Figures S4.3–S4.6, respectively.

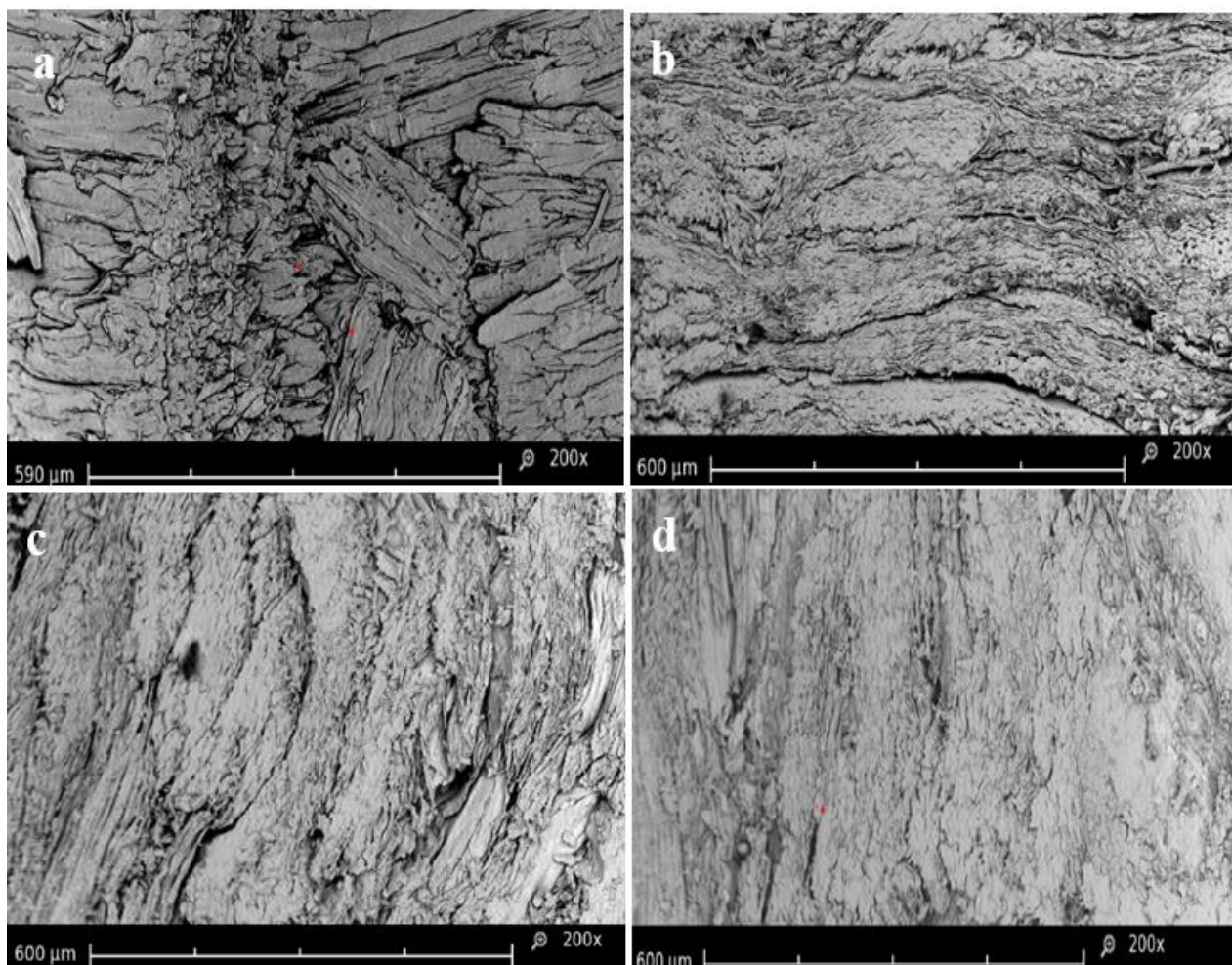


Figure 4.10. SEM micrographs images of WSS pellets: (a) un-torrefied WSS; (b) WSS torrefied at 200 °C; (c) WSS torrefied at 250 °C; (d) WSS torrefied at 300 °C.

4.4.3.5 Differential thermogravimetric analysis (DTG)

To determine the heat stability of the WSS pellet, a DTG analysis was conducted. The degradation, decomposition, and chemical stability of biomass pellets are critical indicators. The DTG analysis graph of the raw and torrefied WSS pellets are presented in Figure 4.11a, b. It was observed that the WSS pellets displayed a similar thermal degradation trail for the studied samples. Four identical regions were observed. The first region is where weight loss occurred at 50–150 °C due to the evaporation of free water and the escape of organic substances with low boiling points (Kang et al. 2018). The second region was the breakdown of hemicellulose and the escape of volatile matter, noted at a temperature of 200–235 °C. The third region was the breakdown of cellulose (250–275 °C), while the final region referred to the oxidation of char at high temperatures (Sarker et al. 2021). This implies that the torrefaction of the WSS slightly reduced its thermal stability. Sarker et al. (2021) made a similar observation when a DTG analysis of some torrefied agricultural residues was performed.

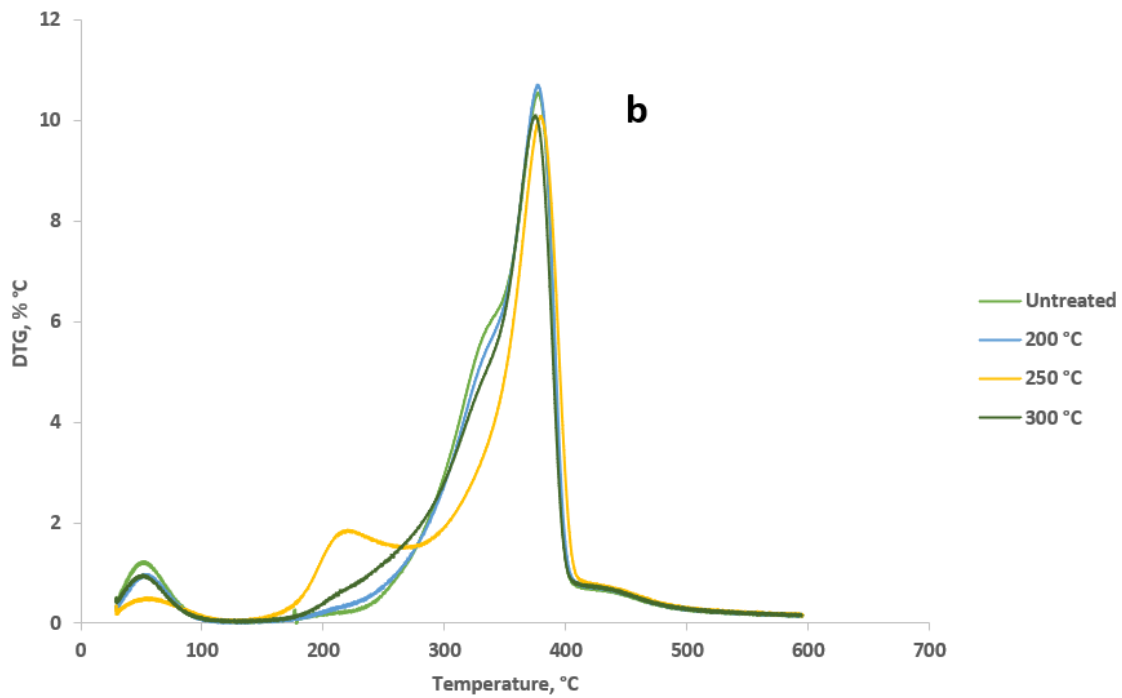
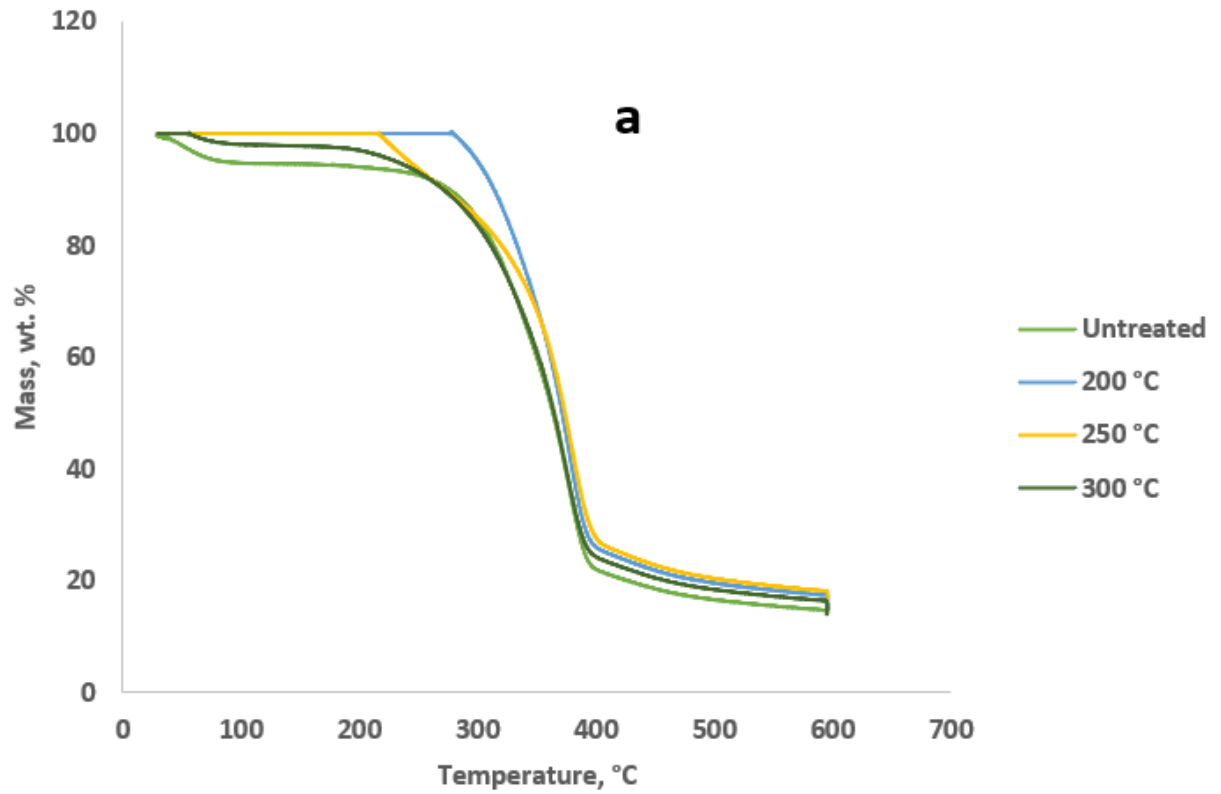


Figure 4.11. The DTG analysis graph of the raw and torrefied WSS: (a) thermogravimetry curves of the untreated and torrefied WSS; (b) differential thermogravimetry curves of the untreated, torrefied WSS.

4.5 Conclusions

The torrefaction pretreatment of white spruce sawdust (WSS) was conducted using a lab-scale microwave oven at temperatures in the range of 200–300 °C and a retention time of 5–9 min in an inert environment to ascertain the effect of these two process variables on the physiochemical properties of the WSS. The products of this process are solid carbon, commonly known as biochar, condensable torrefaction liquid (TL), and non-condensable gases. The result from this study indicates the potential for the torrefaction pretreatment operation to enhance the physiochemical properties of WSS. The hemicellulose content underwent a major degradation as the torrefaction severity increased. Similarly, cellulose also underwent decomposition, but with less degradation. The incomplete depolymerization of cellulose and the disintegration of hemicellulose at the same time increased the lignin content during the pretreatment process, enhancing the FC and drastically decreasing the VM content. This led to an improvement in the higher heating value (HHV), hydrophobicity, bulk, particle density, pellet dimensional stability, and pellet density.

During the torrefaction process, a significant mass loss associated with the decomposition of hemicellulose was observed. The hemicellulose content drastically reduced to approximately 1.8% and the cellulose content reduced by approximately 10%, while the lignin gained approximately 35% as the severity increased. The pellet unit density and pellet tensile strength in this study decreased as the torrefaction severity increased. The breakdown of the hemicellulose and cellulose polymer was suggested to be one reason for the tensile strength loss of the biomass pellets under severe torrefaction treatment. However, the tensile strength of the studied pellets was improved by the introduction of binders. The OH groups in the biomass are destroyed to a limited degree by dehydration. This hinders the formation of H bonds, thereby encouraging the

pretreated biomass to become hydrophobic. From the SEM graphs, it was observed that the torrefied WSS pellets showed more firmly glued surfaces with fewer pore spaces when compared with the raw pellets. The thermogravimetric analysis conducted showed that the torrefaction of the WSS slightly reduced its thermal stability.

Chapter 5

Combined Effects of Torrefaction and Binders on the Quality of Pellets Produced from Oat Straw

- A similar version of this chapter has been published in the journal *BioEnergy Research*: Onyenwoke, C., L.G. Tabil, T. Dumonceaux, D. Cree, and E. Mupondwa. 2023. Combined Effects of torrefaction and binders on the quality of pellets produced from oat straw. *Bioenergy Research*. 16:2357–2370. <https://doi.org/10.1007/s12155-023-10606-6>.

Contribution of the Ph.D. Candidate

The experimental work, data collection and analysis, original drafting, review and editing of the manuscript was done by the Ph.D. candidate, while Drs. L.G. Tabil, T. Dumonceaux, E. Mupondwa, D. Cree, and P. Adapa, provided resources and editorial input. Drs. L.G. Tabil and T. Dumonceaux performed the supervisory roles, while Drs. L.G. Tabil, T. Dumonceaux E. Mupondwa and D. Cree also carried out the project administrative roles.

Contribution of the Paper to the Overall Study

This paper addressed the third objective of this study: to investigate the combined effect of pretreatment and binders on the quality of pellets formed. This study evaluates the physiochemical characteristics of the pellets formed from torrefied oat straw such as measuring tensile strength, moisture uptake rate, bulk density, pellet density, torrefaction liquid analysis and the scanning electron microscope (SEM) analysis. Also, the chemical components and thermochemical analysis of the pellets were measured. These include ultimate analysis, proximate analysis, and measuring of the high heating value. Furthermore, this study determined

the combined effect of pretreatment and binders on the physiochemical properties of pellet. The results of this study showed that torrefaction pretreatment of oat straw led to an improvement in the physiochemical properties of pellet. However, the pellet tensile strength decreased as the torrefaction severity increased. The energy efficiency of this system was improved by the addition of the substance lost during torrefaction as a heat source. The results of this research can be useful to bioenergy plants.

5.1 Abstract

This study was carried out to investigate the combined effects of torrefaction and binders on the quality of pellets produced from oat straw. Torrefaction pretreatment was performed with the aid of a lab-scale microwave oven at temperatures in the range of 200-300 °C and a retention time of 5-9 min in an inert environment to ascertain the effect of these two process variables on the physiochemical, thermal, and mechanical properties of the torrefied biomass. Torrefaction liquid (TL) and sawdust were introduced during pelletization as binding agents in the pellet formulation. From the results in this study, the physiochemical and thermal properties of the oat straw pellet improved after torrefaction except for the tensile strength. The higher heating value (HHV), particle density, and bulk density of the torrefied oat straw improved from 16.84-24.23 MJ/kg, 882.23-1374.87 kg/m³, and 127.87-80.23 kg/m³, respectively. The tensile strength of the pellet after torrefaction ranged between 0.35-1.02 MPa. The introduction of the binder during pelletization improved the pellet tensile strength to about 1.32- 2.28 MPa. Although ash content increased after torrefaction from 5.32-10.22%, there was no significant increase in the ash content after the addition of binders as the value ranged between 8.88-9.00% for the torrefied pellet sample used. The addition of binder further improved the HHV, pellet density, dimensional

stability, and moisture uptake rate from approximately 21-24 MJ/kg, 1032-1393 kg/m³, 2.23-1.02%, and 16-9%, respectively for the torrefied pellet sample used. The energy efficiency of this system was improved by the addition of the substance lost during torrefaction as a heat source.

5.2 Introduction

Canada is a major supplier of oat globally. The global yield of oats in 2021, amounted to about 22.68 Mt (Shahbandeh 2022a). Oat is the second major cultivated cereal crop in Saskatchewan (SK) Canada behind wheat, with a yield of oats in Canada in 2020 of approximately 4.6 Mt (Shahbandeh 2022b). This represents an increase from about 4.23 Mt produced in 2019. The massive production of agricultural outputs leads to a great volume of biomass residue in the field, and the common practice is to leave this residue on the farm after collecting the grain. A minor amount of the residue is utilized in most cases for feeding animals and sheltering these animals, or as mulch to retain moisture in the soil. Although animal consumption forms the majority of oat use globally, because of the awareness of the nutritional benefits of whole grains, the human food market for oats is growing. Most Canadian oats are used for human consumption (POGA 2022).

A huge quantity of waste is generated globally in agri-food processing and agriculture. Husks, shells, straws, and biomass from dedicated energy crops produced yearly are approximately 50 billion tonnes (Szufa et al 2020). Hence, it is essential to find an economically and ecologically sound medium to transform this biomass and its residues into raw materials to produce valuable energy. The utilization of agricultural waste, in this case oat straw for producing biofuel addresses the food versus fuel debate and improves the existing crops (Adapa et al. 2011). Oat straw can serve as a feedstock (raw material) for producing biofuel and bioproducts. The

challenges linked with the utilization of raw biomass that can be detrimental to their industrial usage include too much moisture content, poor grindability, low bulk density, inherently low energy value, and hydrophilicity. These problems affect the storage, handling, transportation, and energy efficiency of this raw biomass (Azargohar et al. 2019a; Azargohar et al. 2019b; Haykiri-Acma et al. 2016; Medic et al. 2012). To overcome these constraints, several pretreatment methods have been investigated for economic cost and environmentally friendly strategies to overcome this drawback caused by lignocellulose matrix to cellulose breakdown (Onyenwoke et al. 2022; Onyenwoke et al. 2023).

Torrefaction is a heat operation of feedstock, associated with the roasting of the biomass sample in a controlled environment at torrefaction temperatures ranging from 200–300 °C. Torrefaction is commonly known as mild pyrolysis; this pretreatment process has been used to make biomass more hydrophobic, reduce moisture content, and increase the amount of energy produced by the biomass, which improves the volumetric energy density of biomass. Torrefaction yields are both the solid fuel (torrefied material) and the torgas. The torgas comprise of torrefaction liquid (TL), and non-condensable gases. The TL is rich in furfural, ketones, organic acids, water, and other minor components, which could be transformed into commercially feasible products (Tumuluru et al. 2011).

Torrefied material is a brown to dark solid yield product that originated from torrefaction or pyrolysis of biomass. It is environmentally safe and could be molded as biofuel pellets for energy production, greenhouse gas reduction, and waste management (Agu et al. 2021; Cha et al. 2016; Hu et al. 2021). Usually, during the densification of torrefied biomass, additives are introduced as a form of binder to improve particle-to-particle adhesion forces. Due to the hardening of the particles of the torrefied biomass and the breaking of the hydroxyl bonds, the efficiency in the

compaction process decreases considerably. The frequently used binders include starch and lignin, but they are expensive, and starch has competing use as food ingredients (Peng et al. 2015) Hence, to improve the thermal and physiochemical properties of solid biofuel pellets from torrefied oat straw, this work used the TL from this process as a binder. This strategy aims to solve the difficulty relating to the promotion of systems and products and equaling energy efficiency and fuel quality. The torrefaction gases (condensable liquid) contain a large amount of energy. Therefore, introducing a closed recycling system has the potential to be economically viable for the torrefaction system. It is significant that this energy is utilized productively in the system (torrefaction and densification) to approach and settle the process stability, system reliability, and safety problems currently averting commercial-scale torrefaction. Sawdust is abundantly available, Peng et al. (2015) recommended it as an economical and effective binder for densification of torrefied material as it is less expensive than lignin and starch. Although sawdust is the main raw material for wood pellet production worldwide, due to its surplus availability, low-cost, effectiveness, and lack of use as a food ingredient it was considered as a binder in this study. The potential of sawdust as a binder has been studied by some researchers (Onyenwoke et al. 2023; Peng et al. 2015; Rahaman and Salam 2017; Alizadeh et al. 2022). This study emphasized the consequence of torrefaction and binders on the mechanical and physiochemical characteristics of pellets formed from torrefied oat straw torrefied material.

5.3 Materials and Methods

5.3.1 Materials

The oat straw that was used for this study was collected from the black soil zone of Maymont in Saskatchewan (SK), Canada. The crop was harvested with a conventional combine and swathed,

and the straw was baled in mid-October 2019. The oat straw variety is Morgan and its moisture content when collected was about 10% (w.b.). Samples were size reduced using a hammermill with a 3.2 mm sieve. The sawdust used as an additive during pellitization was received from NorSask Forest Products Inc. sawmill section near Meadow Lake, Saskatchewan (SK), Canada in 2020. The amount of available moisture in the sawdust sample when received was recorded to be approximately 42% (w.b.). The oat straw (ground) sample as well as the sawdust (binder) were screened to remove irregular particles and then preserved in a fastened plastic container in a conditioned space at a temperature of approximately 4 °C to be used for experimentation.

5.3.2 Experimental procedure

Torrefaction pretreatment of oat straw was carried out to improve its physicochemical and thermal characteristics. Figure 5.1 depicts a schematic diagram of the microwave torrefaction setup. A microwave oven (Cober Electronics, Inc., Norwalk, CT, USA) was used to carry out this experiment. About 100 g of raw sample was introduced into the reactor (500 mL borosilicate vessel), and nitrogen gas was purged into the reactor to remove air. An initial power level of 550 W was set at the beginning of each experiment. Once the required temperature was attained (200, 250, and 300 °C), the microwave power was controlled to hold the temperature until the desired residence time (5, 7, and 9 min) was reached. Equation (5.1) was used to estimate the stage of the pretreatment process (Overend et al. 1987), which is also known as the severity factor (SF). The experiments were performed in triplicate and the results averaged and recorded. Table S5.1 in the supplementary material shows the SF calculated from Equation (5.1).

The products of the torrefaction are solid residue (torrefied oat straw), torrefaction liquid (TL), and non-condensable gas. The torrefaction product yields were estimated using Equation (5.2), (5.3), and (5.4).

$$SF = \log \int \exp \left[\frac{T - 100}{14.75} \right] dt \quad (5.1)$$

where SF represents the intensity of the pretreatment operation (severity factor), T represents the temperature of the pretreatment process (°C), and t represents the residence time of operation (min).

$$M_{SR} = \frac{M_{TS}}{M_I} \times 100 \quad (5.2)$$

$$M_{TL} = \frac{M_{LS}}{M_I} \times 100 \quad (5.3)$$

$$M_{GY} = M_I - (M_{SR} + M_{TL}) \quad (5.4)$$

where M_{SR} , M_{TL} , and M_{GY} represent solid (%), liquid (%) and gas yield (%), respectively. M_{TS} represents mass of torrefied sample (g), M_{LS} represents mass of liquid sample (g), M_I represents mass of raw sample (g).

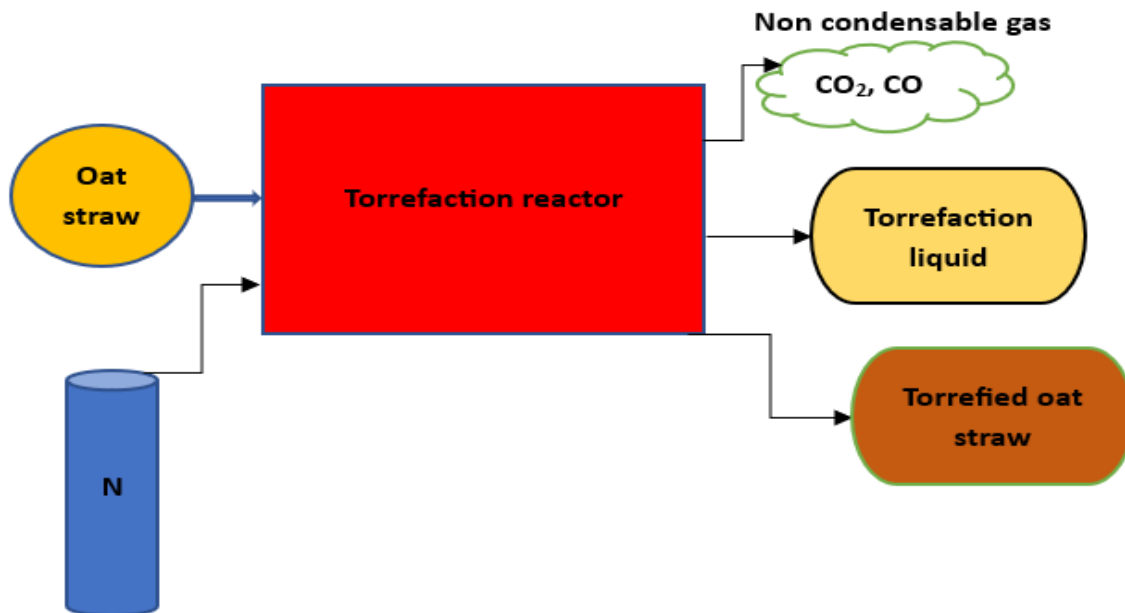


Figure 5.1 Schematic diagram of microwave torrefaction setup.

5.3.2.1 Experimental design

Minitab statistical software version 21.1.0 was used to perform the factorial design of two independent variables (temperature (°C) and residence (min)) three levels were considered for each factor (200, 250, 300 °C, and 5, 7, and 9 min, respectively). Analysis of variance (ANOVA) was used to determine the effect of the pretreatment operation on the response variables (durability, higher heating value (HHV), and pellet density) at 5% level of significance.

5.3.3 Characterization of raw and torrefied material

5.3.3.1 Proximate and ultimate analysis

The proximate analysis of the ground sample (control) and torrefied material was evaluated by the ASTM standard procedures (ASTM 2010; ASTM 1996; ASTM 1997). Fixed carbon (FC) content in the samples was determined by differences (Bai et al. 2018) as given in Equation (5.5).

An Elementar analyzer (Vario EL III, Elementar Americas Inc., Ronkonkoma, NY, USA) was used to estimate the quantity of carbon (C), hydrogen (H), nitrogen (N), and sulfur (S) present in the raw oat straw sample and torrefied oat straw samples. Initially, sulfanilic acid was used to calibrate the equipment as a standard. Then, about 5 mg of the materials was collected in a container and heated with the aid of muffle furnace for analysis. The oxygen content in the samples were evaluated by difference.

$$\text{Fixed Carbon} = 100 - (\text{ash content} + \text{moisture content} + \text{volatile mater}) \quad (5.5)$$

5.3.3.2 Chemical component analysis

ANKOM procedures were used to estimate the chemical content of the torrefied material and the untreated oat straw ANKOM method 5, 6, and 8 (ANKOM 2022a; ANKOM 2022b; ANKOM 2022c) were used to evaluate the acid detergent fiber (ADF), neutral detergent fiber (NDF), and acid detergent lignin (ADL), respectively. ADL contains lignin alone, while hemicellulose and cellulose contents were determined by subtracting ADF from NDF and subtracting ADL from ADF, respectively.

5.3.3.3 Torrefaction liquid (TL) analysis

A gas chromatography-mass spectrophotometer (GC-MS) Agilent 7890 B instrument was used in the analysis of functionalities available in the condensed liquor products after torrefaction with the aid of DB-35MS column (30 m×0.32 mm×0.25 μm). After introduction of the torrefaction liquid sample, the column was heated to approximately 280 °C at a heating rate of 5 °C min⁻¹.

Helium (He) was used in this process as carrier gas at a flow rate of 1 mL min⁻¹ in a divaricate condition. This experiment was performed in triplicate and the results averaged and recorded.

5.3.3.4 Particle size analysis

The particle size analysis of torrefied and untreated oat straw was examined by following the ASABE procedure (2006) About 100 g of the sample was placed on the top layer of a stacked U.S. sieve numbers 16, 20, 30, 50, 70, and 100, then mounted on a Ro-Tap mechanical sieve shaker (W.S. Taylor Inc., Mentor, OH, USA) and allowed to run for approximately 10 min. This experiment was performed in triplicate and the results averaged and recorded. The geometric mean diameter (d_{gw}) and standard deviation of particle diameter (S_{gw}) were evaluated using Equations (5.6) and (5.7), respectively.

$$d_{gw} = \log^{-1} \left[\frac{\sum_{i=1}^n (W_i \log d_i)}{\sum_{i=1}^n W_i} \right] \quad (5.6)$$

$$S_{gw} = \frac{1}{2} d_{gw} \left[\log^{-1} S_{log} - (\log^{-1} S_{log})^{-1} \right] \quad (5.7)$$

where d_{gw} represents the geometric mean diameter (mm); n represents the number of sieves + 1 pan; W_i is the mass on the i^{th} sieve (g); d_i denotes the nominal sieve opening size of the i^{th} sieve (mm); S_{log} denotes the geometric standard deviation of log-normal distribution by mass in the common (base 10) logarithm.

5.3.3.5 Bulk and particle density

Bulk and particle density of torrefied and untreated oat straw samples were determined with the aid of standard 0.5 L cylindrical cup (SWA951, Superior Scale Co. Ltd., Winnipeg, MB, Canada) and gas displacement pycnometer (AccuPyc 1340, Micromeritics Instruments Corp.,

Norcross, GA, USA), respectively. A funnel was used to introduce the samples to the center of the cylindrical cup, the bulk density in kg/m^3 was calculated by the mass per unit volume.

5.3.4 Pelletization of samples

The solid material (torrefied oat straw) after torrefaction was pelletized to explore the influence of residence time and torrefaction temperature on the pellets formed. The effect of binders on pelletization was also studied using different composition of binders (TL and sawdust) Torrefied material at 250 °C temperature and 9 min retention time was selected for the effect of binders on pelletization experiment Table 5.1 shows different formulations that were pelletized to analyze the outcome of binders on the physiochemical and mechanical characteristics of the respective pellets. Pelletization of biomass samples were executed using a single pellet press system mounted to an Instron (Model No. 3366, Instron Corp., Norwood, MA, USA). The system consists of a plunger, thermal element, and a cylindrical die. The plunger which was attached to the motion crosshead of the Instron supplied the compressive pressure required for the compaction, the heating element was tied round the cylindrical die to provide the needed heat (95 °C) and the sample of about 0.75 g was introduced into cylindrical die. The cylindrical die was opened to eject the produced pellet after the required compressive pressure (approximately 128 MPa at a rate of 50 mm/min) was attained.

Table 5.1. Formulations for pelletizing torrefied oat straw (torrefied material) with a torrefaction liquid and sawdust.

| Composition | Torrefied Material (%) | Torrefaction Liquid (%) | Sawdust (%) |
|-------------|------------------------|-------------------------|-------------|
| NTB | - | - | - |
| WBC | 100 | - | - |
| BCSO3 | 33.3 | 33.3 | 33.3 |
| BCSO2 | 75 | 12.5 | 12.5 |
| BCO | 75 | 25 | - |
| BCS | 75 | - | 25 |

NTB: Untreated material without binder; WBC: torrefied material without binder; BCSO3: 33.3 % each of torrefied material, sawdust, and TL; BCSO2: 75% torrefied material with 12.5% each of sawdust and TL as binders; BCO: 75% torrefied material with 25% TL as the binder; BCS: 75% torrefied material with 25% of sawdust as the binder.

5.3.5 Pellet quality analysis

5.3.5.1 Unit density and dimensional stability

The unit density and dimensional stability of the formed pellet at different torrefaction severities was determined. The pellet density is the ratio between the unit weight of the pellet and its volume after production (V_0). The length and the diameter of the pellet was measured using a vernier caliper, while the weight of a single pellet was determined using a lab weighing balance. The individual measured pellet was stored in an airtight Ziploc bag for dimensional stability experiment. The pellets were kept for 14 d under a temperature and relative humidity of about 23 °C and 35%, respectively, the volumetric change after 14 d (V_{14}) was recorded for each pellet. About 10 pellets from each severity condition were used for this study and the result averaged. Equation (5.8) was used to compute the dimensional stability of the pellet.

$$\text{Dimensional stability (\%)} = \frac{V_{14} - V_0}{V_0} \times 100 \quad (5.8)$$

5.3.5.2 Diametral compression test

A diametral compression test was carried out to determine the tensile strength of the pellets formed. In this study the tensile strength was assumed to be a measure of the durability of the pellet. The single pellet system mounted to the Instron for pelletization was replaced with stationary and fixed load for the diametral compression test. First, the pellets were cut into tablets of approximately 2.5 mm, then a vernier caliper was used to measure the thickness (l) and diameter (d) of the tablets. The tablet was placed on the fixed load attached to the Instron while the stationary load (F) was applied until the failure occurred. The tensile strength (δ_x) of the material (MPa) was determined using Equation (5.9).

$$\delta_x = \frac{2F}{\pi dl} \quad (5.9)$$

5.3.5.3 Ash content and the higher heating value (HHV)

The ash content and the higher heating value (HHV) of the pellets formed from both the torrefied oat straw and untreated oat straw was carried out. The amount of ash present in these samples was determined by ASTM procedure (2010), while a bomb calorimeter (6400 Automatic Isoperibol, Parr Instrument Company, Moline, IL, USA) was used to evaluate the HHV of the samples. The protocol for this experiment includes burning about 0.5 g of a sample in a cylindrical metal filled with oxygen and submerged in a known quantity of water, all held in a chamber which is thermally insulated.

5.3.5.4 Moisture uptake test

A humidifier chamber (Espec SH-641 Benchtop chamber, ESPEC Corp., Osaka, Japan) set at a temperature of about 25 °C and relative humidity of 90% was used to test for the moisture uptake capability of the formed pellets. This experiment was important because when pellets are been transported, they sometimes absorb water and become distorted. The average weather condition in Canada for most of the transportation activities (Spring to Fall) is within a temperature range of 22-30 °C and relative humidity of about 60–85% (Bush and Lemmen 2019); in this study, a worst-case scenario was assumed. Approximately 4g of pellets (4-5 pellet samples) from different torrefaction conditions was used to carry out this experiment. The samples were kept in the humidifier for about 72 h, and data were collected at an interval of 10 min for the first hour, 30 min for the next 4 h, and an interval of an hour for the remaining time. The test was carried out in triplicate for each pellet sample and the result was averaged.

5.3.5.5 Scanning electron microscope (SEM) analysis

The surface morphological changes of the produced pellet were examined using a scanning electron microscope (SEM) (SEM Phenom-World, Eindhoven, The Netherlands) with a stereoscope. First the pellets were cut longitudinally and coated with gold of 10 nm thickness, then placed in a chamber in the vacuum.

5.4 Results and Discussion

5.4.1 Product yield composition

5.4.1.1 Effect of reaction time and temperature

Table 5.2 shows the effect of SF on the product yield distribution after torrefaction process. The effect of temperature and time during torrefaction process can best be described as torrefaction severity, the SF is a representation of both reaction time and temperature (Overend et al. 1987). Torrefaction reaction can be classified as light, mild, and severe with temperatures ranging from 200-235 °C, 235-275 °C, and 275-300 °C, respectively (Chen and Kuo 2011). Basically, during torrefaction pretreatment, the following reactions such as devolatilization, depolymerization, and carbonization of cellulose, hemicellulose, and lignin occurred (Tumuluru et al. 2011). Hemicellulose degraded faster than cellulose and lignin during torrefaction as shown in Table 5.3. This was due to its thermal decomposition temperature (215-315 °C). The product yield constitutes of solid residue also known as torrefied material, torrefaction liquid (condensable gas), and non-condensable gas. The solid residue yield reduced as the torrefaction severity intensified, but the case was different in the other two products where their yield increased with increasing severity. When the severity condition changed from light to severe, the torrefied material, liquid, and gas yield were modified by approximately 88.5, 9.88, and 1.62% to 67, 22.22, and 10.78%, respectively. Basically, during torrefaction pretreatment, at least about 70% of torrefied material is retained while 30% accounts for the condensable and non-condensable products (Tumuluru et al. 2011). During the pretreatment operation, the raw biomass lost a significant amount of moisture which resulted in a reduction in the quantity of torrefied material produced and increase in escape of volatile matter (Rousset et al. 2011).

Table 5.2. Effect of SF on product yield distribution during microwave-assisted torrefaction of oat straw.

| Temperature (°C) | Time (min) | Severity factor | Solid yield (%) | Liquid yield (%) | Gas yield (%) |
|------------------|------------|-----------------|-----------------|------------------|---------------|
| 200 | 5 | 3.64 | 88.50 | 9.88 | 1.62 |
| 200 | 7 | 3.79 | 85.40 | 11.58 | 3.02 |
| 200 | 9 | 3.91 | 82.24 | 13.01 | 4.75 |
| 250 | 5 | 5.11 | 80.83 | 14.26 | 4.91 |
| 250 | 7 | 5.26 | 77.45 | 16.00 | 6.55 |
| 250 | 9 | 5.37 | 75.72 | 17.02 | 7.26 |
| 300 | 5 | 6.59 | 72.38 | 18.98 | 8.64 |
| 300 | 7 | 6.73 | 70.56 | 19.76 | 9.68 |
| 300 | 9 | 6.84 | 67.00 | 22.22 | 10.78 |

5.4.2 Characterization of raw and torrefied material

5.4.2.1 Elemental composition, chemical composition, and higher heating value (HHV)

The major elements in biomass are oxygen (O), carbon (C), hydrogen (H), nitrogen (N), and sulfur (S). Table 5.3 depicts the HHV, elemental, and chemical content of oat straw torrefied material. It was observed that as the severity intensified, C content increased while O and H content reduced because of the emergence of carbon dioxide (CO₂), carbon monoxide (CO), and water (H₂O) during the pretreatment operation. At 300 °C the C content of the torrefied material increased to 52.65% from an untreated sample value of 44.24%, while the amount of O and H present in the torrefied material went down from 49.39% to 41.59% and 5.85% to 5.24% respectively, for 9 min retention time of torrefaction. In a similar manner Alizadeh et al. (2022), Sarker et al. (2020), and Valdez et al. (2021) studied torrefaction of sawdust, barely straw, and

oat hull, respectively and observed that increase in severity factor resulted in increase in C and decrease in O and H content. The N and S content of the torrefied material ranged between 0.39-0.41% and 0.02-26% respectively, while the H/C and O/C ratios also reduced as the severity intensified which resulted to lower water-vapor and smoke production during burning. Energy loss is also reduced during combustion because of the reduction in the H/C and O/C ratio in torrefied material after the pretreatment. Previous study findings were also in accordance with the results of this study (Chen et al. 2015b; Ren et al. 2017).

From the result in Table 5.3, the hemicellulose content underwent major degradation as the temperature and torrefaction time increased, while cellulose also underwent decomposition like hemicellulose, but to a lesser extent. The thermal decomposition temperature of lignin made it difficult to decompose during the torrefaction process. The lignin content is seemingly added (insoluble fraction), which may be due to the build-up among phenolic compounds and carbohydrates of torrefied material. Wang et al. (2020) also had similar observations, showing that the real quantity of carbon-rich lignin in torrefied material samples increased when the organic constituent (hemicellulose and cellulose) in lignocellulosic biomass breaks down. Generally, hemicellulose, when compared to cellulose, emits mild volatiles, generating less tar and char (Tumuluru et al. 2011), while depolymerization of hemicellulose releases water and acid which can lead to more cellulose degradation.

The elemental change that occurs in biomass during torrefaction pretreatment was demonstrated by the Van Krevelen diagram. The decrease in atomic ration between H/C and O/C during torrefaction led to less energy consumed and an increase in HHV as the severity intensified. Figure S5.1 illustrates the Van Krevelen diagram, the atomic H/C and the atomic O/C proportions showed a nearly linear relationship. The HHV of the untreated sample was recorded

as 16.84 MJ/ kg, a moderate improvement in the HHV of the torrefied material was observed as the SF of the process intensified, which is due to the dehydration and decarbonization reactions observed in the course of the torrefaction process (Bridgeman et al. 2008). Basically, biomass loses more oxygen and hydrogen than carbon through the pretreatment operation, which improves the HHV of torrefied material. In this study, the range of the HHV of the torrefied material was approximately between 17-24 MJ/ kg as the severity increased. Chen et al. (2015b) similarly reported that the HHV of the torrefied sample improved with increased SF. Sarker et al. (2020) recorded HHV of between 22- 25 MJ/ kg when selected agricultural biomasses were torrefied.

Table 5.3. HHV, elemental, and chemical composition of the torrefied material obtained from the microwave-assisted torrefaction of oat straw.

| SF | Hemi-cellulose (%) | Cellulose (%) | Lignin (%) | Ash (%) | Soluble (%) | HHV (MJ/kg) | C (%) | H (%) | N (%) | S (%) | O * |
|------|--------------------|---------------|------------|---------|-------------|-------------|-------------|------------|------------|------------|-------------|
| NT | 24.70 | 47.45 | 12.92 | 5.32 | 9.61 | 16.84 | 44.24(0.62) | 5.85(0.20) | 0.41(0.08) | 0.11(0.02) | 49.39(0.74) |
| 3.64 | 22.84 | 46.34 | 13.93 | 5.88 | 11.01 | 17.22 | 44.80(0.50) | 5.74(0.00) | 0.39(0.01) | 0.26(0.00) | 48.81(0.42) |
| 3.79 | 21.21 | 44.53 | 15.03 | 6.24 | 12.99 | 17.96 | 45.45(0.00) | 5.55(0.02) | 0.39(0.02) | 0.17(0.01) | 48.44(0.02) |
| 3.91 | 18.23 | 42.54 | 22.12 | 6.50 | 10.61 | 18.04 | 47.17(0.36) | 5.47(0.01) | 0.40(0.04) | 0.02(0.00) | 46.94(0.35) |
| 5.11 | 17.70 | 41.45 | 23.92 | 6.83 | 10.10 | 19.35 | 47.24(0.02) | 5.45(0.00) | 0.41(0.01) | 0.11(0.01) | 46.79(0.01) |
| 5.26 | 15.84 | 41.34 | 28.93 | 7.90 | 5.99 | 21.00 | 48.20(0.05) | 5.42(0.02) | 0.39(0.07) | 0.26(0.01) | 45.73(0.07) |
| 5.37 | 13.65 | 38.42 | 32.63 | 8.80 | 6.50 | 21.87 | 49.87(0.41) | 5.38(0.03) | 0.39(0.02) | 0.15(0.00) | 44.21(0.36) |
| 6.59 | 10.02 | 37.04 | 37.17 | 9.21 | 6.56 | 22.02 | 50.65(0.11) | 5.34(0.02) | 0.41(0.01) | 0.13(0.02) | 43.47(0.08) |
| 6.73 | 6.02 | 35.01 | 40.16 | 9.73 | 9.08 | 22.98 | 52.06(0.10) | 5.29(0.11) | 0.41(0.00) | 0.12(0.02) | 42.12(0.06) |
| 6.84 | 4.02 | 34.04 | 42.12 | 10.22 | 9.60 | 24.23 | 52.65(0.21) | 5.24(0.02) | 0.41(0.02) | 0.11(0.00) | 41.59(0.12) |

SF: severity factor; HHV: Higher heating value; C: carbon; H: hydrogen; N: nitrogen; S: sulfur; O: oxygen; and NT: non-treated; * = values obtained by difference; values in parentheses represent standard deviation from triplicate replication of experiment.

5.4.2.2 Proximate analysis

The proximate analysis result for the torrefied material is presented in Figure 5.2. The presence of too much moisture in the raw biomass reduced the pretreatment performance and increased fuel production costs (Evergreen Renewables 2022). The initial quantity of moisture recorded for oat straw sample in this study was approximately 10.6% which reduced drastically to about 1.02% after severe torrefaction pretreatment. Basically, as the torrefaction severity increased, the moisture content decreased, thereby reducing the risk of biological deterioration and storage, and higher transportation cost (Tumuluru et al. 2011) The quantity of ash present in the torrefied material increased with an increase in SF because of the reduction of organic matter observed during the pretreatment process (Wang et al. 2018). Characterization of biomass solid fuels shows that VM and FC are the two significant measures. Fuels with higher VM normally have a lower FC which results in a lower calorific value and higher reactivity. Subsequently, when the amount of FC in the solid fuel is very large usually the value of VM is very low leading to an improvement in the energy value. The VM linearly reduced from around 81 to 56 wt%, while the FC increased from approximately 5 to 32 wt% as the severity of the pretreatment increased. Similar trends were reported by other researchers (Alizadeh et al. 2022, Sarker et al. 2020; Bridgeman et al. 2008; Wannapeera et al. 2011). The VM drastically reduced because of partial carbonization and dehydration during the torrefaction operation. Thus, incomplete depolymerization of cellulose and the disintegration of hemicellulose increased the proportion of lignin available as the pretreatment process continued (Zhang et al. 2016) which brought about an increase in FC.

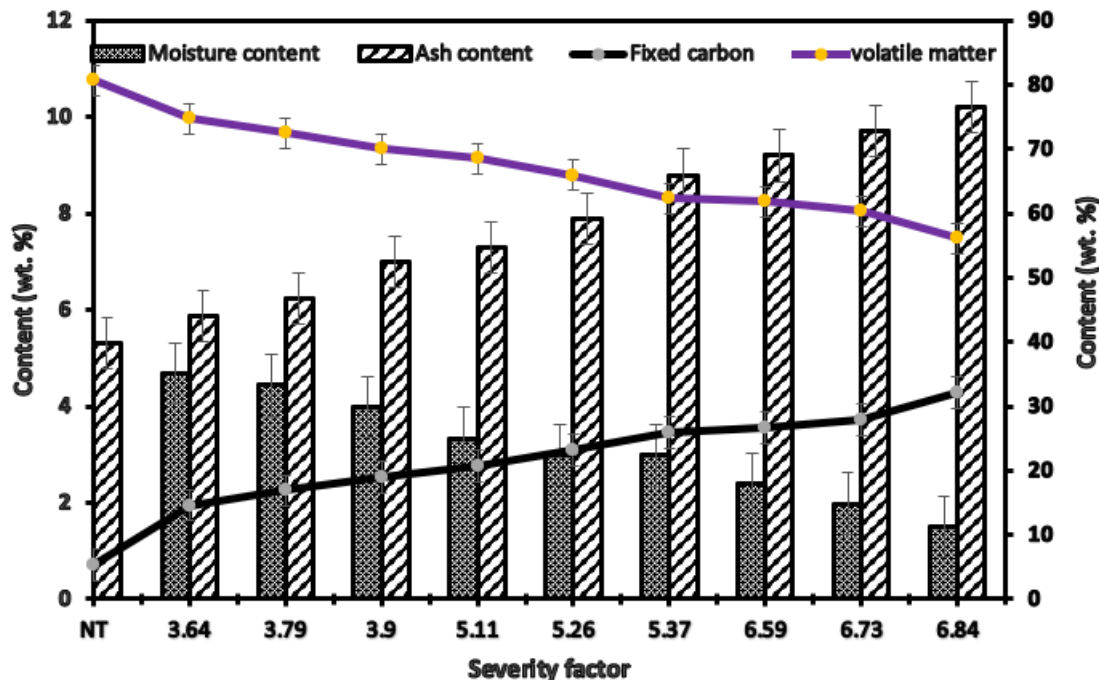


Figure 5.2. Proximate analysis of torrefied material from microwave-assisted torrefaction of oat straw.

5.4.2.3 Torrefaction liquid analysis

The evaluation of the condensable fraction through GC-MS showed that water and acetic acid are the major components of torrefaction liquid (TL) from oat straw torrefaction. Also, a small amount of furfural, formic acid, methanol, hydroxyl acetone, lactic acid, and evidence of phenol were observed. The quantity formed improved with SF. Thus, the products found were mostly from the hemicellulose decomposition during torrefaction. Figure 5.3 represents the condensable yields for oat straw torrefaction pretreatment. A color change from light brown to dark brown, and then black was observed which corresponded to torrefaction severity from light to severe condition respectively. Similar observations were reported previously (Alizadeh et al. 2022; Rudolfsson et al. 2017).

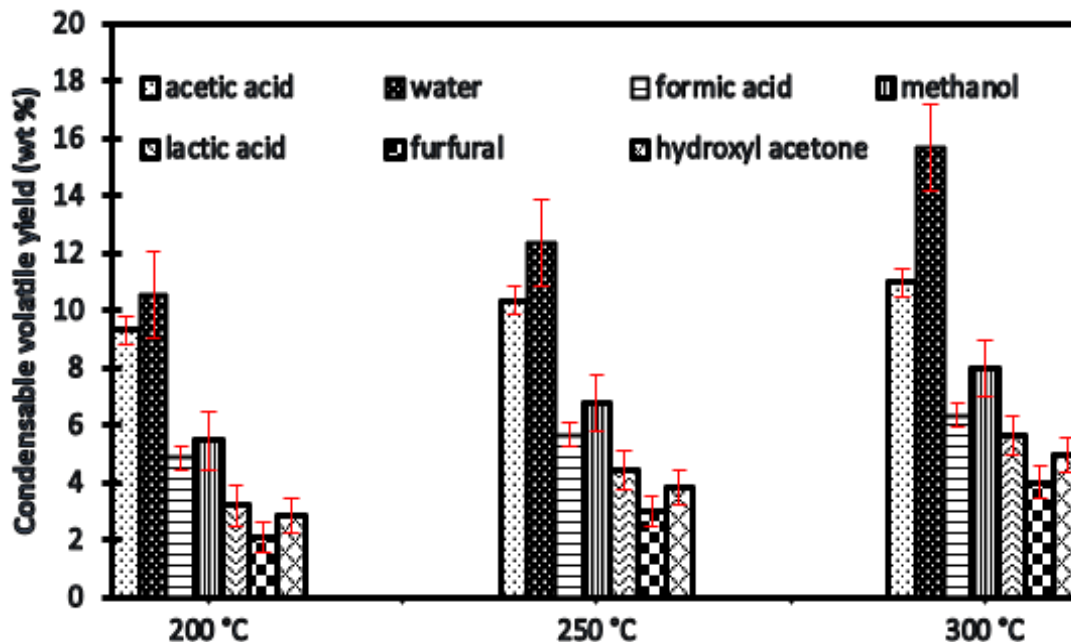


Figure 5.3. Composition of torrefaction liquid from the microwave-assisted torrefaction of oat straw.

5.4.2.4 Particle size, bulk, and particle density of non-treated and torrefied oat straw

The particle size of biomass is one of the essential specifications when considering pelletization, as it helps in the understanding of its flowability and heating value. The pretreated torrefied material particles were notably less than that of the non-torrefied (raw) biomass. This could be because of the pretreatment operation which tended to disintegrate the lignocellulosic arrangement of the torrefied material sample (Iroba et al. 2017), which led to the reduction in shear strength and easy grindability. A narrower particle size distribution (Figure S5.2 in supplementary material) was observed in the torrefied material when set side by side with the untreated ground oat straw due to its brittle characteristics, making it suitable for densification (Onyenwoke et al. 2022). A more durable and denser pellet is formed during densification, when smaller particles reshuffle and fill in the gap between the bigger (coarse) particles (Onyenwoke et al. 2023). Increasing severity condition of the pretreatment process resulted in an increase in

the proportion of fine particles in the sample. This was further confirmed when a Shapiro–Wilk test ($p > 0.05$) was conducted, since the particle size of the samples treated under different severity conditions was approximately normally distributed. Phanphanich and Mani (2011) reported similar trends in the particle size distribution curves with intensity in SF when they studied the pretreatment operation of logging residues and pine chips. Table 4 shows the particle size, bulk, and particle density of non-treated and torrefied oat straw. A geometric average particle size of 0.66 mm was recorded for untreated oat straw. The particle size of the torrefied material reduced with increased SF. After torrefaction, the mean particle size of torrefied material ranged between 0.45 mm and 0.36 mm, while the geometric standard deviation (S_{gw}) ranged between 0.34 mm and 0.24 mm. Onyenwoke et al. (2023) reported similar results after the pretreatment of white spruce sawdust. Bulk density is an essential parameter that provides information about the space required during transportation, handling, and storage of biomass. The bulk density of the samples reduced after torrefaction operation, possibly due to the breakdown of the biomass structure during this process. Also, the decrease in mass (volatiles and moistures) led to a decrease in bulk density. Similarly, Onyenwoke et al. (2023) reported a reduction in bulk density when white spruce sawdust was torrefied at a temperature range of 200-300 °C, and the disintegration of hemicellulose, cellulose, and lignin was the major cause of this reduction. In this study, the particle density of untreated biomass sample was less than the torrefied samples. The increase in particle density after torrefaction could be because of the inter- and intra-particle void created during the torrefaction process (Tumuluru et al. 2011). Also, the decomposition of the oat straw into smaller components and restructuring of its lignocellulosic structure during torrefaction led to an increase in particle density. The bulk density and particle density of torrefied oat straw as shown in Table 5.4 ranged between 117.25-80.23 kg/m³ and

1020.66-1374.87 kg/m³, respectively. Generally, the size and shape of particles can explain the amount of surface area obtainable for integrating particles during pelletization, which affects its mechanical strength (Adapa et al. 2010).

Table 5.4. Particle size, bulk, and particle density of non-treated and torrefied oat straw.

| Severity factor | d_{gw} (mm) | S_{gw} (mm) | Bulk density (kg/m³) | Particle density (kg/m³) |
|------------------------|----------------------------|----------------------------|--|--|
| Untreated | 0.66 | 0.41 | 127.87 | 882.23 |
| 3.64 | 0.45 | 0.34 | 117.25 | 1020.66 |
| 3.79 | 0.41 | 0.32 | 100.32 | 1057.79 |
| 3.91 | 0.40 | 0.32 | 98.56 | 1143.50 |
| 5.11 | 0.40 | 0.31 | 95.67 | 1188.18 |
| 5.26 | 0.39 | 0.29 | 90.54 | 1227.36 |
| 5.37 | 0.36 | 0.28 | 87.27 | 1265.39 |
| 6.59 | 0.38 | 0.27 | 85.44 | 1306.59 |
| 6.73 | 0.35 | 0.26 | 83.32 | 1344.73 |
| 6.84 | 0.32 | 0.24 | 80.23 | 1374.87 |

where d_{gw} represents the geometric mean diameter; S_{gw} is the geometric standard deviation.

5.4.3 Pelletization of samples

The principal aim of pelleting torrefied oat straw is to increase its bulk density which enhances storage capacities and reduces transportation costs. It also facilitates easy feeding during combustion, reduces the formation of dust, and makes the handling process easier. During the densification of biomass, lignin present in the biomass is the main binding source since the pretreatment process exposes many lignin active spots by disintegrating the hemicellulose polymer which resulted in fatty unsaturated structure formation and acts to provide better binding (Tumuluru et al. 2011). Due to the brittle nature of the torrefied oat straw obtained from

the torrefaction process because of the dehydration that occurs during the operation, additional binders were introduced in this study. Torrefaction liquid (TL) and sawdust were used at various proportions as binders for this work, where the TL used is a product of the torrefaction process. Figure 5.4 shows pellets formed by using TL and sawdust at different proportions as a binding agent. After pelletization of the torrefied material, it was observed that pellets with additives appeared to be stronger and no physical breakage or cracks were observed.

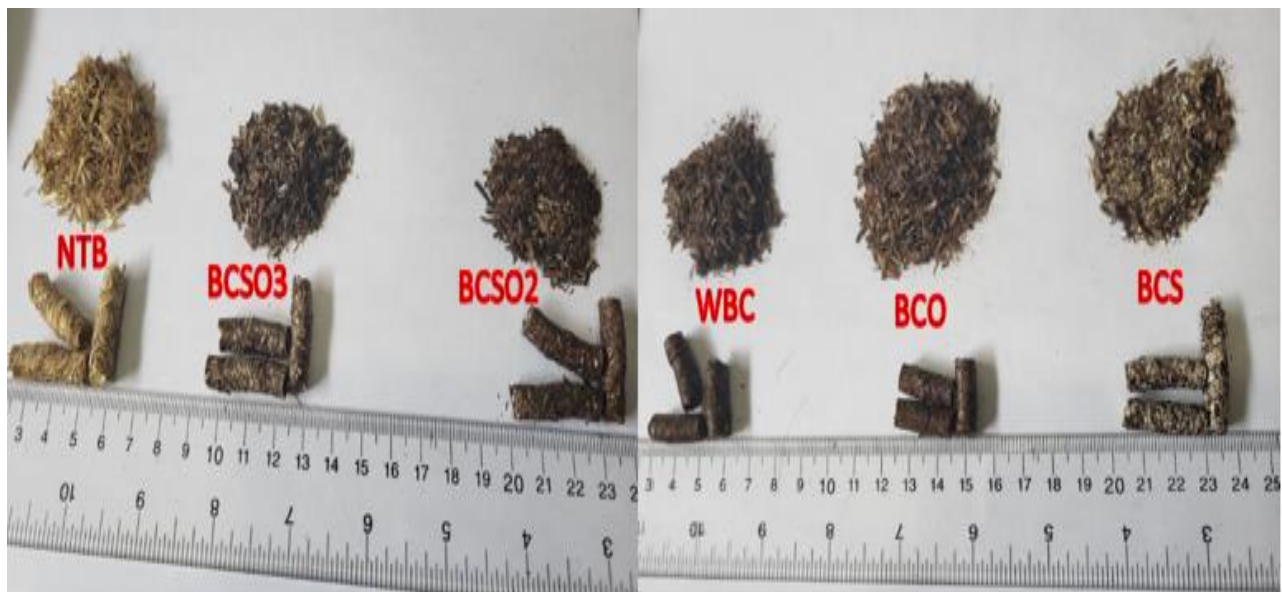


Figure 5.4. Pellets formed from oat straw torrefied material with and without binders.

NTB: Untreated material without binder; **BCSO3:** 33.3 % each of torrefied material, sawdust, and TL; **BCSO2:** 75% torrefied material with 12.5% each of sawdust and TL as binders; **WBC:** torrefied material without binder; **BCO:** 75% torrefied material with 25% TL as the binder; **BCS:** 75% torrefied material with 25% of sawdust as the binder.

5.4.4 Effect of binders on the pellet formed

5.4.4.1 Density of pellet, dimensional stability, and tensile strength of pellet

The effect of binder on the physiochemical, mechanical, and thermal properties of the oat straw pellets formed is depicted in Figure 5.5. The addition of binders in the formulation relatively enhanced the mechanical, thermal, and physiochemical properties of the oat straw pellet formed. The essence of pelletization is to improve the bulk density of the biomass. As stated earlier, an increase in torrefaction severity led to a increase in bulk density which led to an increase in particle density. The pellet density of oat straw improved with the addition of TL in the formulation, which can be because of the binding influence of TL during pelletization. The pellet density of the untreated oat straw pelletized without binder (NTB) was 1031.74 kg/m^3 which improved to 1189.64 kg/m^3 after torrefied and pelletized without binder (WBC). The pellet density of torrefied oat straw pelletized with binder ranged between $1203.76\text{-}1394 \text{ kg/m}^3$. Sarker et al. (2021) reported that the pellet density of most of their samples increased when bio-oil was introduced as a binder during pelletization which was as a result of the binding effect of the bio-oil during densification. The effect of binder on the dimensional stability of the formed pellet is also demonstrated in Figure 5.5. This parameter relates to the stability of pellets during transportation, handling, and storage. This study indicates that torrefaction and pelletization with or without binder improved the dimensional stability of the pellet produced. Lower dimensional stability values are associated with increased pellet stability. The dimensional stability ranged between $8.02\text{-}1.13\%$. Sarker et al. (2021) observed that when bio-oil was added to pellet during pelletization it improves the binding mechanism thereby improves the dimensional stability of pellet. Similarly, the findings of Onyenwoke et al. (2023) agree with this study. The pellet tensile strength was also positively enhanced by the addition of binder, which could be because of the

binding properties of the additives. Pellet density reduced after torrefaction, and this was because of the removal of moisture, volatile matter, and other extractives from the oat straw biomass. The quantity of lignin available in the torrefied material sample determines the pelletability of the torrefied material, whereas sawdust in its raw form has a high amount of lignin. Basically, a higher quantity of lignin saves the cost of operation of the process and enhances binding. It also improves energy density, mechanical strength, and calorific value. The tensile strength of untreated oat straw pellet is 0.35 MPa, this value improved triple fold after torrefaction and introducing binder during pelletization. According to Sarker et al. (2021), the increased pellet tensile strength was due to the presence of fatty acid and protein in the additive which improves the binding system during extrusion.

5.4.4.2 Moisture uptake test

The introduction of TL, as well as sawdust to the torrefied oat straw pellet formulation, ameliorated the surface structure by enhancing the adhesive mechanism of the torrefied material particles. Thus, this lessened the microporous surface tension which decreased the uptake rate of the available water (Onyenwoke et al. 2022; Sarker et al. 2021; Kambo and Dutta 2014). A moisture absorption test is necessary since it authenticates the biodegradable nature of pellets during handling and storage. As stated earlier, the addition of torrefaction liquid is very important in the moisture uptake rate of the formed pellet. The water-repellent rate of pellets formed with the addition of TL and sawdust increased by 5-12%, and 4.5-11%, respectively. In general, the absorption of moisture by torrefied material is because of the presence of OH groups which were broken down during the pretreatment operation resulting in the formation of a hydrophobic product (Onyenwoke et al. 2022; Onyenwoke et al. 2023; Tumuluru et al. 2011;

Lam et al. 2012; Li et al. 2011). An increase in the hydrophobicity of pellets by approximately 11 wt % was observed by Sarker et al. (2021), when bio-oil was introduced, this was associated with the hydrophobic functional groups present in the bio-oil.

5.4.4.3 Higher heating value (HHV) and ash content

The HHV of the pellets formed from different binder additions showed that 75% torrefied oat straw with 12.5% each of sawdust and TL (BCSO2) and 75% torrefied oat straw with 25% TL (BCO) had the highest value of approximately 24 MJ/kg followed closely by BCSO3 and BCS with approximately 23 MJ/kg. Torrefied oat straw pellet without a binder (WBC) had a value of approximately 22 MJ/kg, while the lowest value was observed in the untreated pellet (NTB) with about 17 MJ/kg. Overall, torrefaction pretreatment enhanced the calorific value of pellets when compared to NTB. From the results attained in this study, it can be deduced that the addition of TL in the pellet formulation significantly improved the energy value of the pellet. This result agrees with other studies (Bai et al. 2018; Sarker et al. 2021; Kanwal et al. 2019) using bio-oil as a binder in pellet processing. An increasing pattern was observed in the ash content after torrefaction; however, the result showed that after pelletization with additives the increase in ash content was not significant. The increase in the quantity of ash present after torrefaction was attributed to the mass loss during the process (Onyenwoke et al. 2022; Onyenwoke et al. 2023; Alizadeh et al. 2022; Sarker et al. 2020).

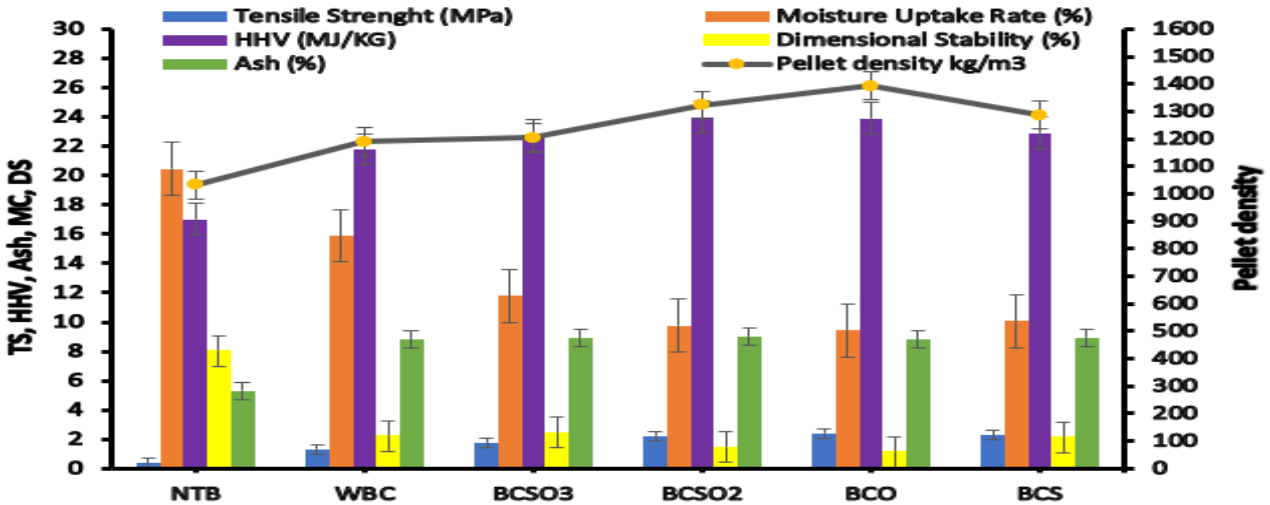


Figure 5.5. Physiochemical properties of pellets from torrefied oat straw with or without binders; TS = Tensile strength; HHV = Higher heating value; MC = moisture uptake rate; DS = Dimensional stability; NTB = Untreated oat straw pellet without binder; WBC = torrefied oat straw pellet without binder; BCSO3 = 33.3 % each of torrefied oat straw, sawdust, and TL; BCSO2 = 75% torrefied oat straw with 12.5% each of sawdust and TL as binders; BCO = 75% torrefied oat straw with 25% TL as the binder; BCS = 75% torrefied oat straw with 25% of sawdust as the binder.

5.4.4.4 Microstructural analysis

The binding mechanism of torrefied material particles when pelleted was further analyzed with the help of scanning electron microscopy (SEM), which aids in the observation of the internal stratification of the pellets formed. The microscale effect of the pretreatment process on the oat straw and the impact of the binder proportions can be observed directly by microstructural examination, which provides a visualization of the structural pattern of the torrefied material pellet samples. Tilay et al. (2015) have previously used SEM for the internal structure examination of pellets. Figure 5.6 shows the SEM micrographs of the torrefied material pellets from different binder additions. It was observed that pellets with TL added as binder showed more firmly glued surface with fewer pore spaces when compared to other pellets. Also, pellets formed with sawdust as a binding agent showed less porosity when compared with torrefied material pellet without binder. The addition of binders improved the binding mechanism during

densification of torrefied material particles, which accounted for the improved tensile strength, HHV, reduced porosity, and low water absorption rate of pellets.

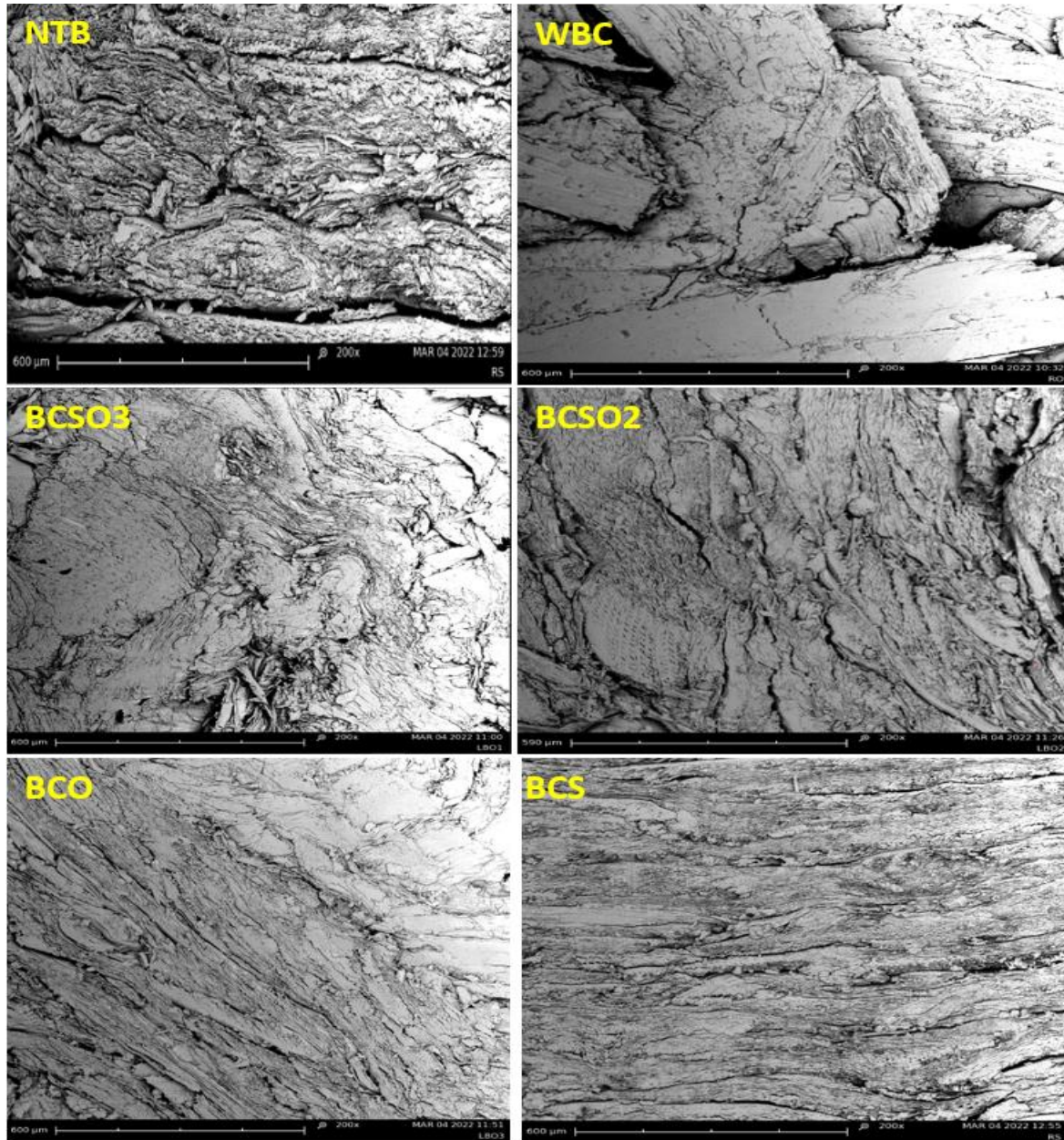


Figure 5.6. SEM micrographs images of oat straw torrefied material pellets.

NTB = Untreated oat straw pellet without binder; WBC = torrefied oat straw pellet without binder; BCSO3 = 33.3 % each of torrefied oat straw, sawdust, and TL; BCSO2 = 75% torrefied oat straw with 12.5% each of sawdust and TL as binders; BCO = 75% torrefied oat straw with 25% TL as the binder; BCS = 75% torrefied oat straw with 25% of sawdust as the binder.

5.5 Conclusions

The combined effects of torrefaction and binders on the quality of pellets produced from oat straw was studied. The results of this study indicate the potential for torrefaction pretreatment to enhance the mechanical, physiochemical, and thermal properties of torrefied oat straw pellets. In addition, there were noticeable improvements in the physiochemical properties of the pellets formed after torrefaction, except for the tensile strength. The introduction of binders improved the binding mechanism among torrefied material particles during pelletization, which resulted in improved pellet tensile strength and further enhanced other pellet properties. Oat straw can be torrefied and pelletized using torrefaction liquid as a binder to make pellets for biofuel production.

Chapter 6

Technoeconomic Analysis of Torrefaction and Steam Explosion Pretreatment Prior to Pelletization of Selected Biomass

- A similar version of this chapter has been published in journal *Energies*: Onyenwoke, C., L.G. Tabil, T. Dumonceaux, D. Cree, E. Mupondwa, X. Li and O. Onu. 2024. Technoeconomic analysis of torrefaction and steam explosion pretreatment prior to pelletization of selected biomass. *Energies* 17(1): 133. <https://doi.org/10.3390/en17010133>.

Contribution of the Ph.D. Candidate

The experimental work, data collection and analysis, original drafting, review and editing of the manuscript was done by the Ph.D. candidate, while Drs. L.G. Tabil, T. Dumonceaux, E. Mupondwa, D. Cree, X. Li, and O. Onu, provided resources and editorial input. Drs. L.G. Tabil and T. Dumonceaux performed the supervisory roles, while Drs. L.G. Tabil, T. Dumonceaux E. Mupondwa and D. Cree also carried out the project administrative roles.

Contribution of the Paper to the Overall Study

Technoeconomic analysis is important for evaluating the economic feasibility of the pathways to commercial production of bioenergy. The economic parameters such as total revenue produced by the plant and payback time are the key metrics that can attract investors to the industry. Information on the large-scale application of steam explosion and torrefaction pretreatments – based pellet production is provided in this chapter. The results from this study can be assessed and implemented in future projects.

6.1 Abstract

Lignocellulosic biomass has demonstrated great potential as feedstock for pellet production, notwithstanding the fact that the industrial production of pellets is faced with some economic challenges. This study presents a technoeconomic analysis of six scenarios to develop a process model for pellet production from sawdust and oat straw that employs torrefaction and steam explosion pretreatment prior to pelletization. SuperPro Designer was used to carry out this evaluation. The pellet plants were designed to have a capacity of 9.09 t/h of sawdust and oat straw each. The pellet yield ranged from 59 kt– 72 kt/year. Scenarios analyzed included variations of steam explosion and torrefaction. In some scenarios materials were lost in the form of liquid and gas due to the pretreatment process. The breakdown of equipment purchase cost showed that the torrefaction reactor is the most expensive unit with about 51% of the purchase cost. Facility-dependent and feedstock costs were the major significant contributors to the pellet production cost. The minimum selling price of pellets obtained from Scenarios 1-6 were \$113.4/t, \$118.7/t, \$283.4/ t \$298.7/t, \$200.5/t, and \$208.4/t, respectively. The profitability of pellet production as determined by the net present value (NPV), internal rate of return (IRR), and payback period and was found to be sensitive to variations in feedstock cost.

6.2 Introduction

In recent decades, the agriculture and forestry industries have encountered many changes because of globalization, financial crisis, high cost of energy, and climate change. Canada, Finland, United States, Sweden, Norway, and some countries in South America have huge, forested areas which can be harvested and used for industrial purposes (Clauser et al. 2018; Lamberg et al. 2012). Co-firing coal with biomass has been shown to be a promising, less

expensive, and low greenhouse gas (GHG) emission alternative for electricity production (Sami et al. 2001). A great number of power generation plants have been established in the United Kingdom, and Europe with co-firing options ranging from 10 to 100%, mainly because of the strict regulations to minimize GHG emissions (Roni et al. 2017). Wood pellets are used as the solid biofuel in power plants in the U.K. and Europe. The use of biomass for power generation in the United States is low (about 2%), nevertheless, this is expected to increase with the establishment of environmental regulations, coal compatible biofuel sources, availability, and low cost of biomass, and lastly, reduced transportation costs (Goerndt et al. 2013).

The United States had an annual wood pellet production approximately 6.6 Mt in 2020 (EIA 2021), making it the world's largest producer of wood pellets utilized for heat and power generation. Canada had an estimated wood pellet production of 3.5 Mt in 2021, with domestic use estimated at 0.5 Mt (USDA 2023). However, there are setbacks in the utilization of wood pellets domestically for power generation due to differences in the higher heating value and dimensional stability of coal versus wood pellets. Pretreatments by torrefaction prior to pelletization have been broadly investigated to improve thermal and physiochemical properties of lignocellulosic biomass as a suitable substitute to coal (Agar 2017; Nanou et al. 2016; Koppejan et al. 2012; Chen et al. 2015b; Cao et al. 2015). Torrefaction is a thermal treatment process of feedstock, which involves the roasting of the feedstock in an inert surrounding at temperatures ranged between 200–300 °C. Torrefaction is sometimes referred to as mild pyrolysis where the operating temperature is less than the pyrolysis process. This pretreatment method reduces the amount of moisture in the biomass sample, which makes the biomass hydrophobic and enhances the calorific value, thereby improving the volumetric energy density of biomass. Torrefaction process produces both the solid char (torrefied material) and the torgas.

The torgas comprises condensable and non-condensable gases. Condensable gas is a torrefaction liquid (TL) which is rich in ketones, organic acids, furfural, water, and trace of other components. These TL components could be transformed into economical feasible products (Phanphanich and Mani 2011). Since the bulk density of torrefied material is reduced because of mass loss and voids formed during torrefaction, pelletization increases the total energy density of the final products. There are a lot of challenges that have limited the development of biomass-based facilities. The characteristics of the biomass from different sources vary significantly, and this obviously is one of the main factors limiting their commercialization in a biomass-based facility. Traditionally, biomass has high moisture content, low energy density, calorific value, and yield. These factors increase the transportation cost of biomass, which also increases biomass conversion costs (Kumar et al. 2003). Pretreatment and pelletization of biomass could reduce the overall cost of biomass conversion.

Steam explosion is a physical and chemical process, otherwise known as physiochemical pretreatment method, that involves the breakdown of lignocellulosic biomass by two procedures: first by the utilization of high-pressure heat (3.4 MPa at 180 °C), which led to the formation of organic acid, and second by shearing forces which prompt moisture expansion and explosive decompression. These processes alter the biomass components via hydrolysis of hemicellulosic components (leading to the release of mono- and oligosaccharides), it modifies the chemical structure of lignin, and enhances the crystallinity index of cellulose. These transformations allow the lignocellulosic biomass structures to unleash and improve the fermentable carbohydrate yield of the next enzymatic hydrolysis steps (Lam 2011; Tooyserkani 2013; Pastre 2002).

Biomass production cost, feedstock transportation cost, and densification cost contribute significantly to the high economic component of downstream production. Therefore, cost

implications of biomass production, transportation, and pretreatment are limitations to the commercialization of pellet production (Shahrukh et al. 2016; Manouchehrinejad et al. 2021).

Biomass pelletization can be defined as a compression or compaction process to eliminate inter- or intraparticle empty spaces. It also reduces the moisture content of biomass during compression. The aim of biomass pelletization is to increase mass per unit volume of the biomass. Pelletization is a vital approach for the biomass market since it can improve the convenience and accessibility of biomass because of the uniform shape and size (Gebreegziabher et al. 2013; Luk et al. 2013). Pelletization also facilitates logistics by improving its storage and handling characteristics and reducing transportation cost (Mupondwa et al. 2012).

Previous research has analyzed the economics of biomass-based energy from the outlook of generic models (Shahrukh et al. 2016; Dassanayake and Kumar 2012; Kumar et al. 2008). The cost of sawdust pellet production has been studied by Mani et al. (2006) who reported that pellets can be produced from sawdust at US\$51/t based on 45 kt plant capacity. Similarly, Shahrukh et al. (2016) performed a technoeconomic assessment of pellets produced from steam pretreatment of three biomass feedstocks (energy crops, forest residue, and agricultural residue). They reported that the cost of production varied from US\$95 – \$105/t for regular pellets and US\$146 – \$156/t for steam pretreated pellets at a capacity of 190 kt and 250 kt, respectively. Pirraglia et al. (2013) carried out a technoeconomic analysis of an industrial scale 100 kt/year wood biomass torrefaction system in United States, reporting a production cost of \$199/t, and concluding that a high capital cost represented the most sensitive factor affecting the net present value (NPV), followed by biomass cost. The torrefaction liquid (TL) which consists of mainly water and other minor components was assumed to sell at US\$ 0.25/L (Badger et al. 2011).

However, there has not been much research to evaluate the production costs of torrefaction and steam explosion pretreated pellets, and how they compare with the production costs of regular pellets (untreated). There is a need to evaluate the economic feasibility of both forms of pretreated biomass-based pellets. There is a need to link research and development (R&D), engineering, and business. By connecting process parameters to economic metrics, it serves as a useful method to screen potential research priorities, it can help understand businesses better and the factors that affect the profitability of their technology development projects.

Therefore, the overall objective of this study is to conduct a comparative technoeconomic assessment of an integrated steam explosion, torrefaction, and pelletization process from two feedstocks sawdust and oat straw relative to the cost of regular pellet production. This can establish cost bottlenecks at the earliest phase of project and present the mass and energy data required to conduct life-cycle environmental assessments.

6.3 Methodology

6.3.1 Process design and parameters

Technoeconomic analysis for sawdust -and oat straw -based pellet production was carried out using SuperPro Designer software v.10 (Intelligen Inc., Scotch Plains, NJ, USA). A plant capacity of 9.09 dry t/h of sawdust and oat straw was selected for the design, equivalent to 72 kt/year, assuming 7,920 h of annual operating time for both scenarios.

The plant size of 72 kt/year was selected based on available sawdust in the NorSask company Meadow Lake (Government of Saskatchewan 2021) and available oat straw in Meadow Lake Saskatchewan Canada with latitude of 54° 7' 27.00" N (52.755529) and longitude of 108° 26' 9.00" W (-107.316492). The unit operations in this study are grouped into three process sections: feeds

tock preprocessing, steam explosion/torrefaction pretreatment, and pelletization. The assumptions, data and process conditions used in the process design of this study were obtained from experimental results reported from our previous research (Onyenwoke et al. 2022; Alizadeh et al. 2023; Onyenwoke et al. 2023a; Onyenwoke et al. 2023b) and Superpro built-in data (Table 6.1).

Table 6.1. Process conditions and data used in process baseline modeling.

| Unit Operation | Parameter | Value | Reference |
|------------------------|-----------------------------|--------------|---|
| Steam explosion | Temperature (°C) | 180 | Onyenwoke et al. 2022, Alizadeh et al. 2023 |
| | Residence time (min) | 9 | |
| Torrefaction | Temperature (°C) | 250 | Onyenwoke et al. 2023, Alizadeh et al. 2022, Valdez et al. 2021 |
| | Residence time (min) | 9 | |
| Binder | Torrefaction liquid (%) | 25 | Onyenwoke et al. 2023b |
| Pelletization | Die temperature (°C) | 95 | Onyenwoke et al. 2023a |
| | Sample moisture content (%) | 10 | |

6.3.2 Process description

6.3.2.1 Feedstock processing and plant location

Sawdust and oat straw were the two feedstocks used in this study. The first step in analyzing the economic cost of producing pellets from oat straw is determination of feedstock cost. In establishing the cost of non - woody biomass, the following were considered: The cost of production (establishment and harvest), storage, drying, and transportation costs.

Biomass Inventory Mapping and Analysis Tool (BIMAT) developed by Agriculture and Agri-Food Canada (AAFC) (BIMAT 2010) was used to identify high oat producing regions in Meadow Lake, Saskatchewan, Canada. Biomass availability was recorded on a 50% farmer participation basis, the tillage type, and competitive usage of straw for livestock and soil

conservation were considered. The available oat straw within 50 km to the plant is approximately 89,409 tonnes per year. This study assumed that trucks were the main mode of transportation for conveying oat straw from the farm to the densification plant within the region. There are no available short line railroads near farms that would be contracted to supply straw, this is the case in most Canadian Prairie. Therefore, trucks offer the requisite responsiveness and flexibility to transport the product as the market demands (Mupondwa et al. 2012). For baled feedstock, conveying is through a segmented truck or flatbed trailer hinged to an agricultural tractor. Chipped material is moved by either a high sided trailer attached to an agricultural tractor or segmented truck and step frame trailer. Agricultural tractor combination is the preferred means of transportation where the travel distances are short (less than 50 km).

Equation 6.1 defines the average transportation distance (L, km) as a function of feedstock collection radius (r, km) of a circular area which consist of farms supplying oat straw to the pelletization plant whose central point is in the circular area.

$$L = \frac{2}{3} r \quad (6.1)$$

The estimated area (A) according to BIMAT is assumed to have area as shown in Equation 6.2

$$A = \pi r^2 \quad (6.2)$$

Using Equation 1, the average transportation distance (L) is determined as a function of plant capacity using Equation 6.3.

$$L = \frac{2}{3} \sqrt{\frac{A}{\pi}} \quad (6.3)$$

Feedstock transportation cost (C_t) is calculated as described by Stephen (2008) and Sokhansanj and Fenton (2006) as indicated in Equation 6.4

$$C_t = C_f + C_v L \quad (6.4)$$

Where, C_f is the fixed cost of transportation which includes loading and unloading, which is \$3.40/t for a bale (Composites Innovation Centre 2008) and \$3.84/t for pellets Sokhansanj and Fenton (2006); C_v represents the variable cost of transporting feedstock (\$/t km).

Bioenergy plants are usually sited in regions where there is abundant feedstock to reduce the cost of transportation and facilitate feedstock supply logistics. Given Saskatchewan's abundance of oat straw feedstock, it was assumed that, without accounting for tortuosity factor, the transportation calculation is acceptable for theoretical analysis (Mupondwa et al. 2012). In this study the biorefinery was strategically situated in the environs of NorSask Forest Products mill in Meadow Lake Saskatchewan, to allow proximity and regular feedstock availability.

The price of sawdust feedstock was obtained from a study by Antonio et al. (2021). It was assumed that the dry oat straw bale was supplied to the bioenergy plant at a cost estimated using BIMAT (2010). The samples were stored indoors under room conditions until ready to use. According to Samson et al. (2016) indoor storage of samples results in 2% loss in sample dry matter. Oat straw samples were conveyed (belt conveyors) to the hammermill of sieve size 3.2 mm for further size reduction. The oat straw particle size (3.2 mm) option chosen in this study has been reported to favor optimum pretreatment performance (Onyenwoke et al. 2022). While sawdust samples did not require a size reduction, instead they were conveyed directly to the pretreatment chamber. The properties of the feedstocks used in this study are presented in Table S6.1 in the Supplementary Material.

6.3.2.2 Pretreatment process

Figures 6.1 and 6.2 show torrefaction and steam explosion pretreatment process. The steam explosion pretreatment process adopted in this study was similarly carried out by Onyenwoke et al. (2022). This process consisted of a steam boiler, reactor, and a discharge at the bottom of the

vessel. Steam-treatment of samples submitted at 180 °C for 9 min was considered for this analysis due to a lower energy consumption. Screw conveyors were used to introduce the feedstocks into the vessel (10.00 bar pressure). The steam was produced by the high-pressure boiler, and when the required reaction temperature was attained, the ball valve was opened manually to allow the saturated steam to enter the reactor chamber (vessel) to treat the sample for a specified period of time. The steam exploded sample was conveyed to the dryer by belt conveyor.

The torrefaction pretreatment process adopted in this study was similarly carried out by Onyenwoke et al. (2023a). This process consisted of the microwave, reactor, and condenser system. The preferred thermal conditions for this study were 250 °C and a retention time of 9 min based on the optimum condition studied (Onyenwoke et al. 2023a). A water-cooled condenser system was designed to trap the torrefaction gases that exited the microwave reactor. The condensable gas was split into a storage tank and a pelletization section to be used as a binder. The torrefied sample was discharged through a water-cooled screw (Teal et al. 2012), which enabled the torrefied samples to cool below 100 °C to prevent auto ignition (Evangelista et al. 2018).

6.3.2.3 Pelletization process

Pelletizer was used to simulate this process. The die temperature was set at 95 ± 2 °C. Screw conveyor was used to introduce the dried steam treated sample into the extruder, then the pellets were ejected and cooled. During pelletization of torrefied samples, the torrefaction liquid was introduced as a binder. The torrefied samples and the torrefaction liquid were thoroughly mixed in a hopper (P-12/HP-101) prior to their introduction into the extruder. A bucket elevator was used to convey the formed pellets to the storage tank, while the pellet crumbles were

reintroduced to the pelletizer to make better pellets. During pelletization, the energy consumed was assumed to be 85.7 kWh t^{-1} as described by Yun et al. (2020). Prior to storage of the pellets formed, the pellets were allowed to pass via a counter flow cooler to reduce the temperature and avoid ignition during storage (Manouchehrinejad and Mani 2019).

6.3.2.4 Scenario

There are six Scenario established in this study as potential bioenergy enterprises : (1) wood pellet production from raw sawdust in a pellet plant, (2) straw pellet production from raw oat straw in a pellet plant, (3) integrated torrefied treated sawdust pelletization plant utilizing the torrefaction liquid (TL) as the pellet binder, (4) integrated torrefied treated oat straw pelletization plant utilizing the torrefaction liquid (TL) as a pellet binder, (5) integrated steam treated sawdust pellet plant and (6) integrated steam treated oat straw pellet plant. The pellet plant was intentionally sited in close proximity to feedstock sources. Biomass Inventory Mapping and Analysis Tool (BIMAT) developed by AAFC (BIMAT 2010) was used to identify high oat straw producing areas around the proposed pellet biorefinery. Scenarios 1 and 2 produce regular wood pellets and agricultural residue pellets, respectively. Figures S6.1 and S6.2 depict the process of pelleting raw sawdust and oat straw, respectively. Figure 6.1 shows the pellet production process for scenario 3. Figure S6.3 depicts the pellet production process for scenario 4. Nitrogen gas was supplied to the reactor to purge the system and provide an inert environment suitable for the torrefaction process. A heating rate of $50 \text{ }^\circ\text{C min}^{-1}$ was assumed in this case to maximize solid yield (Bergman 2005). Figure S6.4 and Figure 6.2 depict the production process of steam exploded sawdust and oat straw pellets, respectively. The generated steam was injected into the reactor to submerge the feedstock. The feedstock was allowed to remain inside the reactor for a period of 9 min as the conditions increase to 1000 kPa and $180 \text{ }^\circ\text{C}$. The steam exploded sawdust

was conveyed to a rotary drum dryer to remove the excess moisture available in the feedstock. During the drying process there is escape of some volatile compounds. Shahrukh et al. (2015) reported that the drying process utilizes a huge amount of energy during the steam pretreatment process, which considerably increases the energy demand. These scenarios were evaluated based on feedstock, procedures, operations, and their mechanisms followed by cost analysis.

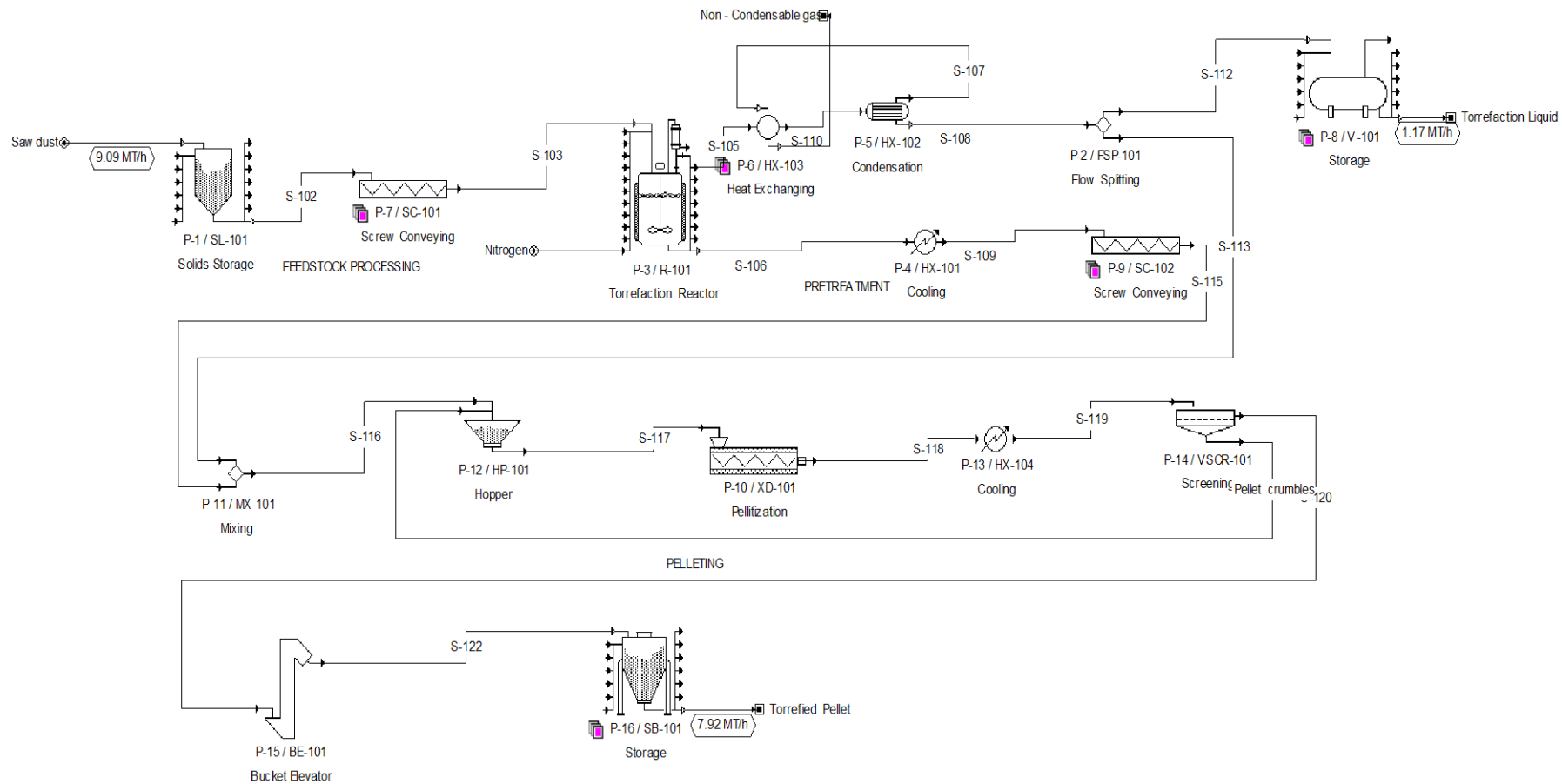


Figure 6.1. Integrated torrefied treated sawdust pelletization plant utilizing torrefaction liquid (TL) as binder (Scenario 3).

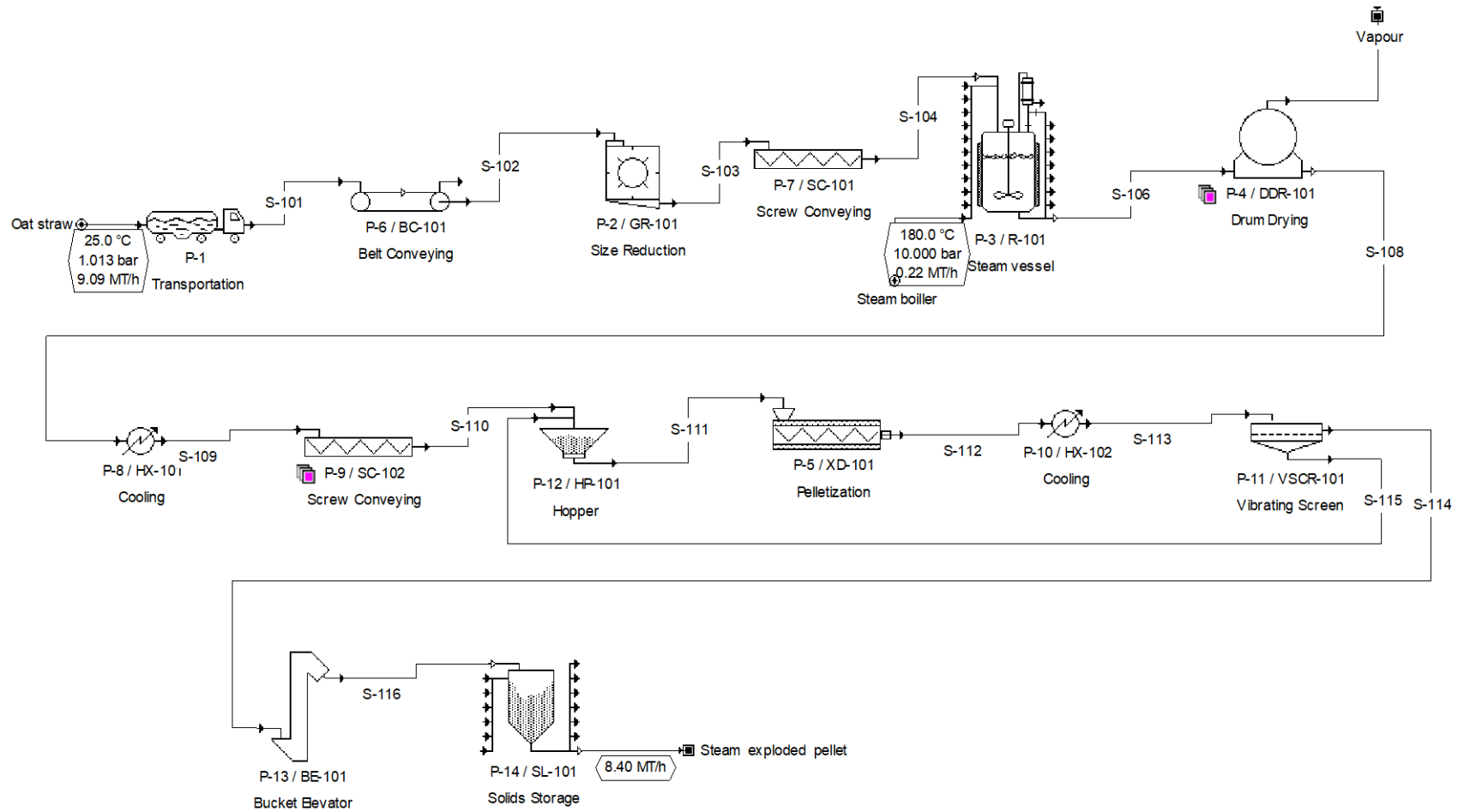


Figure 6.2. Integrated steam exploded treated oat straw pellet plant (Scenario 6).

6.3.3 Cost analysis

The purpose of modeling and simulation of the process was to determine the cost of pellet production and to ascertain the basic economic requirement for prospective research and development. The cost presented in this study was based on 2023 US dollars, Table S6.2 summarizes the cost assumptions and the economic evaluation parameters used in this study. The plant capital investment and operating cost were calculated based on data from other researchers on similar technoeconomic studies (Alizadeh et al. 2023; Valdez 2020; Mupondwa et al. 2017; Vlysidis et al. 2011; Junsittiwate et al. 2022; Fadhilah et al. 2023; Patel et al. 2022) and the built-in cost models in SuperPro designer. Equipment purchase cost (PC) was evaluated on the established equipment sizes acquired from the process model after the materials and energy balance evaluation. Total equipment purchase cost is the summation of listed and unlisted equipment. The summation of direct fixed capital (DFC), start-up and validation cost, and working capital is the total capital investment (TCI). The calculation of the various components of the DFC is shown in Table 6.2.

Startup cost and validation cost is 15% of the DFC (Sarker et al. 2023) while working capital was calculated to be 5% of the DFC (Sarker et al. 2023). Annual plant operating costs used in this study comprise of raw materials, facility dependent, labour dependent, laboratory, and utilities. Facility dependent costs included maintenance (5% DFC), depreciation cost, and miscellaneous cost.

Capital cost and equipment cost for the pellet production capacity were estimated to scale up based on the relationship between capital cost and pellet plant size as represented in Equation 6.5.

$$C_{ne} = C_{ex} \left(\frac{Q_{ne}}{Q_{ex}} \right)^{0.6} \quad (6.5)$$

where C_{ne} denotes estimated capital cost; C_{ex} denotes the empirical capital cost of a baseline pellet plant with size Q_{ex} ; Q_{ne} is the pellet plant size; The exponent 0.60 is selected as scaling factor to depict the economic and financial effect of increasing or reducing pellet plant size (Mupondwa et al. 2012).

6.3.4 Profitability analysis and minimum selling price of pellet

Profitability analysis was carried out to determine the economic viability of the proposed process. Sensitivity analysis provides information on how the alternate form of the economic and process assumed variables influence the economic performance and the sensitive components of the design. The sensitivity analysis of a variable could steer prospective research and development efforts that, in turn, enhance the capacity of the different biorefinery scenarios (Hasanly et al. 2018). The minimal selling price (MSP) of the pellet was estimated using discounted cash flow analysis (DCFA). This study assumed that the plant operates uninterrupted for 24 h a day, 330 days in a year, amounting to 7,920 h per year. The MSP was evaluated using a 10% discount rate, where the net present value (NPV) is equal to zero (selling price at which NPV = 0). Javier and Ortiz (2020) described the profitability index analysis in detail.

The profitability of the pellet biorefinery plant was estimated using the net present value (NPV), which ascertains the enterprise or projects that will yield the greatest return in each period. The NPV was evaluated based on Equation 6.6.

$$NPV = -I_0 + \sum_{j=1}^T \frac{\emptyset_j}{(1 + R)^j} \quad (6.6)$$

where I_0 represents the initial investment in the pellet plant; \emptyset_j denotes annual cash flow; R denotes the discount rate; $j = 1, 2, \dots, T$ is the year with N terminal time.

Table 6.2. Summary of assumed direct fixed capital cost.

| Cost category | Estimation assumption |
|--|--|
| Purchase price of all equipment (PC) | Plant equipment cost + unlisted equipment |
| Installation (I) | 0.50 x PC |
| Piping (II) | 0.35 x PC |
| Instrumentation cost (III) | 0.40 x PC |
| Building and services (IV) | 0.45 x PC |
| Electrical facilities (V) | 0.10 x PC |
| Insulation (VI) | 0.03 x PC |
| Yard improvements (VII) | 0.15 x PC |
| Auxiliary facilities (VIII) | 0.40 x PC |
| Total plant direct cost (TPDC) | PC + I + II + III + IV + V + VI + VII + VIII |
| Engineering (IX) | 0.25 x TPDC |
| Construction (X) | 0.35 x TPDC |
| Total plant indirect cost (TPIC) | IX + X |
| Contractor's fee (XI) | 0.05 x (TPDC + TPIC) |
| Contingency and research & development (XII) | 0.10 x (TPDC + TPIC) |
| Direct fixed capital cost (DFC) | TPDC + TPIC + XI + XII |
| Working capital (WC) | 0.05 x DFC |
| Start-up cost (SC) | 0.15 x DFC |
| Total capital investment (TCI) | DFC + WC + SC |

6.4 Results and Discussion

6.4.1 Capital cost

Tables S6.3, S6.4, and S6.5 outline the purchase cost of major equipment used for each scenario. In Scenarios 1 and 2 (regular pellet process), equipment costs were the most economical when compared to Scenarios 3, 4, 5, and 6 which were the pretreatment process. The pelletizer was the most expensive equipment in Scenario 1 and 2 (US\$212,000) while the bucket elevator (US\$20,000) was the least expensive. The purchase cost of major equipment used for Scenario 1 was approximately US\$1.27 M, while about US\$1.32 M was used to purchase equipment in Scenario 2. Similarly, Alizadeh et al. (2023) estimated the total equipment cost for producing 72 kt/yr of wood pellets at US\$1.7 M based on 2020 US dollars, while Wolbers et al. (2018) estimate for producing 87 kt/yr of wood pellets per year was US\$1.5 M based on 2016 US dollars.

Table S6.4 summarizes the cost of purchasing equipment for producing torrefied pellets (sawdust and oat straw). Scenario 3 (torrefied sawdust pellets) and Scenario 4 (torrefied oat straw pellets) were almost in the same range (US\$7.33 M and US\$7.86 M, respectively) for production capacities of 9.09 t/h of pellets. Similarly, Valdez (2020) reported an estimate of US\$ 6.53 M based on 2023 US dollars for the production capacity of 4.5 t/h of torrefied pellets. In another study, Mobini Dehkordi (2015) estimated that US\$26.08 M based on 2023 US dollars was used to purchase equipment to produce 137 kt/yr of torrefied wood pellets. The breakdown of the equipment purchase cost showed that the torrefaction reactor is the most expensive unit with about 51% of the purchase cost while the least expensive unit was the conveyor. Sarker et al. (2023) reported that torrefaction reactor was the most expensive unit accounting for 32% of the purchase cost of equipment. In a similar manner, Manouchehrinejad et al. (2021) reported the cost share for torrefaction reactor was around 34% of total capital investment (TCI) for torrefaction

before pelleting (TOP) process, while Koppejan et al. (2012) reported that a torrefaction reactor accounts for 45% of the TCI for the TOP. The difference in the cost of the torrefaction reactor may be due to its capacity.

The cost of purchasing equipment for producing steam exploded pellets (sawdust and oat straw) is shown in Table S6.5. Scenario 5 (steam exploded sawdust pellets) and Scenario 6 (steam exploded oat straw pellets) were almost in the same range (US\$2.13 M and US\$2.68 M respectively) for pellet production capacities of 9.09 t/h. According to Alizadeh et al. (2023) the cost of purchasing equipment for producing steam-exploded pellets and electricity was estimated at US\$3.3 M based on 2020 US dollars (\$3.92 M based on 2023 US dollars) with production capacities of 53 kt/yr steam exploded pellets. In a similar manner, Wolbers et al. (2018), reported that the total equipment costs for producing 84 kt/yr steam exploded pellets was estimated at approximately US\$2.3 M based on 2016 US dollars (US\$ 2.94 M based on 2023 rate).

The total capital investment (TCI) for six Scenarios is presented in Table 6.3. TCI for producing regular pellets was approximately US\$9.9 M (Scenario 1 and 2). Alizadeh et al. (2023) reported a TCI of US\$10.5 M based on 2020 US dollars for producing wood pellets from untreated sawdust, with the Cumulative rate of inflation at 18.6% this value stands as \$12.46 M in 2023. Similarly, for a capacity of 80 kt/yr of wood pellet, Peng (2012) recorded a TCI of \$9.1 M on 2012 US dollars (equivalent to \$12.17 M in 2023). Scenarios 3 and 4 have TCI of approximately US\$54.5 M and US\$60.3 M, respectively. The high TCI recorded in Scenarios 3 and 4 was because of the purchase cost of the torrefaction equipment. Peng (2012) recorded a TCI of US\$32.16 M in 2012 US dollars (\$43.10 M in 2023) for torrefied wood pellet production. The TCI for Scenarios 5 and 6 were US\$15.9 M and US\$19.9 M, respectively. Similarly, Alizadeh et al. (2023)

reported a TCI of US\$24.68 M on 2023 US dollars for a plant including combined heat and power (CHP) and steam exploded wood pellets was estimated.

Table 6.3. Total capital investment (TCI) for six Scenarios.

| Cost category | Scenario 1 | Scenario 2 | Scenario 3 | Scenario 4 | Scenario 5 | Scenario 6 |
|---|------------------|------------------|-------------------|-------------------|-------------------|-------------------|
| Purchase price of all equipment (PC) | 1,273,000 | 1,323,000 | 7,335,000 | 7,859,000 | 2,125,000 | 2,675,000 |
| Installation (I) | 636,000 | 662,000 | 3,553,000 | 3,930,000 | 1,063,000 | 1,338,000 |
| Piping (II) | 445,000 | 463,000 | 2,567,000 | 2,751,000 | 744,000 | 936,000 |
| Instrumentation cost (III) | 509,000 | 529,000 | 2,934,000 | 3,144,000 | 850,000 | 1,070,000 |
| Building and services (IV) | 573,000 | 595,000 | 3,301,000 | 3,537,000 | 956,000 | 1,204,000 |
| Electrical facilities (V) | 127,000 | 132,000 | 733,000 | 786,000 | 213,000 | 268,000 |
| Insulation (VI) | 38,000 | 40,000 | 220,000 | 236,000 | 64,000 | 80,000 |
| Yard improvements (VII) | 191,000 | 198,000 | 1,100,000 | 1,179,000 | 319,000 | 401,000 |
| Auxiliary facilities (VIII) | 509,000 | 529,000 | 2,934,000 | 3,144,000 | 850,000 | 1,070,000 |
| Total plant direct cost (TPDC) PC + I + II + III + IV + V + VI + VII + VIII | 4,301,000 | 4,471,000 | 24,677,000 | 26,566,000 | 7,183,000 | 9,042,000 |
| Engineering (IX) | 1,075,000 | 1,118,000 | 6,169,000 | 7,859,000 | 1,796,000 | 2,260,000 |
| Construction (X) | 1,505,000 | 1,565,000 | 8,637,000 | 9,298,000 | 2,514,000 | 3,165,000 |
| Total plant indirect cost (TPIC) IX + X | 2,581,000 | 2,683,000 | 14,806,000 | 17,157,000 | 4,310,000 | 5,425,000 |
| Total Plant Cost (TPC = TPDC + TPIC) | 6,882,000 | 7,154,000 | 39,483,000 | 43,723,000 | 11,492,000 | 14,466,000 |
| Contractor's fee (XI) | 344,000 | 358,000 | 1,974,000 | 2,186,000 | 575,000 | 723,000 |
| Contingency and research & development (XII) | 688,000 | 715,000 | 3,948,000 | 4,372,000 | 1,149,000 | 1,447,000 |
| Direct fixed capital cost (DFC) = TPDC + TPIC + XI + XII | 7,914,000 | 8,227,000 | 45,405,000 | 50,281,000 | 13,216,000 | 16,636,000 |
| Working capital (WC) | 396,000 | 411,000 | 2,270,000 | 2,514,000 | 661,000 | 832,000 |
| Start-up cost (SC) | 1,187,000 | 1,234,000 | 6,811,000 | 7,542,000 | 1,982,000 | 2,495,000 |
| Total capital investment (TCI) = DFC + WC + SC | 9,497,000 | 9,922,000 | 54,486,000 | 60,337,000 | 15,859,000 | 19,963,000 |

Scenario 1: wood pellet production from raw sawdust in a pellet plant; Scenario 2: straw pellet production from raw oat straw in a pellet plant; Scenario 3: integrated torrefied treated sawdust pelletization plant utilizing the torrefaction liquid (TL) as the pellet binder; Scenario 4: integrated torrefied treated oat straw pelletization plant utilizing the torrefaction liquid (TL) as a pellet binder; Scenario: integrated steam treated sawdust pellet plant; and Scenario: integrated steam treated oat straw pellet plant.

6.4.2 Operating cost

The annual operating cost (AOC) includes utilities, the costs associated with purchasing raw materials, chemicals, waste treatment and disposal, facility-dependent, labour-dependent, and laboratory cost. The feedstock cost, facility-dependent, labour-dependent, and the labour cost were the most influential parameters in all six Scenario. Figure 6.3 depicts the annual plant operating costs for each scenario. The main contributors to the operational expenditure (OPEX) in all scenarios were the cost of raw materials and facility dependent cost which ranged from 25 - 43% and 21-36%, respectively. The cost of labour (16-27%) was the third major contributor to the OPEX, while the utilities cost ranged from 9-19% of the OPEX. According to Sarker et al. (2023), raw material cost contributed about 19-48% of overall OPEX cost for both scenarios studied. Pirraglia et al. (2010) studied technoeconomic analysis of wood pellets production for U.S. manufacturers and reported that the cost of feedstock, labour and utilities shared 27%, 24% and 17%, respectively of OPEX. Alizadeh et al. (2023) reported that facility-dependent cost accounts for 56-61% of OPEX and 24% as utilities cost. An economic analysis of pellet production from untreated and torrefied oat hull was carried out by Valdez (2020), in which facility-dependent costs ranged from 28-35%, labour cost was 18%, and utilities cost shared 20%. Similarly, Sarker et al. (2023) and Alizadeh et al. (2023) reported the cost of labour as approximately 20% and 29%, respectively. The difference in the AOC of the compared studies could be attributed to the choice of feedstock and size of the plant.

Manouchehrinejad et al. (2021) conducted research on the technoeconomic analysis of integrated torrefaction and pelletization systems to produce torrefied wood pellets where the feedstock cost and the labour cost were reported to be the most influential parameters in all three Scenario studied. Also, Fadhilah et al. (2023) concluded that feedstock cost and labour cost were the most

expensive sections which accounted for about 27 and 43% of the unit production cost, respectively. Most of the production costs were from facility-dependent, raw material, and utility costs (Fadhilah et al. 2023).



Figure 6.3. Annual plant operating costs for each (1-6) Scenario.

6.4.3 Profitability evaluation

Pellets are the main product in this study, Scenarios 3 and 4 produced torrefaction liquid (TL) which was considered as a credit to the process. From a technoeconomic point of view, torrefaction byproducts, especially torrefaction liquids, have a higher profit potential because they contain formic acid, acetic acid, methanol, and furfural which can be used for the production of green chemicals (Sarkar et al. 2023). Torrefied liquid can also be used to produce “liquid smoke”. The economic value of liquid smoke in terms of its potential uses: natural food ingredient, preservation, flavoring, coloring of treated food, and nonfood uses (e.g., biofertilizer, biopesticide, growth-stimulant, other bioactive compounds (Valdez 2020; Sarkar et al. 2023).

The minimum selling price (MSP) of regular, torrefied, and steam exploded pellet from sawdust and oat straw was calculated for plant capacity of 9.09 dry t/h of feedstock processing at 10% discount rate. The MSP of regular pellets from sawdust and oat straw were \$113.4/t and \$118.7/t, respectively. The MSP of torrefied pellets from sawdust and oat straw were \$283.4/t and \$298.7/t, respectively, while the MSP of steam exploded pellets from sawdust and oat straw were \$200.5/t and \$208.4/t, respectively. These indicate that the project becomes profitable when the pellets’ selling price (PSP) is higher than the break-even point. Manouchehrinejad et al. (2021) reported that \$207/t was the MSP of torrefied wood pellet of 100 kt/yr plant capacity. Similarly, Agar (2017) calculated the MSP of torrefied wood pellet to be \$211/t. Sarkar et al. (2023) evaluated the MSP of pellets with additives and without additives as \$103.4/t and \$105.1/t, respectively. Shahrukh et al. (2016) determined the MSP of steam pretreated forest residue, wheat straw, and switchgrass, and reported \$148.50/t, \$152.63/t, and \$156.61/t, respectively. The difference in the MSP compared could be because of the cost of production for the various studies.

The effect of feedstock cost on NPV at different pellet selling prices is shown in Figure 6.4. Feedstock price is important, and it is attributed to pellet production cost. The assumptions regarding pellet price were made based on literature surveys. Mupondwa et al. (2012) reported an approximate price of \$120/t for wheat straw pellets, which were used for heat applications. Similarly, Pirragila et al. (2013) reported the price of torrefied wood pellet as \$260/t for 100 kt/h of plant capacity. In same manner, Sarker et al. (2023) considered \$170/t for torrefied canola residue pellet, while Shahrukh et al. (2016) conducted a technoeconomic assessment of pellets produced from steam pretreated biomass feedstock and reported a price range of steam exploded pellet and regular pellet from approximately \$140/t - \$160/t and \$80/t - \$95/t, respectively. Feedstock cost of \$70/t and \$75/t were considered for oat straw and sawdust, respectively in the baseline model, which resulted in a negative NPV at regular pellet selling price (PSP) which is below \$140/t (Figure 6.4a) and pretreated pellet selling price is below \$200/t (Figure 6.4b, c). A positive NPV was obtained at a PSP of \$140/t for untreated pellets, while steam exploded pellets sold for above \$200/t which resulted in a positive NPV. Figure 6.4b showed that if the PSP of torrefied pellet was \$260/t the NPV remained negative. The sensitivity of NPV to feedstock cost was performed by varying the cost of sawdust and oat straw by 25% (above and below the baseline price). Figure 4a showed that if the was PSP below \$120/t, the reduction of feedstock cost by 25% did not result in profitability of pellet production. However, when the PSP was increased to \$120/t and further to \$140/t and the feedstock cost was maintained at 25% less than the baseline price (\$75 and \$70 for sawdust and oat straw, respectively) these led to about 60% increase in NPV, which was approximately 2.3 times of the NPV baseline model. Similarly, Figure 6.4c showed that at a PSP below \$180/t for steam exploded pellets, reducing feedstock cost by 25% did not improve the economics of pellet production. However, increasing the PSP to

\$200/t and further to \$220/t and maintaining feedstock cost at 25% (\$56.25 and \$52.50) less than the baseline price (\$75 and \$70 for sawdust and oat straw, respectively) resulted in a 14% increase in NPV, which was approximately 1.2 times of NPV of the baseline model. Generally, increasing the feedstock cost by 25% (\$93.75 and \$87.50) more than the baseline price (\$75 and \$70 for sawdust and oat straw, respectively) did not result in the profitability of pellet production. Although Scenario 5 showed a positive NPV when the PSP was increased to \$220/t, it was 50% less than the NPV of the baseline model. Figure 6.4b depicts the NPV at a 7% discount rate for torrefaction pretreated pellets at various PSP and feedstock prices. The negative NPV was found in both scenarios when PSP was increased from \$180/t - \$260/t. Similarly, Valdez (2020) reported that the inclusion of the torrefaction equipment to the direct fixed capital cost made the project unprofitable. Figure 6.5 depicts the profitability analysis and the maximum pellet capacity of this study. The PSP of \$ 140/t, \$ 240/t and \$260/t were considered for regular pellets, steam pretreated pellets, and torrefied pellets, respectively. Scenarios 1 and 2 have 72 kt/year of regular pellets produced, which is equivalent to the initial feedstock (no loss). Scenarios 3-6 (steam explosion and torrefaction) resulted in the loss of some materials in the form of non-condensable gas due to the pretreatment process.

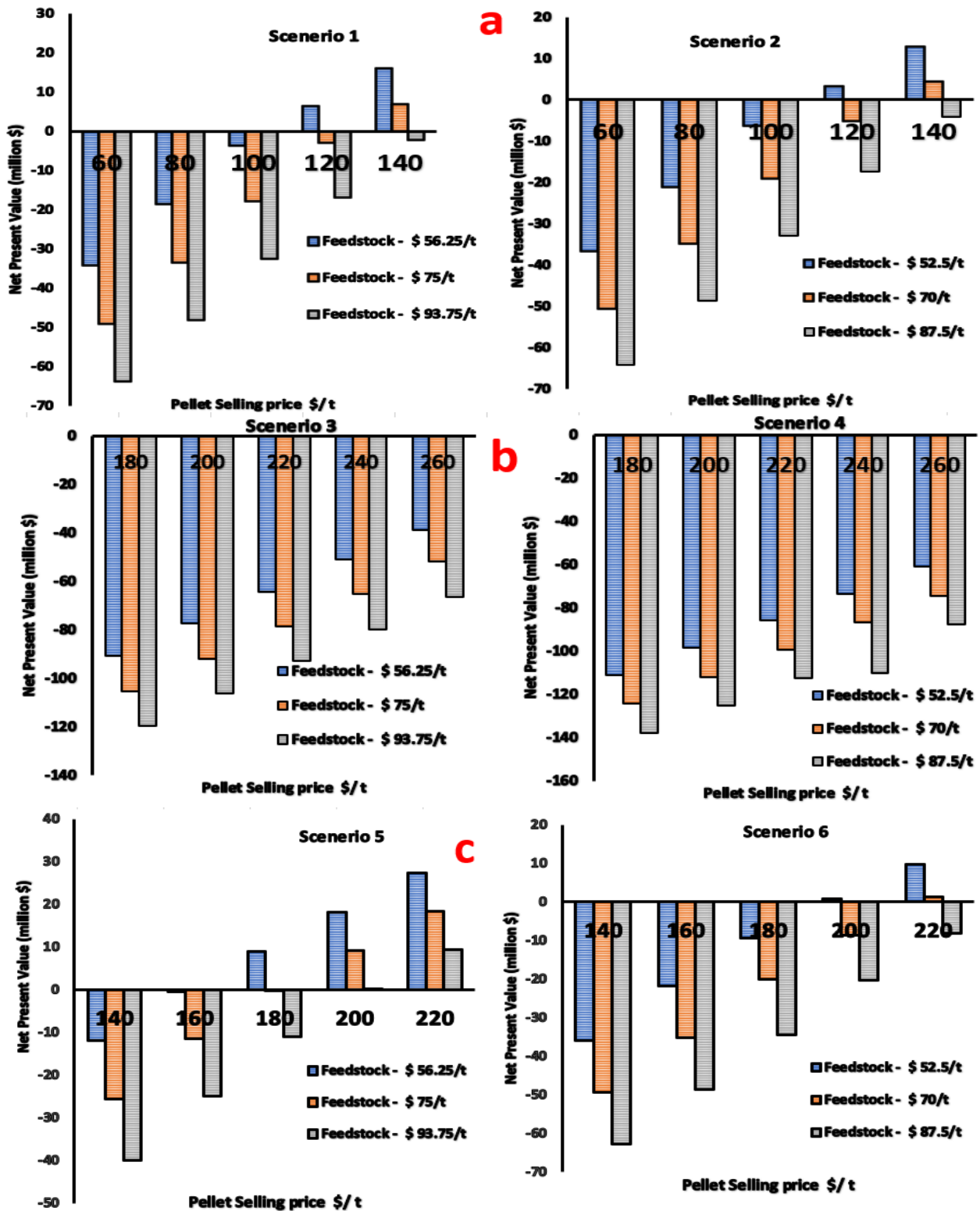


Figure 6.4. Net present value (NPV) at 7% discount rate for pellet at various pellet selling price and feedstock price; a) NPV for regular pellets; b) NPV for torrefied pellets; c) NPV for steam exploded pellets.

By analyzing the NPV, internal rate of return (IRR), and payback period (PBP), the profitability of fuel pellet production from various conditions was determined. Cash flow is generated as soon as the construction is concluded, and the project begins. These funds make it possible that eventually, the cash flow becomes positive. The PBP is the period required to recuperate the original investment. IRR is a metric that is used to evaluate the return of an investment. Any business must have a PBP that is less than the period of the project for it to be economically feasible. Considering the baseline models, this study found the PBP was lower for undiscounted and discounted cases for most of the scenarios when compared side by side with the entire life of the project. It can be highlighted that some of the proposed projects (Scenario 1, 2, 5, and 6) were economically profitable for production of fuel pellets. However, Scenarios 3 and 4 were not economically profitable to produce fuel pellets. The PBP only measures the duration to recover the initial investment and does not ensure the project's feasibility beyond that point. Therefore, additional profitability indicators such as NPV and IRR were considered to complement the PBP. The estimated IRR values were 19%, 11%, 10%, and 8% for scenarios 1, 2, 5, and 6, respectively. Since the same calculation applies to the various Scenarios, it is evident that all mentioned scenarios were profitable. The PBP for scenarios 1 and 2 were the lowest (2.48 yr and 2.59 yr, respectively), followed by scenarios 3 and 4 (4.07 yr and 4.64 yr, respectively) and finally scenarios 5 and 6 (6.27 yr and 6.78 yr, respectively). Although scenarios 3 and 4 had a higher investment but lower PBP than Scenarios 5 and 6, this may be because of the co-products from Scenarios 3 and 4 which generated more revenue. Similarly, Alizadeh et al. (2023) recorded PBP of 2.49 yr, 6.35 yr, and 3.38 yr for untreated wood pellet, steam exploded pellet, and torrefied pellet, respectively. In the same way, Sarker et al. (2023) studied techno-economic analysis of torrefied fuel pellet production from agricultural residue via integrated torrefaction

and pelletization process, reporting an estimated IRR value of 25% and 22% at 10% discounted rate for both scenarios.

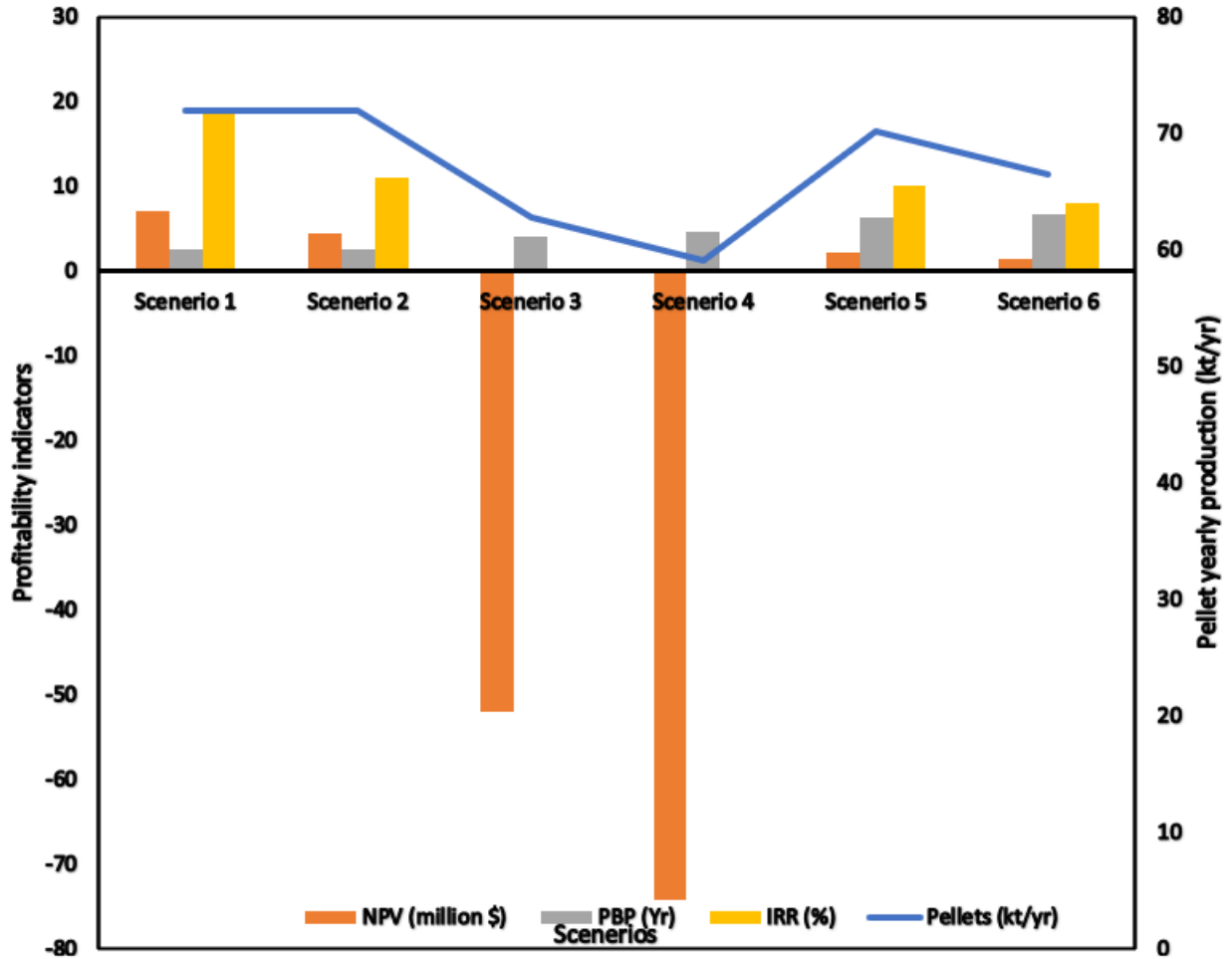


Figure 6.5. Profitability analysis and the maximum pellet capacity.

6.5. Conclusions

A technoeconomic analysis to produce regular and pretreated pellets via integrated steam explosion, torrefaction, and pelletization system was conducted. Four scenarios of treated pellets were estimated and compared with two conventional pellets processes. For the base scenario, the model indicated a plant size of 72 kt/year for both regular pellet scenarios and approximately 59

kt – 67 kt and 67 kt – 70 kt for torrefied and steam pretreated pellets, respectively. From the profitability analysis, facility-dependent and feedstock cost were the major significant contributors to the pellet production cost. The minimum selling price of pellet for the six Scenarios were evaluated using a 10% discount rate, where the net present value (NPV) is equal to zero. The profitability of the pellet production, as determined by the NPV, internal rate of return (IRR), and payback period, was found to be sensitive to the variations in feedstock cost. The baseline model discussed in this study does not represent an existing pellet plant. Nevertheless, this analysis is poised to establish a benchmark for industry stakeholders intending to implement the new technology in the pelletization of oat straw and sawdust in Saskatchewan.

Chapter 7

General Discussion

7.1 Overall Ph.D. Project Discussion

As the use of lignocellulosic biomass has increased due to its potential for producing biofuels and bio-based chemicals in an environmentally sustainable manner, researchers have been challenged with developing a better knowledge of lignin structure, quantity, and potential uses. The breakdown of lignin in the cell walls of cellulosic biomass is necessary to provide ease of access to cellulose fibers, and to loosen inherent binders to enhance the mechanical strength of biomass pellets. Therefore, biomass pretreatment modifies the structure of the biomass feedstock, which facilitates pelletization. This study explored the possibilities of steam explosion and torrefaction pretreatment as strategies for improving the physiochemical and mechanical properties sawdust and oat straw with the aim of maintaining an economic value and reducing the negative environmental impact of pellet production. These two pretreatment techniques are well-established, steam explosion has been used in forest feedstocks, this study used oat straw to establish its processing parameters.

Steam explosion is a physiochemical pretreatment process that breaks down lignocellulosic biomass by the application of high-pressure heat, which results in the formation of organic acid, and shearing forces causing moisture expansion and explosive decompression are the two stages involved in the steam-explosion process. These processes modify the biomass components through hydrolysis of hemicellulosic components (resulting in the release of mono- and oligosaccharides), alterations to the chemical structure of lignin, and enhancement of the crystallinity index of cellulose. These conditions allow the lignocellulosic biomass structures to

unlock and improve the quality of pellets generated from these feedstocks. Residence time during steam explosion has a major effect on the amount of degradation of the products that are observed, which must be minimized. Moreover, pressure is another important parameter that is directly related to temperature and influences the kinetics of the production of degradation products and hydrolysis of cellulose fractions. Additionally, the difference between the atmospheric pressure and that of the reactor is proportional to the severity of the shearing forces applied to the biomass when the pressure is suddenly and explosively released. Therefore, the application of steam explosion pretreatment to lignocellulose must take into consideration the variation of the operation conditions to maximize the key effects of process. Optimization of steam pretreatment conditions for oat straw and sawdust was conducted in this study. Temperature and residence time are significant factors for the steam explosion pretreatment of sawdust and oat straw.

Similarly, torrefaction is a biomass heat operation associated with roasting a biomass sample in a controlled environment at torrefaction temperatures, which range from 200 to 300 °C. Torrefaction is commonly known as mild pyrolysis, and it has been investigated for the pretreatment of biomass to make the biomass more hydrophobic, reduce its moisture content, and increase its calorific value, resulting in an increase in the volumetric energy density of the biomass. In addition, torrefaction with pelletization can reduce the transportation costs of biomass and intensify its heating value and hydrophobicity. These changes make biomass more combustible, storable, and attractive. Torrefaction yields are both the solid fuel (torrefied material) and the torgas. The torgas is composed of torrefaction liquid (TL), and non-condensable gases. The TL is rich in furfural, ketones, organic acids, water, and other minor components, which could be transformed into commercially feasible products. Hence, to

improve the thermal and physiochemical properties of solid biofuel pellets from torrefied oat straw, this work used the TL from this process as a binder. This strategy aims to solve the difficulty relating to the promotion of systems and products and equaling energy efficiency and fuel quality. The torrefaction gases (condensable liquid) contain a large amount of energy. Therefore, introducing a closed recycling system has the potential to be economically viable for the torrefaction system. It is significant that this energy is utilized productively in the system (torrefaction and densification) to approach and settle the process stability, system reliability, and safety problems currently averting commercial-scale torrefaction. The combined effect of torrefaction and binders on the mechanical and physiochemical characteristics of pellets formed from torrefied biomass was also studied.

Laboratory-scale application of torrefaction pretreatment needs low energy input; however, at the industrial scale, it is expensive due to high energy required. Technoeconomic analysis is important for evaluating the economic feasibility of the pathways to commercial production of bioenergy. The economic parameters such as total revenue produced by the plant and payback time are the key factors that investors will assess for taking investment decisions. Therefore, technoeconomic analysis was carried out using this study's optimum steam explosion and torrefaction pretreatment conditions.

7.2 Achievement of Research Objectives

The main objective of this study is to investigate the effect of steam explosion and torrefaction pretreatment strategies to improve the accessibility of the energy potentials and enhance the densification process of sawdust and oat straw for biofuel production. The goal of this study was accomplished by realizing the following specific objectives as

listed in section 1.5. The thesis chapters in which the specific objectives are met are included in parentheses after each numbered listing for reference.

1. Investigate the steam explosion pretreatment of sawdust and oat straw to improve their quality as biofuel pellets; (Chapters 2 and 3)
2. Investigate torrefaction pretreatment of sawdust and oat straw to improve their quality as biofuel pellets; (Chapters 2 and 4)
3. Investigate the combined effect of pretreatment and binders on the quality of pellet formed; (Chapters 4 and 5) and
4. Carry out a comparative techno-economic analysis of pellet production from sawdust and oat straw using different pretreatment methods (Chapter 6).

7.3 Contribution to Knowledge Advancement

The novel contribution of this study to academic knowledge advancement is the exploration of the most current issue regarding the practicality of torrefaction systems and products for industrial applications. The problems concerning the promotion of systems and products and balancing energy efficiency and fuel quality remain unsolved. During torrefaction pretreatment, several reaction products are formed. The yield depends on torrefaction conditions (temperature and time) and on biomass properties. These torrefaction gases contain a large amount of energy. Introducing a closed recycling system has been shown to be economically viable for the torrefaction system, and it is important that this energy is utilized efficiently in the system (torrefaction and densification). The use of these gases throughout the torrefaction system has

addressed and resolved the process stability, system reliability, and safety issues currently preventing commercial-scale torrefaction.

The results of the study will provide useful information on the optimum steam explosion and torrefaction pretreatment process parameters studied for improved quality of bio-pellets. This development will encourage investment in biofuel production. Furthermore, information on cost implications in this research for biofuel production will guide the research and development of these technologies for efficient cellulosic biorefinery concepts. The minimum selling price of pellets obtained from Scenarios 1-6 were \$113.4/t, \$118.7/t, \$283.4/ t \$298.7/t, \$200.5/t, and \$208.4/t, respectively. The profitability of pellet production as determined by the net present value (NPV), internal rate of return (IRR), and payback period and was found to be sensitive to variations in feedstock cost.

Chapter 8

Summary, Conclusions and Recommendations

8.1 Overall Summary and Conclusions

Lignocellulosic biomass is a renewable resource that can be used in the production of biofuels and platform chemicals in addition to bioenergy. As the most abundant carbon-neutral resource, using biomass as a replacement for fossil fuels can help mitigate environmental pollution. However, several physical, chemical, and structural factors can hinder effective and efficient conversion of biomass into fuels and chemicals. Among the processes involved in transforming lignocellulosic biomass to useful biofuel at large scale, pretreatment of feedstock is seen as the most critical due to its negative impact on the overall cost of biofuel production. Because of the high economic cost and severity of most of these pretreatment methods, their applications at the industrial scale are very limited. Knowledge of the properties of biomass will be relevant in the allocation of feedstock to the appropriate end-use. An ability to determine these properties promptly is also necessary for the commercialization of biomass-based products. Thus, there is a need for high throughput methods and equipment in the characterization and monitoring of the feedstock as conventional methods have been destructive and laborious. However, the effect of various preprocessing and pretreatment methods on the lignocellulosic matrix is not well understood. Applications of pre-processing methods such as size reduction or increasing porosity, and pretreatment techniques such as steam explosion and torrefaction on agricultural biomass have demonstrated an improvement in pellet (compact) quality that can be linked to the changes in the lignocellulosic components and distribution.

This work investigated the effects of temperature, time, and moisture content on the mechanical properties of biomass (oat straw and spruce sawdust) pellets. These variables showed a significant effect on all the response variables studied for sawdust but not for oat straw. This study indicated that the most significant ($p < 0.05$) factors in the steam explosion of the biomass for pellet production were temperature and time. The p-values associated with the regression models for the dimensional stability, pellet unit density, and tensile strength of the steam exploded pellets were significant ($p < 0.05$). Investigation of the steam explosion technology shows that some factors influence the productivity of the process, including retention time, initial sample moisture content, and pressure. Steam explosion breaks up the lignocellulosic structure of biomass, leading to lower bulk densities of pellets. After steam explosion pretreatment the carbon and the nitrogen content of the steam-exploded sample increased significantly ($p < 0.01$), concomitant with temperature and residence time, while biomass hydrogen and oxygen content decreased. Hydroxyl groups are linked to biomass structural components, and their volatilization during steam explosion leads to an increase in the energy density of the residual carbon-rich solids. In addition, the carbonization and autohydrolysis process in the steam explosion removes the extractives with low volatility, resulting in increased HHV. Higher moisture content negatively affects the HHV of the solid pellet, while increased steam explosion temperature positively affects the HHV. During the steam explosion pretreatment, the lignocellulosic structure of the straw and sawdust are disintegrated by the sudden pressure change and the lignin and hemicellulose are hydrolyzed by this process. This process tends to destroy the OH group, thereby causing the biomass sample to lose the ability to form a hydrogen bond and increasing the hydrophobic characteristic of the sample. This study showed that steam-exploded pellet samples became more hydrophobic as the treatment severity condition increased. Extensive

defibrillation of fibers was observed after the steam explosion pretreatment, mainly because of the mechanical effect from the adiabatic expansion of absorbed water during the process. SEM images of the pellets from steam-exploded pellets reveal more tightly bonded particles and cemented surfaces with fewer pores when compared with the untreated pellets. In this study, steam explosion pretreatment resulted in positive effects on the quality of pellets.

The torrefaction pretreatment of white spruce sawdust (WSS) was conducted using a lab-scale microwave oven at temperatures in the range of 200–300 °C and a retention time of 5–9 min in an inert environment to ascertain the effect of these two process variables on the physiochemical properties of the WSS. The products of this process are solid carbon, commonly known as biochar, condensable torrefaction liquid (TL), and non-condensable gases. The result from this study indicates the potential for the torrefaction pretreatment operation to enhance the physiochemical properties of WSS. The hemicellulose content underwent a major degradation as the torrefaction severity increased. Similarly, cellulose also underwent decomposition, but with less degradation. The incomplete depolymerization of cellulose and the disintegration of hemicellulose at the same time increased the proportional lignin content during the pretreatment process, enhancing the FC and drastically decreasing the VM content. This led to an improvement in the higher heating value (HHV), hydrophobicity, bulk, particle density, pellet dimensional stability, and pellet density.

During the torrefaction process, a significant mass loss associated with the decomposition of hemicellulose was observed. The hemicellulose content drastically reduced to approximately 1.8% dw and the cellulose content reduced by approximately 10% dw, while the lignin gained (by proportion) approximately 35% as the severity increased. The pellet unit density and pellet tensile strength in this study decreased as the torrefaction severity increased. The breakdown of

the hemicellulose and cellulose polymer was suggested to be one reason for the tensile strength loss of the biomass pellets under severe torrefaction treatment. However, the tensile strength of the studied pellets was improved by the introduction of binders. The OH groups in the biomass are destroyed to a limited degree by dehydration. This hinders the formation of H bonds, thereby encouraging the pretreated biomass to become hydrophobic. From the SEM graphs, it was observed that the torrefied WSS pellets showed more firmly glued surfaces with fewer pore spaces when compared with the raw pellets. The thermogravimetric analysis conducted showed that the torrefaction of the WSS slightly reduced its thermal stability.

The combined effects of torrefaction and binders on the quality of pellets produced from oat straw were studied. The results of this study indicate the potential for torrefaction pretreatment to enhance the mechanical, physiochemical, and thermal properties of torrefied oat straw pellets. In addition, there were noticeable improvements in the physiochemical properties of the pellets formed after torrefaction, except for the tensile strength. The introduction of binders improved the binding characteristics among torrefied material particles during pelletization, which resulted in improved pellet tensile strength and further enhanced other pellet properties. Oat straw can be torrefied and pelletized using torrefaction liquid as a binder to make pellets for biofuel production.

Technoeconomic analysis of six case scenarios to develop a process model for bioenergy production from sawdust and oat straw that uses torrefaction and steam explosion pretreatment prior to pelletization was carried out using SuperPro Designer. The pellet plants, with processing capacities of 9.09 t/h of sawdust and oat straw had pellet yield ranging from 59 kt – 72 kt annually. Scenarios 1 and 2 had 72 kt regular pellet produced, which is equivalent to the initial feedstock (zero waste). Scenarios 3-6 (steam explosion and torrefaction) simulated the loss of

some materials in the form of liquid and gas due to pretreatment process. Scenarios 1, 2, 5, and 6 generated positive net present value at baseline model. Facility-dependent and feedstock cost were the major significant contributors to the pellet production cost. The profitability of the pellet production, as determined by the net present value (NPV), internal rate of return (IRR), and payback period, was found to be sensitive to the variations in feedstock cost. The baseline model discussed in this study does not represent an existing pellet plant, rather it is a hypothetical virtual pellet plant with few details for a more precise approximation of the process economics.

8.2 Recommendations for Future Work

The following recommendations are made for future studies:

1. The dark brown liquor from steam exploded pretreatment slurry can be separated via filtration and analyzed to find further uses of this byproduct.
2. The results of this study showed that torrefaction liquid is a suitable binder to make pellets for biofuel production as it improved pellet tensile strength and further enhanced other pellet properties. It is important to study the inherent binder characteristics of TL and its role in pellet bonding mechanisms.
3. A comparative study on different kinds of binders for pelletization as compared to TL should be performed.
4. A study on the relationship between lignocellulosic biomass components and the quality of pellet produced, for instance mechanical strength of pellet, is recommended for future work.

5. The utilization of pellets produced from steam explosion and torrefaction pretreatment in co-firing with coal can be examined with different ratios and in the same burners as coal-fired power plants.
6. A blending of steam exploded and torrefied biomass should be explored for pellet production, and their thermochemical and physiochemical properties can be determined and compared with individual process.
7. Assessment of the environmental performance of torrefaction and steam explosion pretreatment of biomass for pellet production through life cycle assessment should be carried out.
8. Microstructural characterization in different orientation and how particles are arranged should be performed using high resolution imaging.
9. The relationship between pellet tensile strength and durability can be determined and compared.

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Appendix A

Supplementary data for Chapter 3. Investigation of steam explosion pretreatment of sawdust and oat straw to improve their quality as biofuel pellet.



Figure S3.1. Photographs of the steam explosion system used in the experiments. (a) steam generator; (b) pressure vessel (chamber); (c) cyclone.

Table S3.1. Severity factor of steam exploded biomass.

| Temp (°C) | time (min) | severity factor |
|--------------|---------------|--------------------|
| 160 | 5 | 2.47 |
| 160 | 7 | 2.61 |
| 160 | 9 | 2.72 |
| 180 | 5 | 3.05 |
| 180 | 7 | 3.20 |
| 180 | 9 | 3.31 |
| 200 | 5 | 3.64 |
| 200 | 7 | 3.79 |
| 200 | 9 | 3.90 |

Appendix B

Supplementary data for Chapter 4. Effect of torrefaction on the physiochemical properties of white spruce sawdust for biofuel production.

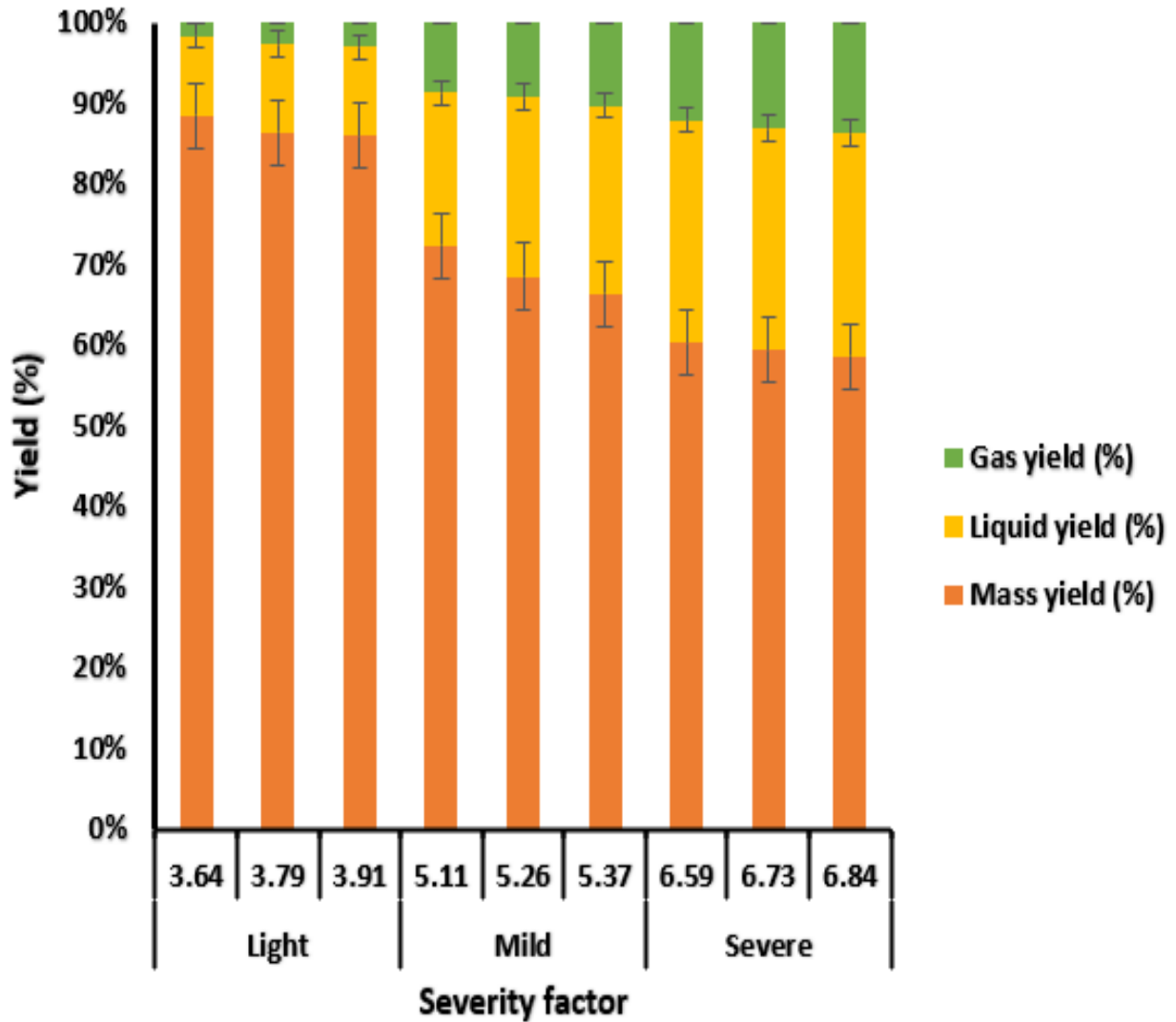


Figure S4.1. Product yields of torrefied white spruce sawdust (WSS).

Table S4.1. Elemental and proximate composition of untreated and torrefied WSS.

| Temp (°C) | RT (min) | R | M.C (%) | C (%) | H (%) | N (%) | S (%) | O ^a (%) | Ash ^b | VM ^b | FC ^b |
|--------------|-------------|------|------------|----------|----------|----------|----------|-----------------------|------------------|-----------------|-----------------|
| NT | NT | NT | 8.69 | 47.47 | 6.66 | 0.11 | 0.21 | 45.41 | 0.14 | 86.42 | 13.44 |
| 200 | 5 | 3.64 | 8.62 | 48.80 | 5.64 | 0.38 | 0.20 | 44.69 | 0.29 | 83.56 | 16.15 |
| 200 | 7 | 3.79 | 8.60 | 49.85 | 5.52 | 0.40 | 0.18 | 43.76 | 0.29 | 81.22 | 18.49 |
| 200 | 9 | 3.91 | 8.55 | 50.17 | 5.48 | 0.40 | 0.15 | 43.50 | 0.30 | 79.59 | 20.11 |
| 250 | 5 | 5.11 | 8.51 | 50.27 | 5.44 | 0.41 | 0.13 | 43.43 | 0.32 | 78.02 | 21.66 |
| 250 | 7 | 5.26 | 8.49 | 51.22 | 5.41 | 0.43 | 0.13 | 42.43 | 0.38 | 76.50 | 23.12 |
| 250 | 9 | 5.37 | 8.41 | 51.87 | 5.39 | 0.43 | 0.15 | 41.74 | 0.42 | 75.44 | 24.14 |
| 300 | 5 | 6.59 | 8.33 | 52.06 | 5.32 | 0.44 | 0.12 | 41.52 | 0.54 | 70.43 | 29.03 |
| 300 | 7 | 6.73 | 8.27 | 52.69 | 5.27 | 0.44 | 0.12 | 40.92 | 0.58 | 68.89 | 30.53 |
| 300 | 9 | 6.84 | 8.24 | 54.25 | 5.22 | 0.44 | 0.11 | 39.34 | 0.61 | 67.66 | 31.73 |

Temp = temperature; RT = residence time; R = severity factor (which is a function of temperature and time); M.C = moisture content; C = carbon; H = hydrogen; N = nitrogen; S = sulfur; O = oxygen; VM = volatile matter; FC = fixed carbon; ^a= calculated by difference; ^b= dry basis; and NT = non-treated.

Table S4.2. Pellet properties of torrefied WSS.

| Temp (°C) | RT (min) | R | Pellet Density (kg/m ³) | Dimensional Stability (%) | Tensile Strength (MPa) |
|--------------|-------------|------|---|---------------------------------|------------------------------|
| NT | NT | NT | 1048.56 | 2.87 | 1.68 |
| 200 | 5 | 3.64 | 1022.34 | 2.12 | 1.82 |
| 200 | 7 | 3.79 | 1017.67 | 1.61 | 1.86 |
| 200 | 9 | 3.91 | 1001.89 | 1.22 | 1.84 |
| 250 | 5 | 5.11 | 997.31 | 1.06 | 1.34 |
| 250 | 7 | 5.26 | 992.00 | 0.82 | 1.26 |
| 250 | 9 | 5.37 | 973.08 | 0.94 | 1.23 |
| 300 | 5 | 6.59 | 934.84 | 0.67 | 0.85 |
| 300 | 7 | 6.73 | 930.61 | 0.54 | 0.81 |
| 300 | 9 | 6.84 | 915.42 | 0.66 | 0.78 |

Temp = temperature; RT = residence time; R = severity factor; NT = untreated.

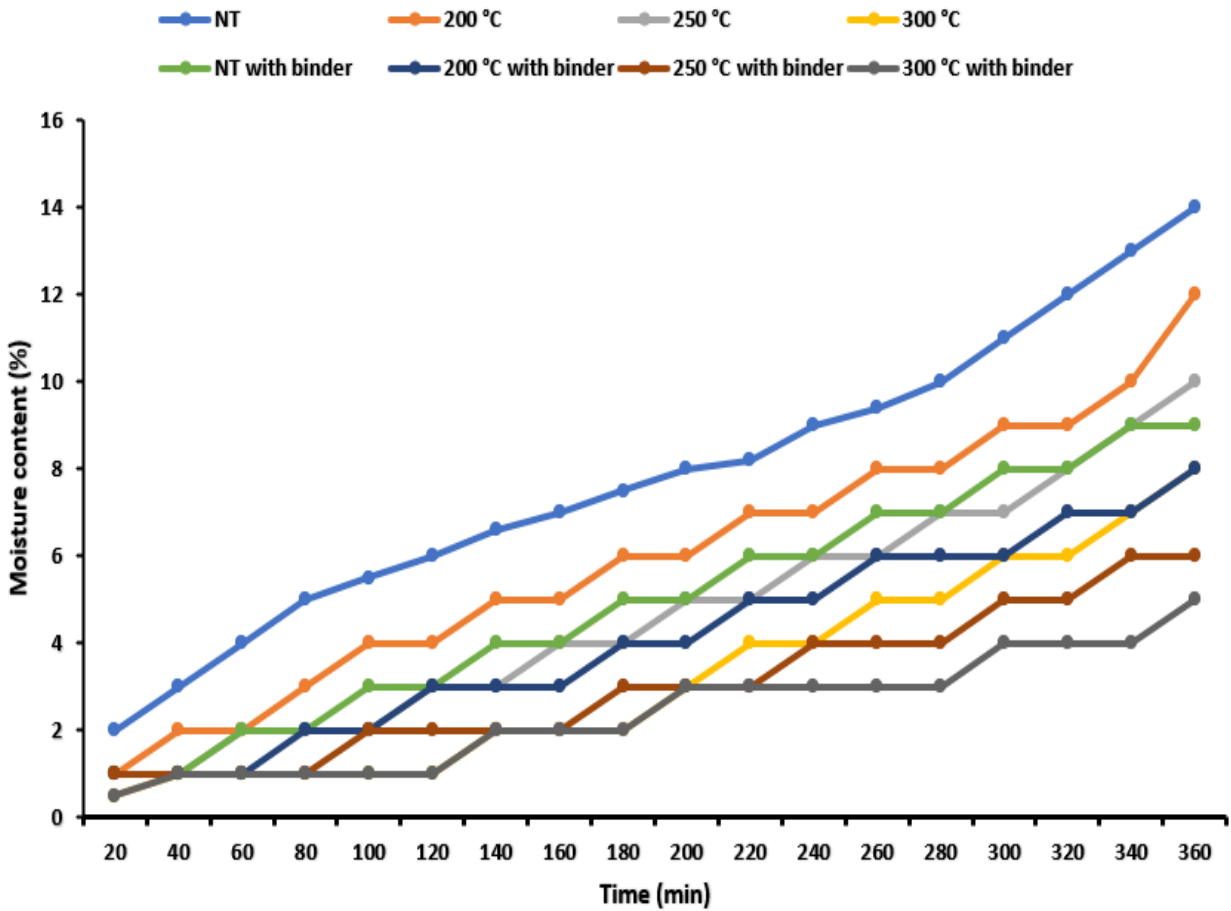


Figure S4.2. Effect of the binders on the moisture absorption rate of the pellets.

NB: NT = Untreated.

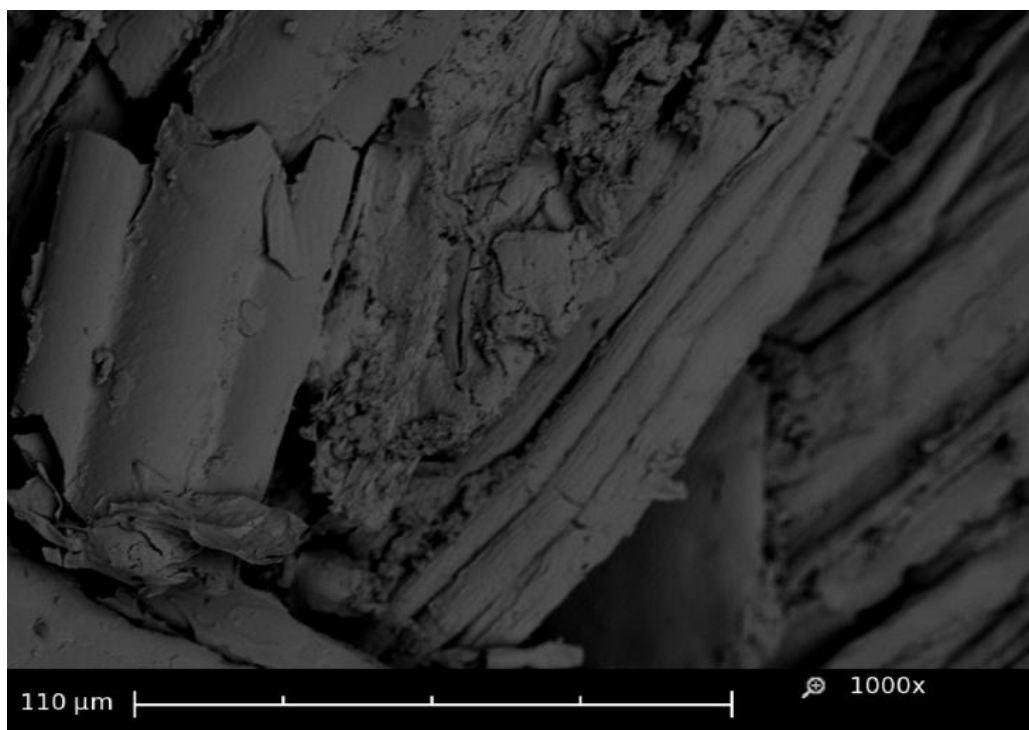


Figure S4.3. Higher magnification of scanning electron microscope image of the longitudinal cross-section of the untreated pellet.

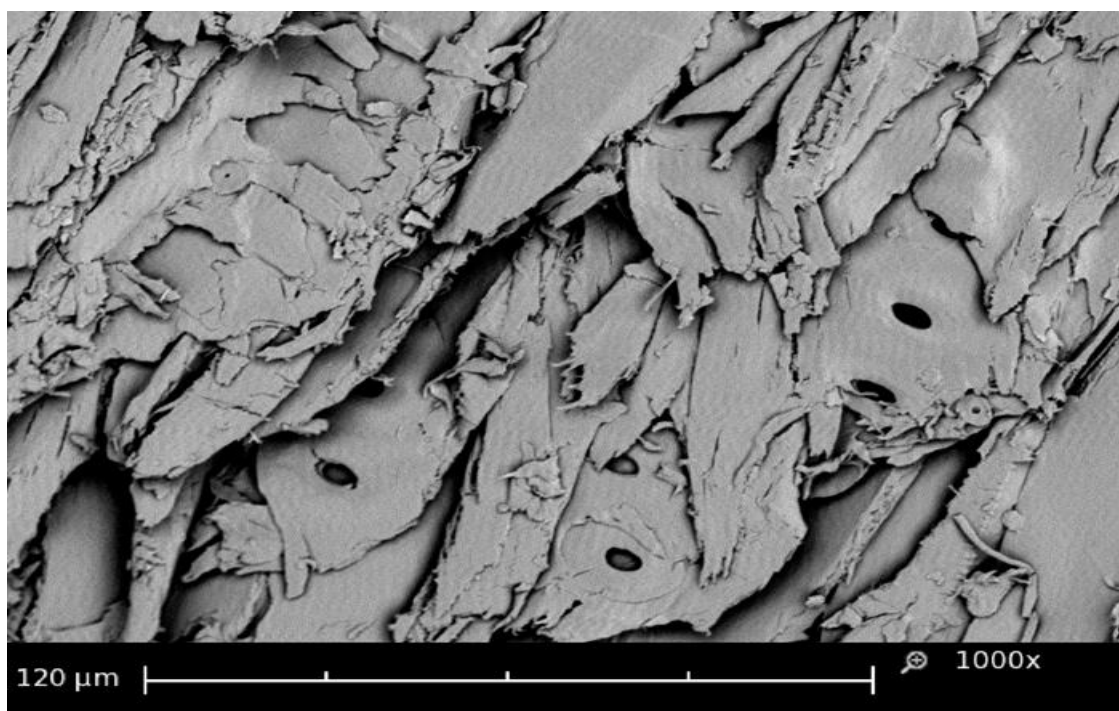


Figure S4.4. Higher magnification of Scanning electron microscope images of the longitudinal cross-section of WSS torrefied at 200 °C.

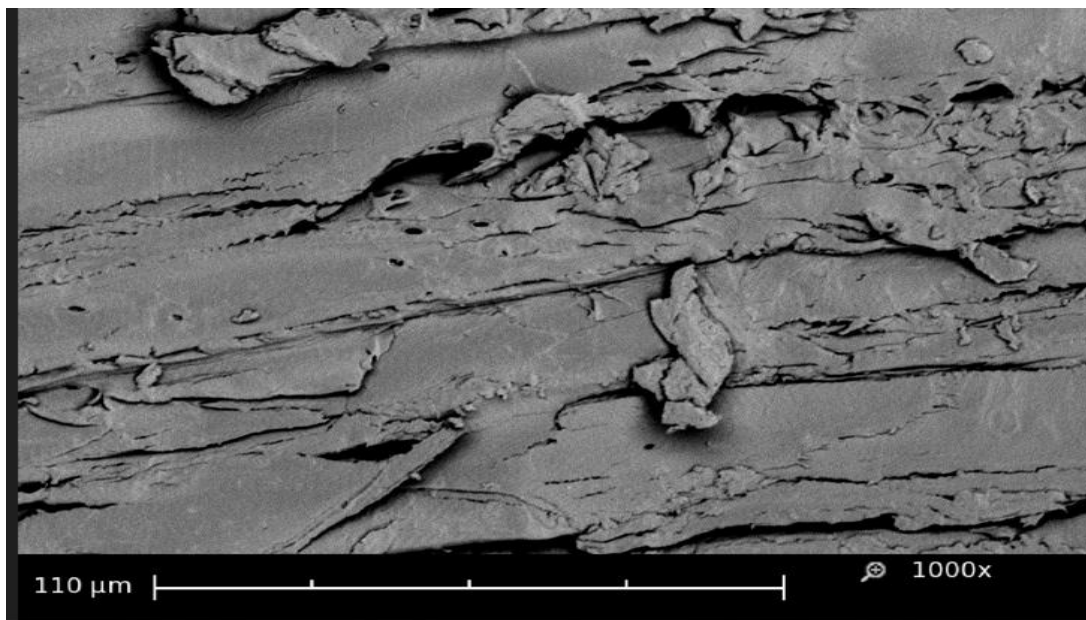


Figure S4.5. Higher magnification of Scanning electron microscope images of the longitudinal cross-section of WSS torrefied at 250 °C.

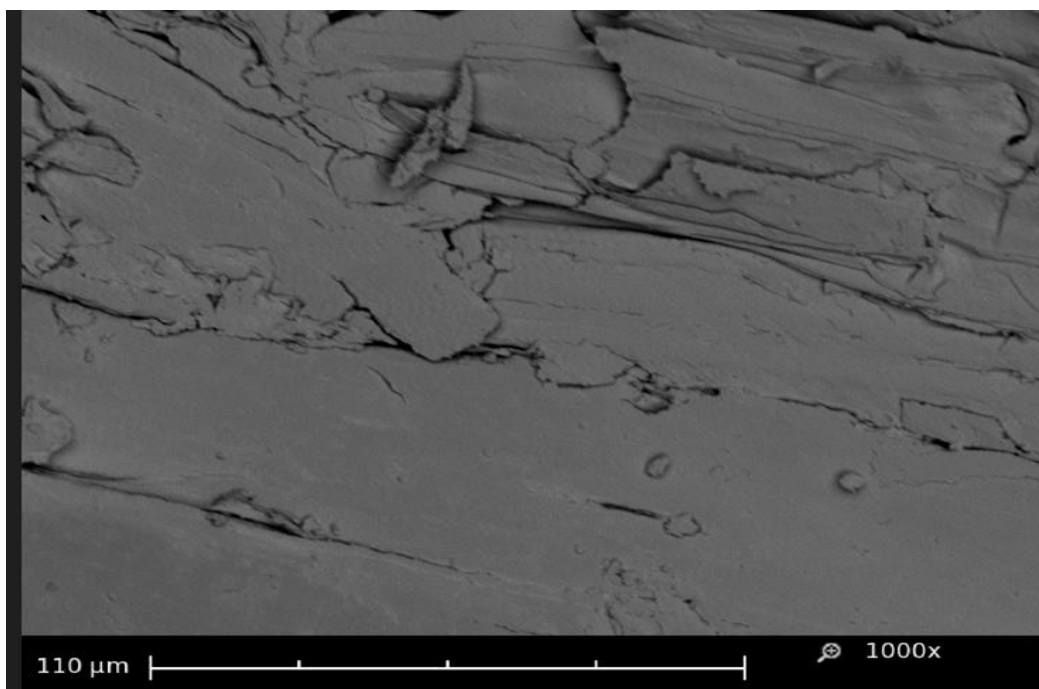


Figure S4.6. Higher magnification of Scanning electron microscope images of the longitudinal cross-section of WSS torrefied at 300 °C.

Appendix C

Supplementary data for Chapter 5. Combined effects of torrefaction and binders on the quality of pellets produced from oat straw.

Table S5.1. Severity factor of microwave-assisted torrefaction of oat straw.

| Temperature (°C) | Time (min) | Severity factor |
|------------------|------------|-----------------|
| Untreated | - | - |
| 200 | 5 | 3.64 |
| 200 | 7 | 3.79 |
| 200 | 9 | 3.91 |
| 250 | 5 | 5.11 |
| 250 | 7 | 5.26 |
| 250 | 9 | 5.37 |
| 300 | 5 | 6.59 |
| 300 | 7 | 6.73 |
| 300 | 9 | 6.84 |

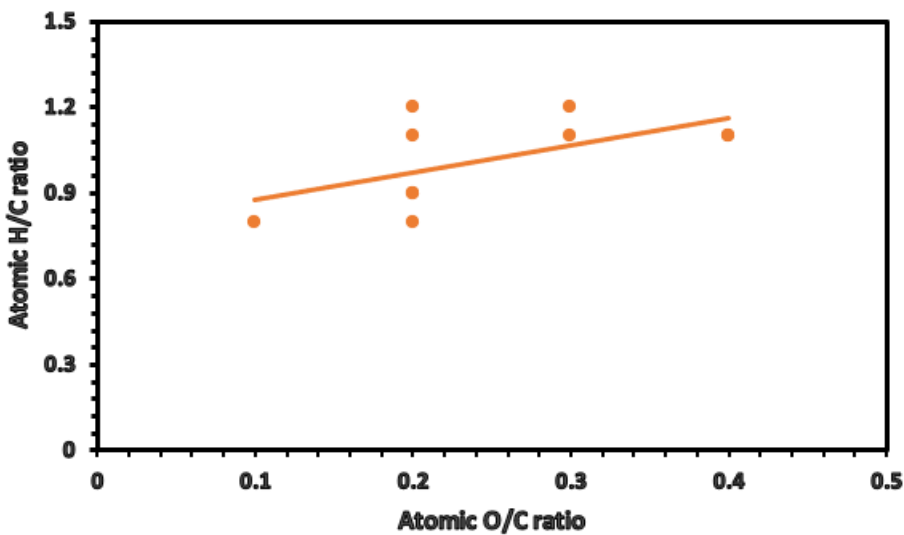


Fig. S5.1. The Van Krevelen diagram.

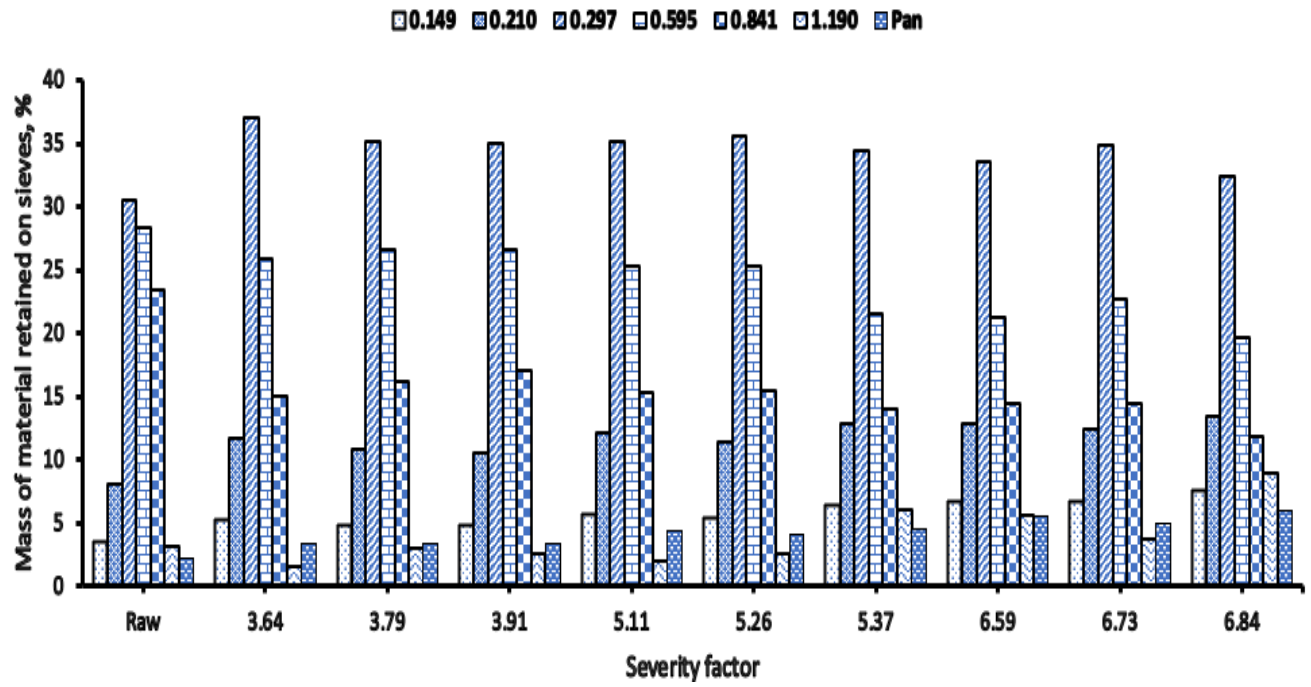


Fig. S5.2. Particle size distribution of untreated and torrefied oat straw at different severities.

Appendix D

Supplementary data for Chapter 6. Technoeconomic analysis of torrefaction and steam explosion pretreatment prior to pelletization of selected biomass.

Table S6.1. Properties of the feedstocks.

| Properties | Oat straw | Sawdust | Source |
|-----------------------------------|------------------|----------------|---------------|
| Bulk density (kg/m ³) | 127.87 | 158.00 | [29,31] |
| HHV (MJ/kg) | 16.84 | 18.02 | [29,31,32] |
| Cellulose (%) | 47.45 | 41.24 | [29,31] |
| Hemicellulose (%) | 24.70 | 15.72 | [29,31,32] |
| Lignin (%) | 12.92 | 25.16 | [29,31] |
| Ash (%) | 5.32 | 0.54 | [29,31] |
| Other solids (%) | 9.61 | 7.34 | [29,31] |
| Moisture content (%) | 10 | 10 | [29,31] |

HHV: Higher heating value.

Table S6.2. Cost assumptions and economic evaluation parameters.

| Parameters | Value | Source |
|--------------------------------------|------------------------|---|
| Time parameters | | |
| Year of analysis | 2023 | |
| Currency | US Dollars (\$) | |
| Construction period | 12 months | |
| Startup period | 6 months | |
| Project lifetime | 20 years | |
| Inflation | 2% | |
| Financial parameters | | |
| Depreciation style | Straight line | |
| Depreciation time | 10 years | |
| Revenue tax rate | 40% | |
| Salvage value | 5% of DFC | |
| Discount rate | 10% | |
| Operating cost | | |
| Operation capacity | 90% | |
| Labor | \$ 69/h | SuperPro designer databank |
| Sawdust | \$ 75/t | Antonio et al. (2021) |
| Oat straw | \$ 70/t | BIMAT (2010) |
| Nitrogen gas | 0.11 / Nm ³ | EIA (2021) |
| Selling price of pellet | \$60/t - \$260/t | (Shahrukh et al. 2016; Pirraglia et al. 2013; Sarker et al. 2023) |
| Selling price of torrefaction liquid | \$0.25/L | Badger et al. (2011) |
| Utilities | | |
| Steam (High P) | \$ 20/t | SuperPro designer databank |
| Steam | \$ 12/t | SuperPro designer databank |
| Cooling water | \$ 0.05/t | SuperPro designer databank |
| Electricity | \$ 0.1/kW-h | SuperPro designer databank |

DFC: Direct fixed capital cost.

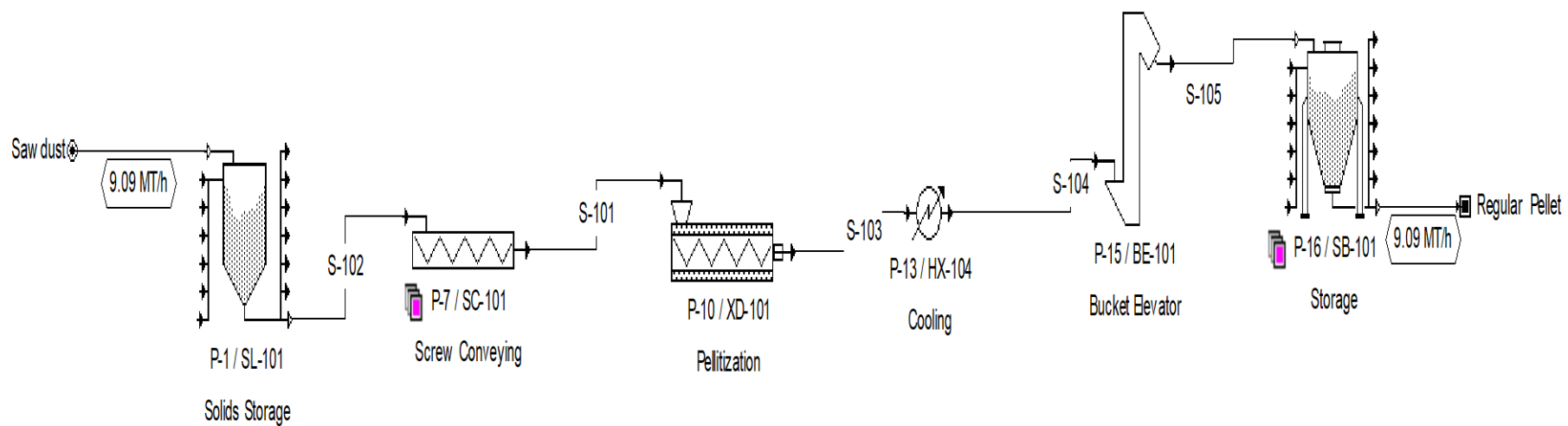


Figure S6.1. The process of pelleting raw sawdust (Scenario 1).

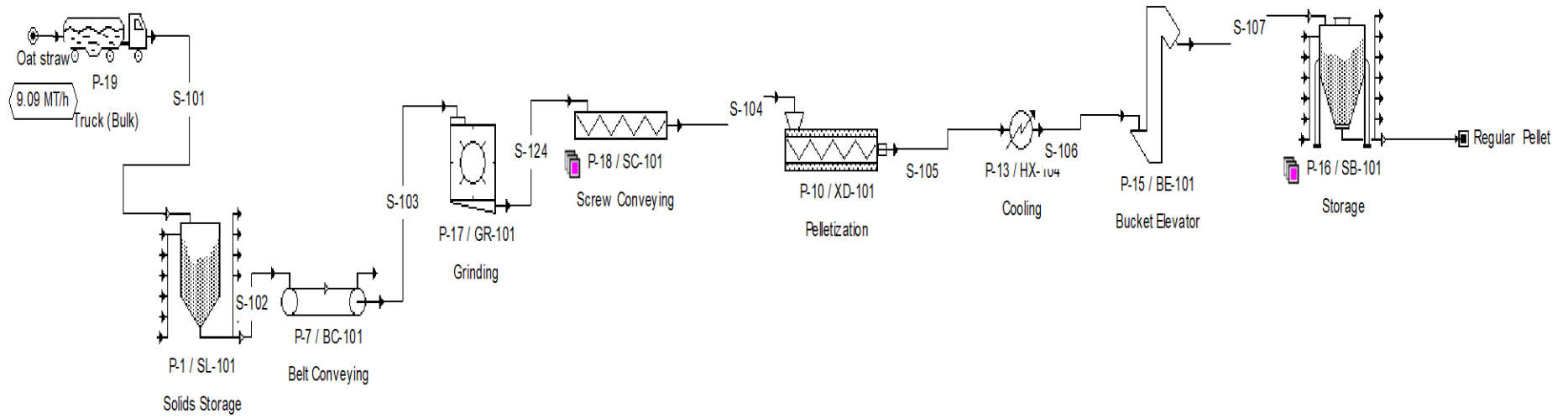


Figure S6.2. The process of pelleting raw oat straw (Scenario 2).

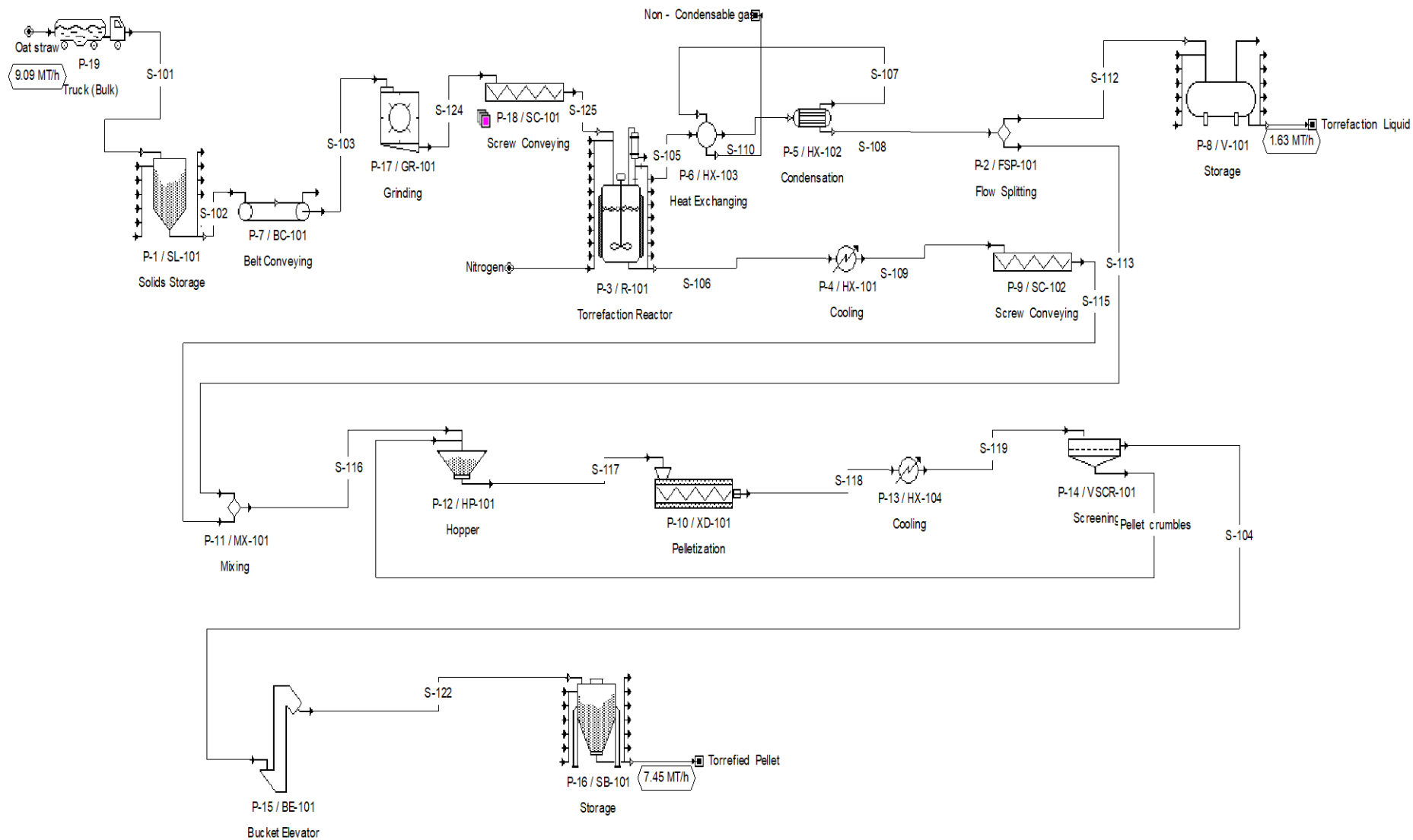


Figure S6.3. Integrated torrefied treated oat straw pelletization plant utilizing torrefaction liquid (TL) as binder (Scenario 4).

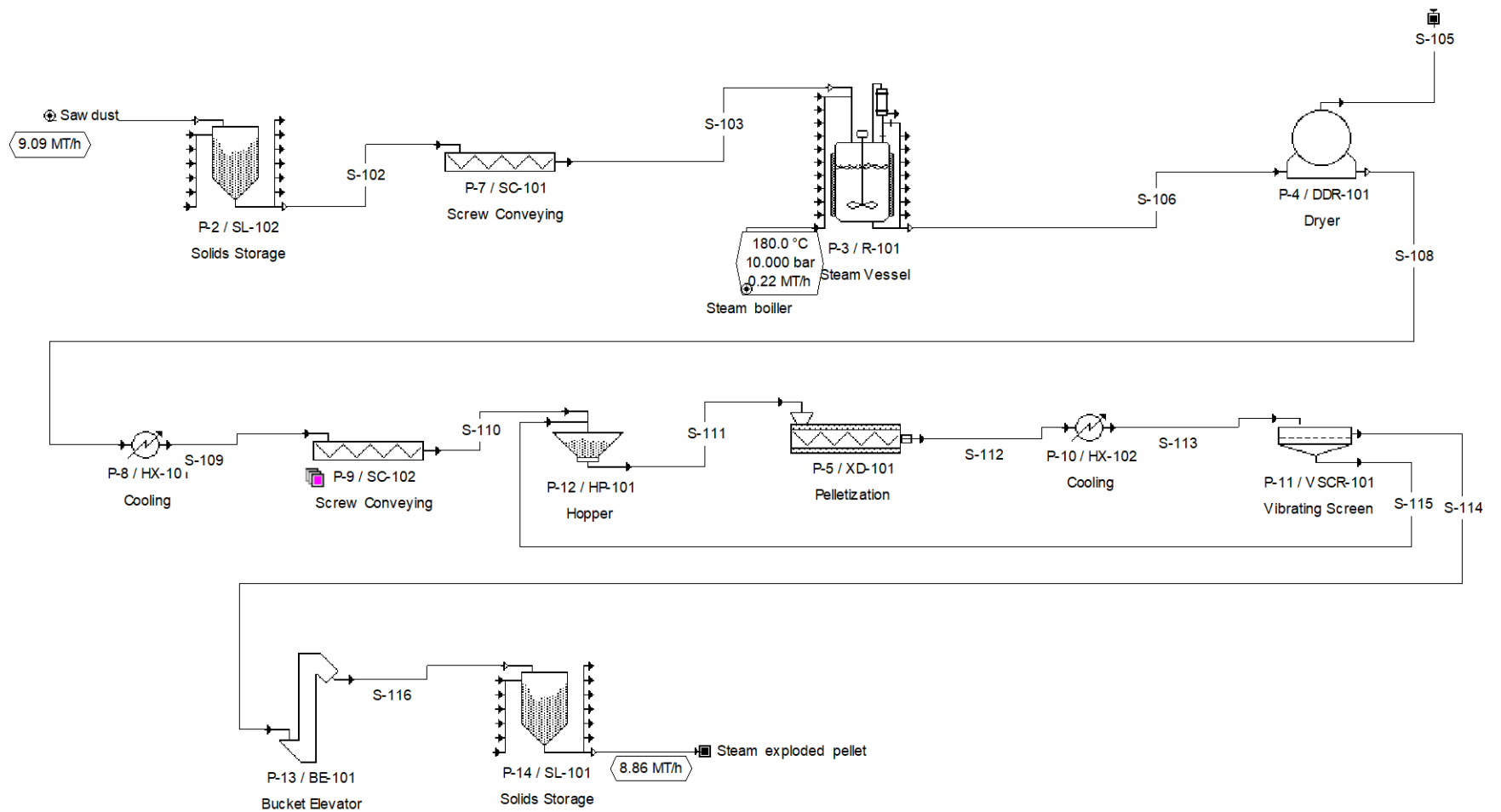


Figure S6.4. Integrated steam exploded treated sawdust pellet plant (Scenario 5).

Table S6.3. Major equipment costs (2023 prices in US\$) for Scenarios 1 and 2.

| Description | Cost (\$) (number of units in parentheses) | |
|--------------------|--|------------------|
| | Scenario 1 | Scenario 2 |
| Silo/bin | 477,000 (4) | 247,000 (2) |
| Grinder | | 154,000 (1) |
| Screw conveyor | 176,000 (2) | 162,000 (2) |
| Belt conveyor | | 177,000 (1) |
| Pelletizer | 212,000 (1) | 212,000 (1) |
| Heat exchanger | 23,000 (1) | 23,000 (1) |
| Bucket elevator | 20,000 (1) | 20,000 (1) |
| Unlisted equipment | 230,000 | 265,000 |
| Total | 1,148,000 | 1,323,000 |

Table S6.4. Major equipment costs (2023 prices in US\$) for Scenarios 3 and 4.

| Description | Cost (\$) (number of units in parentheses) | |
|----------------------|--|------------------|
| | Scenario 3 | Scenario 4 |
| Silo/bin | 477,000 (4) | 338,000 (3) |
| Grinder | | 301,000 (2) |
| Screw conveyor | 374,000 (4) | 254,000 (3) |
| Torrefaction reactor | 3,875,000 (1) | 3,875,000 (1) |
| Condenser | 34,000 (1) | 34,000 (1) |
| Flow splitter | 163,000 (1) | 163,000 (1) |
| Horizontal tank | 48,000 (2) | 24,000 (1) |
| Belt conveyor | | 789,000 (3) |
| Pelletizer | 185,000 (1) | 212,000 (1) |
| Vibrating screen | 33,000 (1) | 38,000 (1) |
| Heat exchanger | 420,000 (8) | 236,000 (4) |
| Bucket elevator | 20,000 (1) | 23,000 (1) |
| Unlisted equipment | 1,410,000 | 1,572,000 |
| Total | 7,048,000 | 7,859,000 |

Table S6.5. Major equipment costs (2023 prices in US\$) for Scenarios 5 and 6.

| Description | Cost (\$) (number of units in parentheses) | |
|--------------------|---|-------------------|
| | Scenario 5 | Scenario 6 |
| Steam generator | 638,000 (1) | 641,000 (1) |
| Dryer | 131,000 (1) | 264,000 (2) |
| Silo/bin | 140,000 (2) | 70,000 (1) |
| Grinder | | 109,000 (1) |
| Screw conveyor | 272,000 (3) | 275,000 (3) |
| Belt conveyor | | 263,000 (3) |
| Pelletizer | 273,000 (1) | 273,000 (1) |
| Vibrating screen | 13,000 (1) | 12,000 (1) |
| Heat exchanger | 210,000 (4) | 363,000 (6) |
| Bucket elevator | 23,000 (1) | 23,000 (1) |
| Unlisted equipment | 425,000 | 535,000 |
| Total | 2,125,000 | 2,675,000 |