



Mitigating Off-Gassing and Self-Heating in Fuel Wood Pellets Storage

A Raw Materials Selection and Pre-Treatment Centred Approach



Workson Siwale

Faculty of Health, Science and Technology

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Abstract

Over the years, wood pellets have become a preferred solid biomass fuel for heat and power generation due to their standardized nature, known properties, and consistent quality. However, pellets produced from fresh sawdust sometimes undergo self-heating and off-gassing, and this poses a challenge for their transportation and storage. The aim of this thesis was to increase the body of knowledge towards understanding the mechanisms underlying the self-heating and off-gassing of wood pellets, and offer solutions for producing wood pellets from freshly generated sawdust, but with reduced self-heating and off-gassing tendencies.

Two separate studies (papers I and II) were conducted to investigate the effects of total wood extractive content and the specific types of extractives present in the raw material on the off-gassing of wood pellets. The results from paper I showed that the total amount of extractives in the raw material has little effect on off-gassing. The concentrations of off-gasses; CO, CO₂ and CH₄ reduced in pellets produced from Scots pine sawdust which had lower extractive contents (stored and acetone extracted). However, increasing the extractive content of the sawdust through the addition of additive oils did not result in increased off-gas emissions. The results of pure cellulose pellets with added additive oils showed that off-gassing is influenced by the type of extractives in the raw material (paper II). The highest concentrations of off-gasses; CO, CO₂ and CH₄ were recorded from pure cellulose pellets with added linseed oil due to their high content of unsaturated fatty acids of 73.9% linolenic and 7.6% linoleic.

The effects of raw material composition and pre-treatments on self-heating and off-gassing of Scots pine wood pellets were also investigated through two separate studies (papers III and IV). The results from both studies indicated significant effects of both raw material composition and drying temperature. There was a strong linear correlation between off-gassing and the sapwood content of the raw material, with correlation coefficient (R) values greater than 0.9 at $p < 0.001$ for all the off-gases (paper III). An increase in sapwood content of fresh Scots pine sawdust led to an increase in off-gassing of CO, CO₂ and CH₄, and O₂ consumption. Storing of sawdust for over six

months prior to pellet production, and increasing the temperature of drying the sawdust resulted in a significant reduction of off-gassing for sapwood pellets. For heartwood pellets, increasing the drying temperature increased the off-gassing while raw material storage had no effect. In the other study (paper IV), the pellets produced from Scots pine mature wood sawdust were more prone to self-heating and off-gassing compared to those produced from juvenile wood sawdust. Steam drying the sawdust at high temperature led to a significant reduction in heat and gas generation for both materials. Furthermore, the study established a notable connection between self-heating and off-gassing, the storage piles with high temperature increase also exhibited high concentrations of off-gases.

The results from this thesis have shown that a biological process, in combination with the chemical oxidation of unsaturated fatty acids lay behind the self-heating and off-gassing of wood pellets. The other notable effect was that methane formation is dependent on anaerobic conditions, whereas formation of carbon oxides can occur both under aerobic and anaerobic conditions. While storing of fresh sawdust for a period of time prior to pellet production remains the most effective method for mitigating the self-heating and off-gassing of wood pellets, sorting and separating the raw materials at source can facilitate the development of storage schedules tailored to specific raw materials, thereby reducing on the raw material storage time.

Keywords: Solid Biofuels, Densified Biomass, Pellet Quality, Heat Generation, Off-gas Emissions, Carbon Oxides, Methane, Fatty and Resin Acids, Safe Pellets

List of Publications

This thesis is a summary of the following appended papers.

Paper I:

W. Siwale, S. Frodeson, J. Berghel, G. Henriksson, M. Finell, M. Arshadi, C. Jonsson, *Influence on off-gassing during storage of Scots pine wood pellets produced from sawdust with different extractive contents*, Biomass and Bioenergy. 156 (2022) 106325.
<https://doi.org/10.1016/j.biombioe.2021.106325>.

Paper II:

W. Siwale, S. Frodeson, M. Finell, M. Arshadi, C. Jonsson, G. Henriksson, J. Berghel, *Understanding Off-Gassing of Biofuel Wood Pellets Using Pellets Produced from Pure Microcrystalline Cellulose with Different Additive Oils*, Energies. 15 (2022) 2281.
<https://doi.org/10.3390/en15062281>.

Paper III:

W. Siwale, S. Frodeson, M. Finell, M. Arshadi, G. Henriksson, J. Berghel, *Influence of Sapwood/Heartwood and Drying Temperature on Off-Gassing of Scots Pine Wood Pellets*, Bioenergy Res. 17 (2024) 479–490. <https://doi.org/10.1007/s12155-023-10668-6>.

Paper IV:

W. Siwale, M. Finell, S. Frodeson, G. Henriksson, J. Berghel, *Fuel Wood Pellets Produced from Sawdust of Scots Pine Mature and Juvenile Wood: Self - Heating and Off - Gassing Tests at Industrial Scale*, BioEnergy Res. (2024). <https://doi.org/10.1007/s12155-024-10736-5>.

Paper V:

W. Siwale, M. Finell, S. Frodeson, *Effects of Mature/Juvenile Wood Composition and Drying Temperature on Scots Pine Wood Pellet Quality Properties*. (Manuscript).

Author's Contributions to the Publications

Paper I: I was involved in planning the study and choosing the appropriate methodology. With the help of my co-authors at Karlstad University, we prepared the raw materials and produced the pellets while off-gassing measurements were done by my co-authors at Swedish University of Agricultural Sciences. I analysed the data and wrote the original draft manuscript.

Paper II: I was involved in planning the study and choosing the appropriate methodology. With the help of my co-authors, I was involved in all data collection stages of raw material preparation, pelletization, off-gassing measurement & fatty and resin acids analysis. I analysed the data and wrote the original draft manuscript.

Paper III: I participated in planning the study and choosing the appropriate methodology. I played a crucial role in every phase of data collection, including raw material preparation, pelletization and off-gassing measurement. I conducted data analysis and wrote the original draft manuscript.

Paper IV: I participated in planning the study and choosing the appropriate methodology. Together with two of my co-authors, I was involved in setting up the industrial experiments to collect self-heating and off-gassing data. I analysed the data and wrote the original draft manuscript.

Paper V: I was involved in planning the study and choosing the appropriate methodology. Together with my co-authors, I contributed to organizing industrial experiments and collecting samples for pellet quality measurement, conducted by my colleagues at the Swedish University of Agricultural Sciences. I also wrote part of the original draft manuscript.

Conferences and Seminars

In addition to the appended publications, results related to this thesis were presented at conferences and seminars listed below.

1. **W. Siwale**, S. Frodeson, M. Finell, G. Henriksson, M. Arshadi, C. Jonsson, J. Berghel, *Effect of Raw Material Sterilization and Contamination on Methane and Carbon Oxides Emissions of Scots Pine Fuel Wood Pellets*. Presented as a conference poster at the World Sustainable Energy Day, 5-8 April 2022, Wels/Austria.
2. **W. Siwale**, S. Frodeson, M. Finell, M. Arshadi, C. Jonsson, J. Berghel, G. Henriksson, Variations in Extractive Content of Scots Pine Sapwood and Heartwood and the Effect on Off-Gassing during Storage of Wood Pellets. Proceedings of the 2023 SWST International Conference, 25 – 30th June 2023, Asheville, North Carolina, USA.
3. Five presentations made at SVINPELS and InnoPels Project Seminars between 2020 and 2023 in Umeå, Karlstad & Gävle, Sweden.

List of Abbreviations

Acronyms & Formulas

AES	Acetone Extracted Sawdust
CH ₄	Methane
CHP	Combined Heat and Power
CO	Carbon monoxide
CO ₂	Carbon dioxide
COC	Coconut Oil plus Cellulose
DP	Degree of Polymerisation
EU	European Union
FPS	Fresh Pine Sawdust
GC-MS	Gas Chromatograph Mass Spectrometry
LOC	Linseed Oil plus Cellulose
LOS	Linseed Oil plus Sawdust
MC	Moisture Content
MCC	Microcrystalline Cellulose
O ₂	Oxygen Gas
PRC	Pine Rosin plus Cellulose
PRS	Pine Rosin plus Sawdust
SPC	Synthetic Pure Cellulose
SPS	Stored Pine Sawdust
TOC	Tall Oil plus Cellulose
TOS	Tall Oil plus Sawdust
VOCs	Volatile Organic Compounds

Units & Symbols

db	Percentage on dry basis
EJ	Exajoule
h	Hours
kg	Kilogram
kg/m ³	Kilogram per Cubic Metre
kWh/kg	Kilowatt-hour per Kilogram
L	Litre
mm	Millimetre
ppm	Parts per Million
TWh	Terawatt-hour
w.b.	Percentage based on wet or as-is condition
wt.	Weight
°C	Degree Celsius
%	Percentage
β	Beta
α	Alpha

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1.0 INTRODUCTION

The global energy supply is still to a great extent, based on fossil fuels; oil, natural gas and coal [1]. This situation has been raising concerns regarding energy availability and sustainability. Challenges such as the depletion of fossil fuel reserves, price volatility of fossil based energy, and environmental impacts such as climate change and global warming [2], emphasize the need to transition towards more sustainable and environmentally friendly energy sources. Wood fuel pellets present a promising solution to address these challenges. They are a renewable energy source and can be produced locally, thus offering a highly price stable alternative compared to conventional energy sources.

Wood fuel pellets are utilized both in small furnaces for residential heating, and on an industrial scale, either as a direct substitute for coal or part of co-fired systems for electricity generation, as well as in combined heat and power (CHP) plants [3,4]. They have emerged as a preferred solid biomass fuel for heat and power generation due to their standardized nature, known properties, and consistent quality. The important pellet quality parameters are moisture, ash and fine contents, mechanical durability, bulk density and pellet size [5]. When compared to other solid biomass fuels, the desirable properties of wood fuel pellets include: high transport and energy storage density, high heating value and low ash and moisture content [3,6]. Additionally, their size and uniformity make them easy to handle and transport, while their low moisture content renders them resistant to degradation by microbiological activities.

This thesis focused on the production and storage of wood pellets, with particular interest on advancing our understanding of the phenomena of self-heating and off-gassing associated with wood pellets. Self-heating and off-gassing refer to the uncontrolled generation and release of heat and gasses by wood pellets and other biomass materials [7–10], and this present challenges during transportation and storage. While there have been a number of research studies on the self-heating and off-gassing of wood pellets, questions still remain regarding the mechanisms underlying the heat and gas generation, as well as the development of sustainable measures for mitigation.

1.1 Wood Pellets Supply

The production and use of wood pellets have been increasing rapidly over the years. Figure 1 shows that the global wood pellets production increased from about 18 million tonnes in 2012 to about 46 million tonnes in 2022 [11]. The European Union is the primary consumer of globally produced wood pellets, representing a significant share of 75% of total wood pellet consumption [4]. However, the demand of wood pellets within the European Union cannot be met by local supply, only slightly over 50% of the 75% consumption share can be produced within Europe (Figure 1). Therefore, huge amounts of wood pellets are imported mainly from North America. Because of this trade situation, the amounts of wood pellets in transportation and storage have increased to ensure continued supply. For instance, between 2010 and 2020, Sweden consistently produced in excess of one million tonnes of wood pellets annually. However, for most years this output typically fell short of the country's annual pellet consumption (Figure 2) [12].

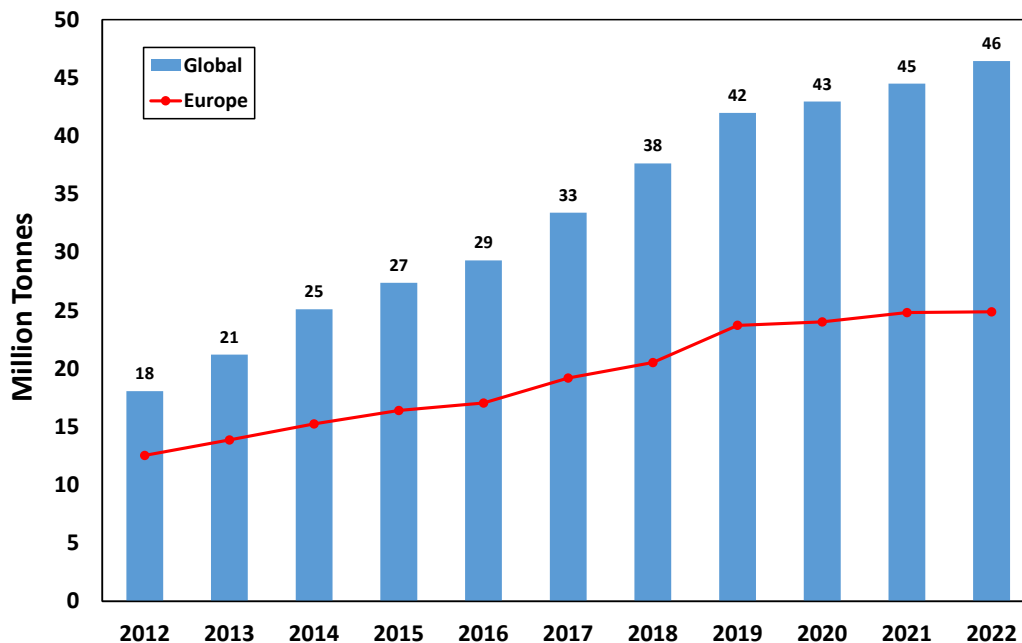


Figure 1. Global and Europe wood pellets production by year. Data obtained from Food and Agriculture Organization (FAO), wood pellet production and trade statistics [11].

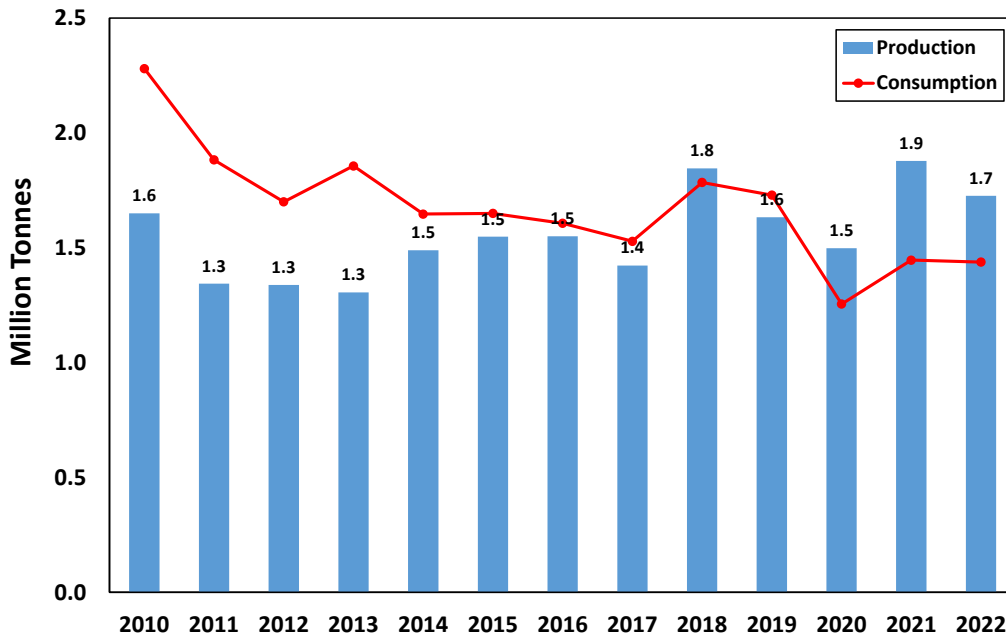


Figure 2. Production and consumption of wood pellets in Sweden by year. Data obtained from the Swedish Pellets Association (PelletsFörbundet) [12].

1.2 Self-heating and Off-gassing of Wood Pellets

Wood pellets sometimes undergo self-heating and off-gassing during storage. The excessive heat accumulation due to self-heating may lead to spontaneous ignition, resulting in fires [8,13,14] while formation and emission of toxic gases such as carbon monoxide and some volatile organic compounds [15–18], is an environmental and human health hazard. According to Larsson et al. [19] eight fire incidents caused by self-heating and spontaneous ignition of wood pellets occurred in different parts of Sweden between 2004 to 2012. There have also been reports from different parts of the world of fatal incidents [15,20,21] attributed to suffocation due to oxygen depletion and increased carbon monoxide from off-gassing. The increased heat due to self-heating can also cause disintegration of the pellets and result in dry matter losses and reduction in quality properties such as bulk density and durability [22].

In a study to determine temperature changes in a wood pellets silo with capacity of 4500 tonnes, temperatures around 65 – 70 °C were reached at the top of the silo over a 7 month storage period [23]. Another study with a similar focus recorded a temperature increase inside a wood

pellets silo from 20 – 57 °C in 10 days [24]. Off-gassing studies of different wood pellets [18,25–27] have shown high concentrations of carbon monoxide, carbon dioxide, methane and volatile organic compounds.

The self-heating and off-gassing of biomass can be caused by either biological processes, chemical oxidation, physical transition reactions (e.g. water sorption) or a combination of these processes [10,28]. Chemical oxidation is believed to be the main contributor to self-heating and off-gassing of stored wood pellets [29–33]. The intensity of heat and gas generated is influenced by various factors including wood species, duration of storage, and pellet quality and quantity [14,34]. Generally, pellets made from freshly processed sawdust are more prone to self-heating and off-gassing compared to those from aged sawdust. Among different species, pellets produced from spruce sawdust tend to be less susceptible to self-heating and off-gassing than those made from pine sawdust [35]. However, regardless of the species, freshly produced pellets emit more gases and heat than those that have been stored for a period [31,36].

1.3 Justification for Research

Previous studies have suggested the oxidation of wood extractives as the primary cause of self-heating and off-gassing in wood pellets [29–33]. However, there is no consensus over the types of extractives involved and the exact oxidation mechanism that generates the heat and gases. Moreover, self-heating is also known to occur in carbonaceous materials like coal and agricultural crops such as maize, wheat, and soybeans, where traditional extractives are absent or present in minimal amounts [37–39]. This suggests that the generation of heat and gas in wood pellets may involve other reaction mechanisms beyond just the oxidation of wood extractives. Therefore, further research is necessary to deepen our understanding of the mechanisms underlying self-heating and off-gassing.

To address the problems of self-heating and off-gassing in wood pellets storage, earlier studies proposed storage and handling guidelines which included recommendations for storage conditions with respect

to air circulation, duration of storage and the size of storage facilities [13,20,36,40]. However, the increase in production volumes, leading to the adoption of larger silos and storage sheds, may pose implementation limitations for these guidelines.

The wood pellets industry currently mitigates self-heating and off-gassing by incorporating a higher proportion of stored sawdust in the raw material blend. The sawdust is typically stored for a duration of 6 to 12 months before utilization. Storing the sawdust serves to reduce the extractive content and enhance the inter-particle binding strength of the resulting pellets [41]. However, the growing demand for wood pellets is making it increasingly impractical to store sawdust for such extended periods. The storage of sawdust is associated with logistical costs, including tying up raw material capital for an extended duration, and losses in total wood weight due to biodegradation. Additionally, in order for big pellet factories to meet annual productions of close to 100 000 tonnes, large storage spaces are required due to the low bulk density of sawdust of about 100 – 200 kg/m³ [42]. Hence, there is need to explore alternative methods for preventing self-heating and off-gassing in wood pellets produced from fresh sawdust.

1.4 Research Objectives

The purpose of this thesis was to contribute to the existing body of knowledge on the self-heating and off-gassing tendencies of wood pellets during storage. It aimed to enhance knowledge on the mechanisms underlying heat and gas generation by examining these phenomena from a production perspective, particularly raw material selection and pre-treatment. The overall goal was to offer solutions for producing wood pellets using freshly generated sawdust, that exhibit decreased tendencies for self-heating and off-gassing, without compromising on pellet quality. The following were the specific research objectives.

1. Produce wood pellets from differently pre-treated raw materials across different production and storage scales, including laboratory and industrial setups. Evaluate the degree of self-

heating and off-gassing exhibited by these pellets, and establish correlations between self-heating and off-gassing tendencies.

2. Enhance the knowledge on understanding the impact of total wood extractive content in the raw material on the off-gassing of wood pellets. The research question for this objective was: To what degree does the total extractive content influence the off-gassing of wood pellets?
3. Enhance the knowledge on understanding the effect of different types of wood extractives on off-gassing of wood pellets. The research question to this objective was: To what extent does the different types of extractives influence the off-gassing of wood pellets?
4. Enhance raw material selection and pre-treatments to mitigate self-heating and off-gassing in wood pellets while evaluating pellet quality properties. The research questions to this objective were:
 - a. Are there differences in off-gassing tendencies between wood pellets produced from sapwood and heartwood sawdust?
 - b. Are there differences in self-heating and off-gassing tendencies between wood pellets produced from mature wood and juvenile wood sawdust?
 - c. How does the drying process (low-temperature or high-temperature drying) in combination with the type of wood raw material influence the self-heating and off-gassing of wood pellets?
 - d. How does the drying process (low-temperature or high-temperature drying) in combination with the type of wood raw material impact the pellet quality properties?

2.0 BIOMASS FOR ENERGY

Bioenergy is a form of renewable energy derived from organic materials, collectively known as biomass. Modern bioenergy comes in solid, liquid and gaseous forms, with solid bioenergy dominating production [43–48]. Biomass refers to organic matter, such as plants, crop residues, and wood, that has stored solar energy through the process of photosynthesis, converting it into chemical energy. It is regarded as a carbon neutral energy source since the carbon dioxide released during processing is absorbed through photosynthesis by the growing trees and plants [49]. Therefore, bioenergy presents environmental advantages of reducing pollutant emissions and maintaining carbon neutrality compared to fossil-based energy.

There are numerous methods for converting biomass into energy or biobased products. Combustion, which involves burning biomass directly to generate heat and/or electricity, is the easiest and most common conversion process. Others include thermochemical conversion processes such as gasification and pyrolysis, and biochemical conversions involve the fermentation/digestion of biomass to produce liquid and gaseous biofuels [50,51]. The choice of processing technology depends on various factors, including the desired energy product (heat, power, or fuels), the available feedstock, and the chemical/physical characteristics of the feedstock. Woody biomass with higher lignin content of 25 – 35% is more solid and denser than agricultural biomass, and this give woody biomass more bioenergy content for heat and power generation through combustion processes [52]. However, in biochemical conversions, lignin inhibits access to cellulose and hemicelluloses, which are sources of sugars necessary for the production of liquid and gaseous biofuels [53].

2.1 Structure and Chemical Composition of Wood

The chemical composition of wood greatly impacts its characteristics, performance, and behaviour across a range of applications, including its utilization for energy [54,55]. The chemical composition of wood is, in turn, affected by the presence, abundance, and distribution of various wood tissues.

2.1.1 Macrostructure of Wood

The primary structural parts of a tree include the stem, tree tops branches, roots, bark, and foliage, collectively forming the biomass of the tree; its accumulated mass both above and below ground level [56]. The macrostructure of wood refers to the overall visible organization and arrangement of wood cells and tissues and this can be described in terms of various features [57]. One prominent feature of wood macrostructure is the presence of growth rings. These rings are concentric circles visible in cross-sections of a tree trunk and are formed as a result of annual growth cycles, each ring represents a year of growth.

2.1.1.1 Sapwood and Heartwood

Sapwood and heartwood are among the essential macrostructural elements in wood. These two wood classifications are easily identifiable in a cross-section of a stem due to their differing colours. Typically, sapwood which is the outer layer of the xylem appears lighter in colour compared to the inner heartwood which is darker [58,59]. However, the sapwood and heartwood of certain species like poplars and spruces exhibit uniform colouring.

Sapwood contains physiologically active and living cells, and performs three primary functions of support, conduction, and storage in a living tree. In softwoods, the tracheids play a dual role, supporting the tree and facilitating the transport of water and minerals from the soil and up. Meanwhile, parenchyma cells store energy rich molecules/nutrition in the form of starch and fats, also serving as a biological defense mechanism by releasing biologically active substances, including antimicrobial toxins [60]. When a tree is felled and the wood dries, the starch and the fat is retained in the wood cells as a permanent deposit. Heartwood on the other hand is the oldest part of the wood and lacks living cells. It has no conduction and storage functions, but only provides structural support. It is also impregnated with antimicrobial molecules, contributing to its natural resistance [61].

The transition of sapwood into heartwood involves changes in the chemical composition of the extractives and primary chemical components of the wood cell walls [60,62,63]. In general, heartwood has a much higher extractive content than sapwood. Across various pine species, the total amounts of extractives were found in the ranges of 2.3 – 8.9% for heartwood, and 0.76 – 3.7% for sapwood [54,64–66]. Additionally, heartwood extractives are more diverse, and they include compounds such as phenolic compounds, resin acids, and free fatty acids and these extractives mainly have as a role to work as antimicrobial agents, whereas sapwood extractives predominantly consist of triglycerides, with a role of nutrient storage [59,67]. This reflects the different biological functions of the two types of wood; sapwood extractives are mainly for energy storage while heartwood extractives prevent microbial decay. There are contradictory conclusions regarding the composition of other wood chemical compounds in sapwood and heartwood [68–70]. Generally, in most softwoods, sapwood contains more cellulose than heartwood, whereas the composition of lignin, hemicelluloses and other non-cellulosic carbohydrates is slightly higher in heartwood compared to sapwood.

2.1.1.2 Juvenile and Mature Wood

The formation of wood in the life cycle of a tree has two phases. During the early stages of growth, immature juvenile wood is produced just after establishment of the cambial zone, and as the tree grows, it undergoes a gradual transition towards producing mature wood [57]. The transition age is estimated to be in the range of 11 – 27 years at breast height with juvenile wood proportion of 15.3 – 47.5% [71]. A young tree predominantly forms juvenile wood, and as it ages, a distinct pattern emerges where mature wood is produced primarily in the lower section of the stem (the butt end), while the upper part of the stem continues to generate juvenile wood [57]. As a result, a tree stem has a consistent core diameter of juvenile wood throughout its length. The proportion of juvenile wood in a tree increases with increasing tree height [72]. Therefore, when trees are felled and bucked, pulpwood, top sawlogs, and thinnings tend to have a much higher content of juvenile wood than butt sawlogs. Additionally, the composition of sapwood and

heartwood varies among these log segments. According to Yang *et al.* [73], the thickness of sapwood in the lower section of a tree stem (up to approximately 10 m) remains relatively consistent and considerably greater than that of the upper section. This suggest that mature wood contains a higher proportion of sapwood compared to juvenile wood.

2.1.2 Wood Chemical Composition

Generally, in terms of chemical composition, woody biomass, which is the main feedstock for pellets production, is a polymeric composite of cellulose, hemicelluloses and lignin. In addition to these, wood comprises other structural elements like starch, pectins, and proteins, along with small quantities of various non-structural, low molecular mass substances such as extractives and inorganic elements [55,74]. The proportions and specific composition of these chemical constituents varies among different wood species (Table 1). Furthermore, within the same tree stem, variations in chemical composition exist between juvenile wood and mature wood, normal wood and reaction wood, as well as sapwood and heartwood. There are even greater variations between different parts of tree; for instance, bark and branch material can have much higher extractive content compared to stem wood. Table 2 provides a comparative overview of the general chemical composition of various tree parts, including stem wood, bark, and forest residues.

Table 1. Example of the chemical composition of some wood species (mass %) [74].

Species	Extractives	Lignin	Cellulose	Glucoman- mannan	Xylan	Other polysacch.	Others
Softwoods							
Norway Spruce (<i>Picea abies</i>)	1.7	27.4	41.7	16.3	8.6	3.4	0.9
Scots Pine (<i>Pinus sylvestris</i>)	3.5	27.7	40	16	8.9	3.6	0.3
Hardwoods							
Birch (<i>Betula verrucosa</i>)	3.2	22	41	2.3	27.5	2.6	1.4
Beech (<i>Fagus sylvatica</i>)	1.2	24.8	39.4	1.3	27.8	4.2	1.3
River red gum (<i>Eucalyptus calimaldulensis</i>)	2.8	31.3	45	3.1	14.1	2	1.7
Red maple (<i>Acer rubrum</i>)	3.2	25.4	42	3.1	22.1	3.7	0.5

Table 2. Average chemical composition of stem wood (inner and outer bark), bark and forest residues (branches and tree tops) [55]. (% based on dry solids)

Component	Stem Wood	Bark ^a	Forest Residues ^a
Cellulose	40 – 45	20 – 30	35 – 40
Hemicelluloses	25 – 35	10 – 15	25 – 30
Lignin	20 – 30	10 – 25	20 – 25
Extractives	3 – 4	5 – 20	~5
Other Organics	~1	5 – 20 ^b	~3
Inorganics	< 0.5	2 – 5	~1

^a Depends greatly on the wood species.

^b Containing mainly suberin (2 - 8%) and polyphenols (2 - 7%) as well as proteins and starch (1 - 5%).

2.1.2.1 Cellulose

Cellulose is the most abundant organic chemical on earth. It is the major chemical component in wood occupying 40 – 50% of the wood dry weight [55]. Wood cellulose is located in the cell wall in association with hemicelluloses and lignin. Chemically, cellulose is a homopolysaccharide made up of only glucose anhydride ($C_6H_{10}O_5$) units. The β -D-glucopyranose units are joined end to end by (1→4) glycosidic bonds in a 4C_1 conformation to form a linear long chain $(C_6H_{10}O_5)_n$ cellulose polymer. The glucopyranose units are 180° towards each other, this makes cellobiose residue, which consists of two adjacent glucopyranose units to be the repeated structural unit of cellulose instead of glucopyranose (Figure 3) [74]. A wood cellulose molecule in its natural state has an average degree of polymerization (DP) in the range of 9 000 – 10 000. Because of the linearity of their structure, individual cellulose molecules are organized within the cell wall in close association with one another to form long strands of microfibrils. The microfibrils contain 60 – 80% highly ordered crystalline regions and the rest is less ordered [56]. The presence of the crystalline structure and the intra and inter-molecular hydrogen bonds on the hydroxyl side groups stiffens cellulose, and this makes it to be more thermally and chemically stable than other wood chemical components. Because of this stability, cellulose is less likely to have an influence on self-heating and off-gassing processes.

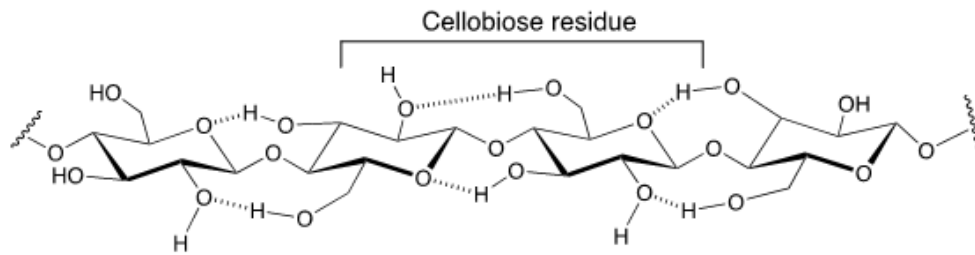


Figure 3. Basic molecular chemical structure of cellulose [74].

2.1.2.2 Hemicelluloses

Hemicelluloses are heteropolysaccharides formed from glucose and other C6 and C5 sugar monomers. They occupy 25 – 35% of wood dry weight. The primary hemicelluloses both in softwoods and hardwoods are the glucomannans and xylans [56,74]. Softwood glucomannans constitute about 15 – 20% of wood dry weight. Their chemical structure is built up of a linear framework of β -D-glucopyranose (β -D-Glcp) and β -D-mannopyranose (β -D-Manp) units, which are branched by α -galactopyranose (α -D-Galp) units. The softwood xylans are made up of a linear framework of β -D-xylopyranose (β -D-Xylp) units, containing branches of 4-O-methyl- α -glucuronic acid (4-O-Me- α -GlcpU) and α -L-arabinofuranose (α -L-Araf) (Figure 4). The xylans in softwoods constitute about 5 – 10% of wood dry weight. In hardwoods, the primary hemicelluloses components are the xylans also referred to glucuronoxylan and constitute 20 – 30% of the dry wood mass. Hardwood glucuronoxylan have a linear framework of β -D-xylopyranose units that contains 4-O-methyl- α -glucuronic acid branches. Hardwood glucomannans have the same linear framework as softwood glucomannans containing β -D-glucopyranose (β -D-Glcp) and β -D-mannopyranose (β -D-Manp) units but it is unbranched (Figure 5). The glucomannans in hardwoods constitute less than 5% of the dry wood mass.

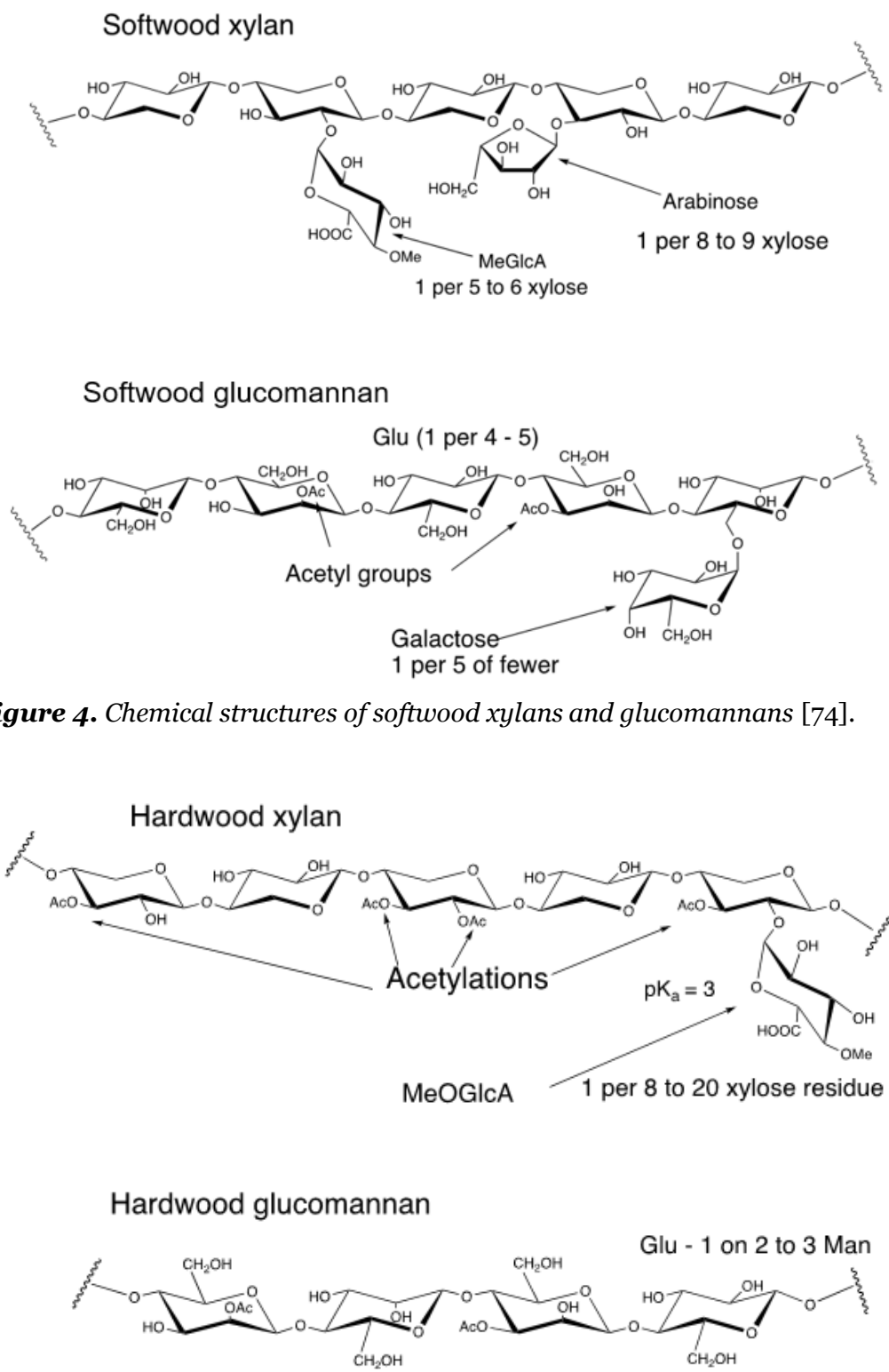


Figure 4. Chemical structures of softwood xylans and glucomannans [74].

Figure 5. Chemical structures of Hardwood xylans and glucomannans [74].

The chemical and thermal stability of hemicelluloses is generally lower than cellulose due to low molecular weight, low degree of

polymerisation (average DP of 100 – 200) and lack of crystallinity. The amorphous structure of the hemicelluloses favours the OH groups to be more reactive when subjected to heat and chemicals, and their saccharide related molecules start to undergo various oxidations even at relatively low temperatures [75]. Pelletization being a thermal process may degrade some hemicelluloses and, in the process, form compounds that may influence self-heating and off-gassing.

2.1.2.3 Lignin

Lignin is an amorphous phenolic heteropolymer with a chemical structure that distinctly differs from the other macromolecular constituents of wood. It is deposited both within the cell wall and in the compound middle lamella, and occupies 20 – 30% of wood dry weight [55]. Lignin biosynthesis occurs through the oxidative radical polymerization of three hydroxy and methoxyl substituted phenylpropane units: sinapyl, coniferyl and *p*-coumaryl alcohols (Figure 6), the so-called lignin precursors. Coniferyl alcohol is the dominant structural building monomer in softwood lignin while equal proportions of sinapyl alcohol and coniferyl alcohol make hardwood lignin. The lignin precursors are joined together by ether (C-O-C) linkages and carbon-carbon (C-C) bonds, and of these linkages the C-O-C dominates with the most prominent type being the β -O-4 structure [74]. Unlike wood carbohydrates, the chemical structure of lignin is irregular in the sense that the monomeric structural elements are not linked in any systematic order. Lignin has a high molecular mass but low degree of polymerization. The molecular mass of softwood and hardwood milled wood lignin is believed to be in the range of 15 000 – 20 000 Dalton while the DP is in the range of 75 – 100 [56].

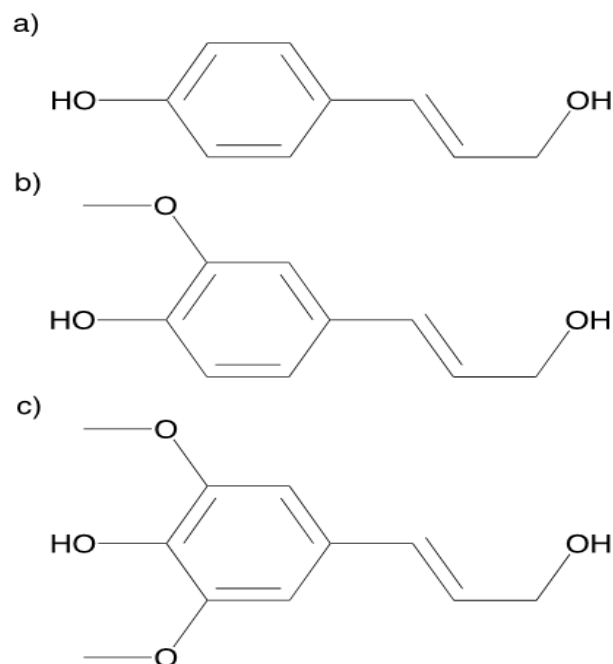


Figure 6. The three precursors of lignin p-coumaryl alcohol a), coniferyl alcohol b), and sinapyl alcohol c) [74].

There are chemical linkages between lignin and the carbohydrates; hemicelluloses and cellulose due to their close association in wood. The covalently bonded aggregates of lignin and the carbohydrates form what is called lignin-carbohydrate complexes (LCC). The carbohydrates linked to lignin in LCC are mainly hemicelluloses; xylans and glucomannans, and the most frequently suggested lignin-hemicelluloses bonds are benzyl ether, benzyl ester and phenyl glycosidic linkages [55,76,77]. The structural function of lignin in wood is to bind the wood cells and to impart rigidity to the cell walls. When it comes to wood pelletization, lignin's thermoplastic properties [78] contribute to inter-particle bonding. As a result, biomass with high lignin content tends to make pellet with high durability.

2.1.2.4 Extractives

Extractives are numerous low molecular mass non-structural organic compounds present in wood that can be extracted using polar and non-polar solvents [79]. They include compounds such as fats, fatty acids, fatty alcohols, phenols, terpenes, steroids, resin acids, rosin, waxes and also more hydrophilic compounds such as sugars and lignans [64]. Table 3 shows the classification of wood extractives into aliphatic

compounds, phenolic compounds and other compounds depending on their chemical character.

Table 3. Classification of organic extractives in wood [55]

Aliphatic Compounds	Phenolic Compounds	Other Compounds
Terpenes and Terpenoids (including Resin Acids and Steroids)	Simple Phenols	Sugars
Esters of Fatty Acids (Fats and Waxes)	Stilbenes	Cyclitols
Fatty Acids	Lignans	Tropolones
Alkanes	Isoflavones	Amino acids
	Flavonoids	Alkaloids
	Condensed Tannins	Coumarins
	Hydrolyzable Tannins	Quinones

Extractives perform a wide range of physiological and defensive functions in trees [74,79]. Extractives such as sugars, starch, fats and waxes are energy sources of the biological functions in the wood cells. Other compounds like terpenes, resin acids, and phenols are responsible for the tree's defence mechanism against biological attack. Although wood extractives cover a wide range of individual chemical compounds, their amount is generally small and varies both in nature and concentration within and between wood species and within the same tree. Total extractive contents of not more than 5% of dry weight of wood have been reported in temperate softwoods and hardwoods while in some tropical hardwoods relatively high amounts of extractives are found [80].

The extractive content can be considerably higher in parts like bark and branches and is normally increased in wounded wood regardless of the tree species [54,64,80]. According to Alen [55], the total extractive contents based on wood dry weight for pine and spruce are in the ranges of 2.5 – 4.5% and 1.0 – 2.0% respectively, and the variations are due to factors like tree age, tree part and growth conditions. A comprehensive study on wood extractives by Nisula [64] showed that pines are very rich in extractives with average concentrations of 2.3 – 8.9%, 0.76 – 3.7% and 0.82 – 30% for heartwood, sapwood and

knots respectively. The corresponding values for spruces were 0.54 – 2.0% heartwood, 0.30 – 1.4% sapwood and 2.6 – 18% knots.

2.1.2.5 Inorganic Materials

Wood contains small amounts of inorganic materials, i.e., the ash content after burning. The ash content of stem wood from the temperate zone is 0.1 – 1.0% of wood dry weight but it is increased during handling and it is much higher in needles, leaves and bark [74]. Agricultural residues such as grasses also contain high levels of inorganics particularly silica [55]. The inorganic materials originate mainly from metal salts such as carbonates, phosphates and silicates deposited in wood cell walls, with calcium and potassium salts being the most common ones. Although some of the inorganic elements are essential for tree growth, ash is unwanted in thermal conversion processes of biomass because it causes ash-related problems like slagging and corrosion. Generally, standardised wood pellets have very low ash content (0.7 – 2.0%) [81] compared to other solid biomass fuels.

2.2 Sustainable Biomass Sources

The widespread availability of biomass globally makes it a potential substitute for a significant portion of fossil fuel usage if well managed. The global production of biomass is estimated to be in the range of 10 to 50 billion dry tonnes annually [82]. Sustainable sources of biomass for energy purposes can be derived from various waste streams, including forestry (logging, thinning, and wood processing residues), agricultural (harvest and processing residues), and significant portions of food and municipal solid wastes [83]. Additionally, dedicated bioenergy crops, such as short rotation trees like poplar (*Populus*), willow (*Salix*), and Eucalypt (*Eucalyptus*) with a growth rotation age of 3 to 15 years, along with non-woody perennial grasses [3], are another viable and sustainable source of biomass for energy production. To ensure the sustainability of biomass sources, it's essential to adopt responsible harvesting and cultivation practices, avoid overexploitation, and consider the overall environmental and social impacts of biomass production.

2.3 Wood Pellets Production and Storage

The utilization of solid biomass for heat and power generation is the leading bioenergy application, primarily due to its cost competitiveness in terms of installation and maintenance compared to fossil fuel alternatives [2]. However, handling raw solid biomasses can present significant difficulties due to their low bulk density and uneven particle size, leading to logistical and technical challenges in transportation, storage, and utilization. Additionally, the high moisture content of biomass renders it prone to mould growth and other forms of biodegradation [3,84]. Biomass densification overcomes all these challenges.

Pelletization is the most prevalent biomass densification technology for solid fuel applications [85–88]. Wood pellets are a densified solid biomass in the form of short cylindrical units with the diameter of 6 – 12 mm and length not exceeding 40 mm [89]. The characteristic features of wood pellets include having low moisture content, high energy density and homogenous size and shape. The European Pellet Council certification standard; ENplus has set the quality thresholds for wood pellets for industrial and non-industrial use [81]. The standard classifies the pellets into three ENplus quality classes; A1, A2 and B (Table 4) and defines quality requirements for different pellet properties.

Table 4. Threshold values of the selected important pellet properties [81]

Property	Unit	ENplus A1	ENplus A2	ENplus B
Diameter	mm	6 ± 1 or 8 ± 1	6 ± 1 or 8 ± 1	6 ± 1 or 8 ± 1
Length	mm	3.15 < L ≤ 40	3.15 < L ≤ 40	3.15 < L ≤ 40
Moisture Content	w-% ^a	≤ 10	≤ 10	≤ 10
Mechanical Durability	w-% ^a	≥ 98	≥ 97.5	≥ 97.5
Bulk density	kg/m ³ ^a	600 ≤ BD ≤ 750	600 ≤ BD ≤ 750	600 ≤ BD ≤ 750
Fines (< 3.15 mm)	w-% ^a	≤ 1	≤ 1	≤ 1
Ash content	w-% ^b	≤ 0.7	≤ 1.2	≤ 2
Net Calorific Value	kWh/kg ^a	≥ 4.6	≥ 4.6	≥ 4.6

^a: as received; ^b: dry mass basis

The raw materials used to produce wood pellets are mainly residues such as, sawdust, shavings and wood dust from wood processing industries, as well as log wood from logging residues, thinnings, pulpwood and short rotation trees [90]. The required process steps in pelletizing these materials differ depending on their particle size and moisture content (Figure 7). The thinnings, pulpwood and short rotation trees are first debarked, chipped and course milled before joining the rest of the production process. Sawdust, shavings and wood dust are the mostly used raw materials for pellet production. Wet sawdust is first dried before fine milling while dry wood shavings and wood dust go straight into fine milling as a first process step. Bark containing materials such as forest wood chips and logging residues cannot be used for the production of ENplus classified non-industrial use wood pellets due to the high ash content of bark. However, they are used to produce industrial wood pellets.

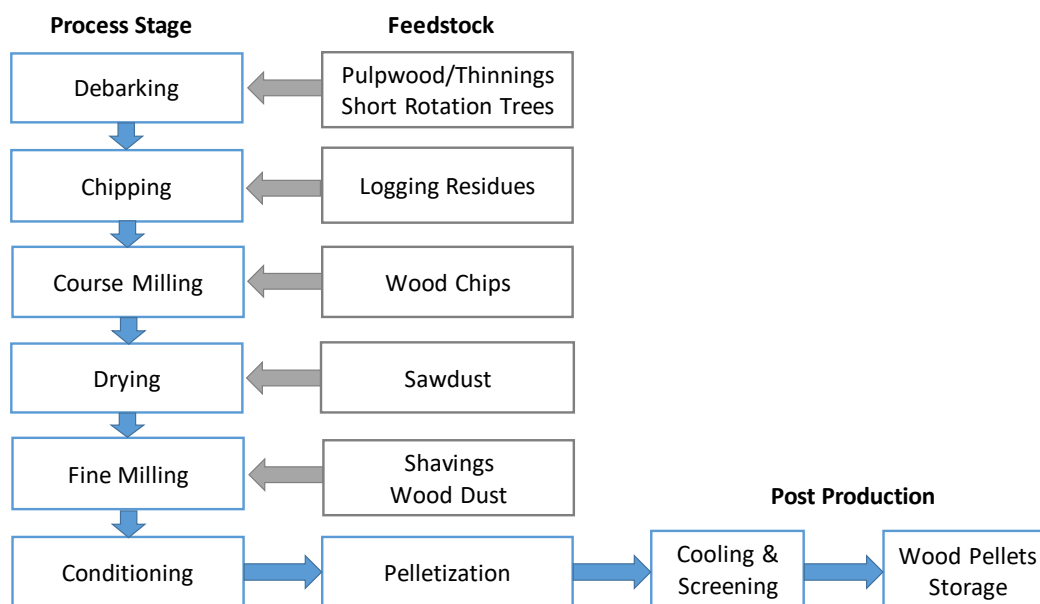


Figure 7. Overview of the wood pelletization process [90].

The densification of the material occurs in the pelletizing process stage. In this stage, conditioned biomass feedstock is compression and extruded through die holes under heat and pressure, resulting in the formation of dense cylindrical pellets. During the entire production process, the moisture content of the raw material is reduced from approximately 50% to around 8 – 12% after drying and conditioning, and further decreases post-pelletization [91]. On the other hand, the

low bulk density of the feedstock is increased from about 40 – 250 kg/m³ depending on feedstock characteristic, to 600 – 800 kg/m³ [42,92].

There are two types of pelletizer systems; flat-die and the ring-die. In a flat die, the material flows into the pelletizer from the top and falls down to a fixed circular perforated disk (die) where two or more roller wheels rotate and force the material through the holes in the die [88] (Figure 8 left). In a ring die, the material is fed in the middle and distributed over the inner surface of the ring die. When the die is rotating, the rollers that are mounted inside the die force the material through the press channels to produce the pellets (Figure 8 right). In some designs, both the die and the rollers rotate.

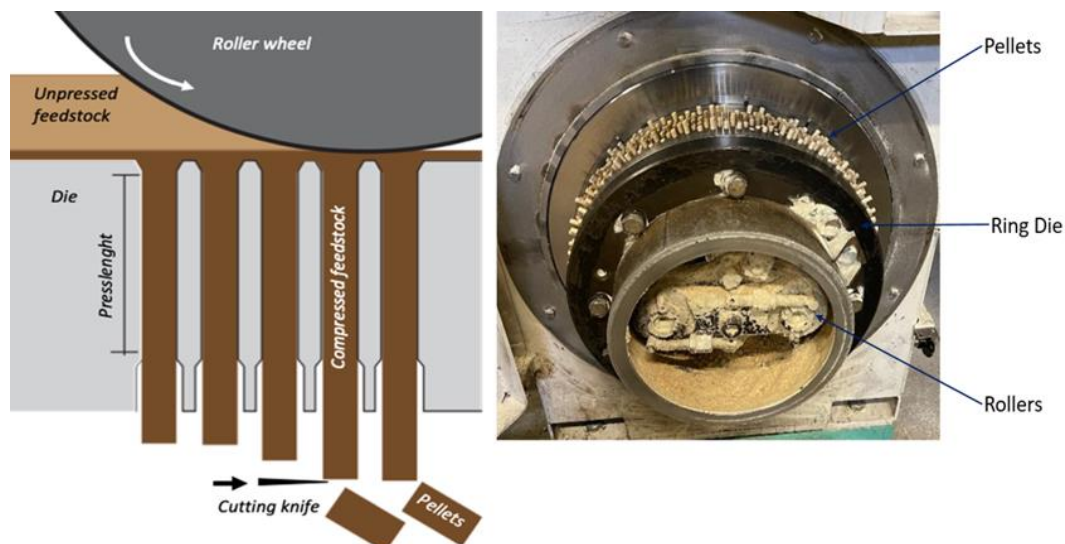


Figure 8. Schematic representation of densification of biomass in a flat die pelletizer - left [88]. A ring die showing the rollers mounted inside the die and pellets protruding out of press channels - right (Photo by author).

The wood pellets produced are stored indoors under dry conditions because of their hydrophilic nature, requiring protection from moisture [93]. Exposure to high humidity could result in pellet disintegration, thus defeating the purpose of increasing the bulk density. Additionally, improper storage increases the risk of self-heating and off-gassing. Wood pellets are initially stored at production facilities before being transported to end users for further storage and eventual use. At these production sites, the pellets are normally stored in huge storage sheds, stack in large piles or packed in big sacks or

small 16 kg bags (Figure 9). Large-scale users such as municipal heating companies and housing cooperatives, store their pellets in large silos, whereas individual households store them in domestic storage rooms.

The duration of pellet storage can vary widely, ranging from a few weeks to a year, depending on seasonal energy demands and international trade dynamics that might affect pellet availability [40]. During periods of low heating demand, such as in the summer, pellets may be stored for longer periods, whereas in winter, when heating demand peaks, storage times are typically shorter.



Figure 9. Pile of wood pellets in a storage shed (left), and stacks of small 16 kg bags of wood pellets (right), at Härjeåns Energi AB pellet factory in Sveg, Sweden. (Photos by author)

2.4 Bioenergy Outlook

The World Energy Outlook 2023 [94] indicates that the overall bioenergy production increased by 5% in 2022, reaching 40 EJ. This accounted for more than half of the global renewable energy share, with North America and Europe collectively contributing over 50%. The increase in renewable energy adoption across Europe is largely attributed to policy initiatives spearheaded by the European Commission, such as the Renewable Energy Directive legal framework. The prevailing directive, (EU) 2018/2001 sets a binding target of at least 32% for the overall share of energy sourced from renewable sources in the European Union's gross energy consumption by 2030 [95]. Additionally, EU industrial policies are oriented towards fostering a sustainable circular bioeconomy, encompassing innovative

technologies and value-added products derived from biomass. These policies are aligned with the objectives of the United Nations Sustainable Development Goals [96], particularly Goal 7, which aims to ensure access to affordable, reliable, sustainable, and modern energy; as well as Goal 13, which focuses on addressing climate change and its impacts.

As of 2022, renewable energy accounted for 23% of the EU's energy consumption, with Sweden leading at 66% share [97]. Driven by its climate policy framework that aspires to achieve zero net greenhouse gas emissions by 2045 [98], Sweden utilises a lot of bioenergy. The long-term strategies are to contribute towards fulfilling the Paris Agreement of reducing anthropogenic greenhouse gas emissions and enhance removals by sinks, and to promote increased carbon sequestration. The nation aspires to evolve into a low-carbon society, particularly aiming to eliminate reliance on oil in the transportation sector by 2030. As a result, the country has experienced a substantial shift in its national energy system, favouring renewable energy, particularly bioenergy [99]. According to statistics from the Swedish Energy Agency, the contribution of biomass to Sweden's energy mix has seen a consistent increase since the 1970s. In 2010, biofuels constituted 135 TWh of the total nation's energy supply, compared to 97 TWh in 2000, representing an increase of over 214% since 1970 [100,101]. By 2020, out of a total energy supply of 508 TWh, 141 TWh came from biofuels [102]. Although this represented a slight decrease from the previous year, primarily attributed to the impact of COVID-19 pandemic, which led to reduced energy consumption, especially in the transportation and industrial sectors. This significant proportion of renewable energy stems from the extensive use of solid biofuels such as wood pellets in the industrial sector and district heating, alongside the use of bioethanol, biodiesel, and biogas in the transportation sector.

3.0 MATERIALS AND METHODS

This section provides an overview of the research design and describes in detail the primary materials and methods employed in the five papers comprising this thesis.

3.1 Research Design

This research employed an exploratory experimental design to investigate how factors like raw material type and pre-treatments influence the self-heating, off-gassing, and quality properties of wood pellets. The experimental designs depicted in Figures 10 – 13 were customized to align with the individual study's specific objectives.

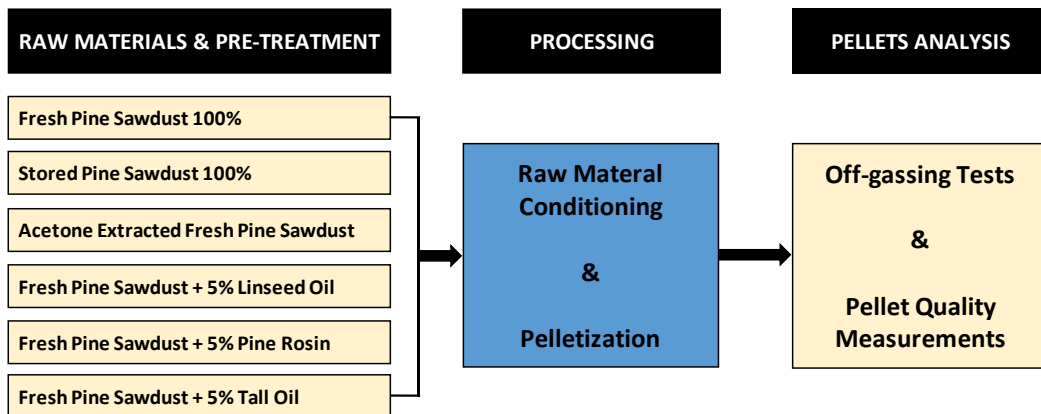


Figure 10. Experimental design of the study for paper I showing the raw materials and pre-treatments used to produce the wood pellets.

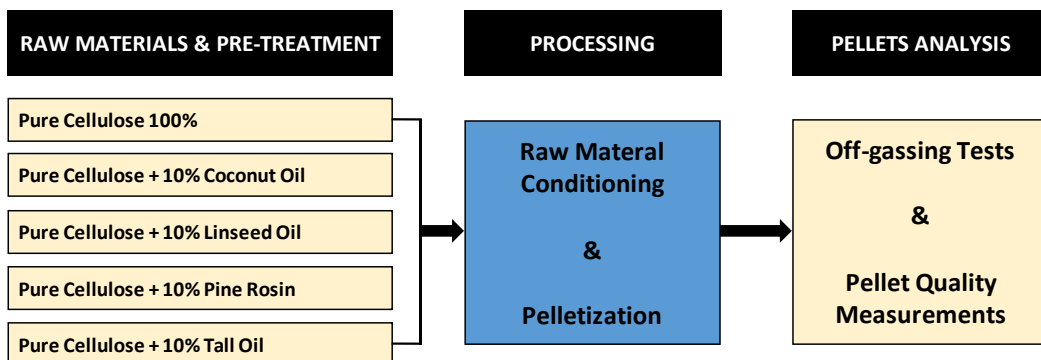


Figure 11. Experimental design of the study for paper II showing the raw materials and pre-treatments used to produce the pellets.

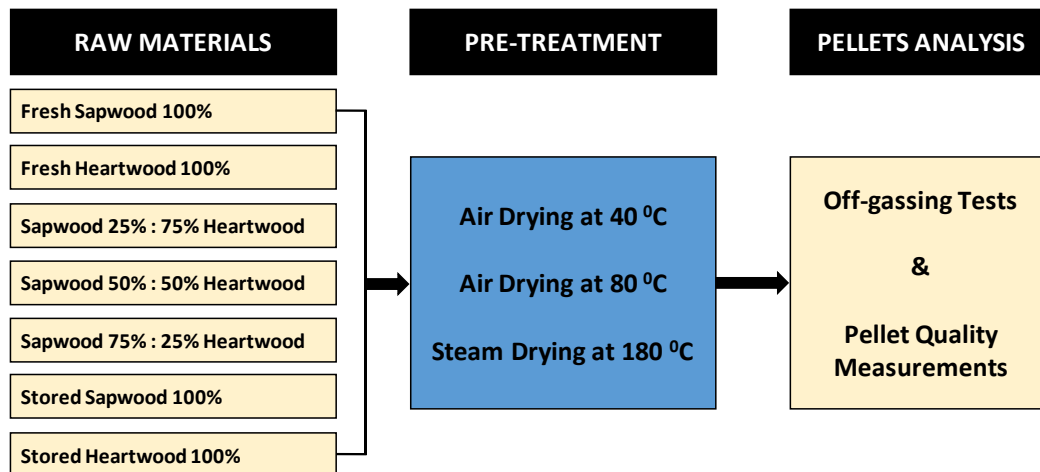


Figure 12. Experimental design of the study for paper III showing the raw materials and pre-treatments used to produce the wood pellets.

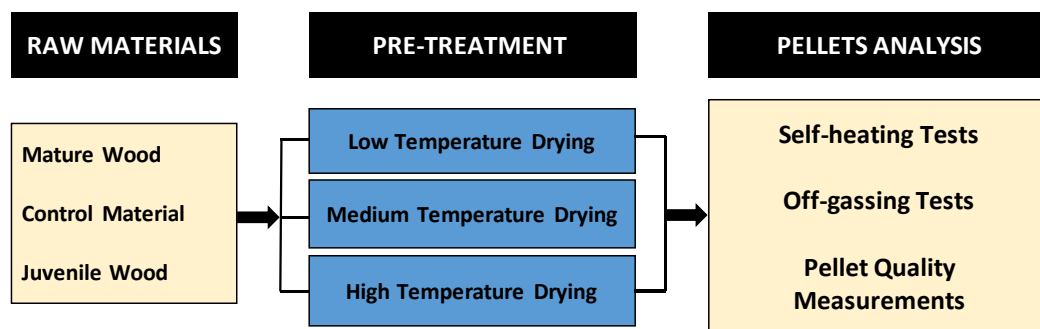


Figure 13. Experimental design of the study for papers IV & V showing the raw materials and pre-treatments used to produce the wood pellets.

3.2 Materials and Preparation

The materials used included Scots pine sawdust, synthetic pure cellulose powder, and different additive oils. Pre-treatments of the raw materials varied and were adapted to the specific objectives of the studies reported in individual appended papers. For the laboratory experiments in papers I, II, and III, about 20 – 25 kg was prepared for each type of the raw material. The target was to have enough material to produce about 15 – 20 kg of wood pellets required for off-gassing analysis.

3.2.1 Sawdust Preparation – Paper I

The sawdust used in paper I was freshly produced Scots pine sawdust sourced from Laxå pellets in central Sweden. At delivery, the sawdust

had a moisture content of 54% w.b. (percentage based on wet or as-is condition). The sawdust was air dried in a bed drying kiln at 40 °C to about 8.5% w.b. moisture content, sieved through a 5 mm sieve and kept in sealed plastic bags until use. The sawdust was pre-treated to generate six different types of raw materials as follows:

1. Fresh pine sawdust without any treatment (FPS)
2. Stored pine sawdust (SPS)
3. Fresh sawdust plus pine rosin (PRS)
4. Fresh sawdust plus linseed oil (LOS)
5. Fresh sawdust plus tall oil (TOS)
6. Acetone extracted fresh sawdust (AES).

Fresh pine sawdust (FPS) was prepared by just conditioning dried sawdust to about 12% w.b. moisture content prior to pelletization. Stored pine sawdust (SPS) was obtained by storing wet fresh pine sawdust under standard room conditions for 9 months, thereafter dried and conditioned to 12% w.b. moisture content. For the three sawdust materials involving added extractives (PRS, LOS & TOS), 5% db (percentage on dry basis) of linseed oil, tall oil, and pine rosin were individually added to dried fresh pine sawdust and the resulting mixtures conditioned 12% w.b. moisture content

To prepare acetone extracted sawdust (AES), special fabricated 30 L capped containers were used for the extraction process (Figure 14). Fresh sawdust was extracted in 3 kg batches, using 25 L of acetone, over a 72-hour period at room temperature. This extraction method was adopted from [103] but modified to accommodate larger quantities of sawdust. In this modified procedure, 3 kg of sawdust was placed in a 30 L capped container, and 20 L of acetone was added. The mixture was allowed to stand for 48 hours with occasional stirring at 12-hour intervals. After the initial 48 hours, 5 L of the solvent was filtered and drained through the valve at the bottom of the container, and a fresh 5 L of acetone was introduced. The mixture was left to stand for an additional 24 hours, after which all the solvent was filtered and drained off. The remaining extracted sawdust residue was then rinsed with 25 L of water to neutralise any remaining acetone. All filtrates from the extraction process were kept in sealed containers for safe disposal.



Figure 14. Capped containers used for extraction (a) and extracted pine sawdust (b). The containers are fitted with a valve at the bottom for easy draining of the solvent. (Photos by author)

3.2.2 Sawdust Preparation – Paper III

The raw material used in paper III consisted of fresh and stored Scots pine (*Pinus sylvestris*) sapwood and heartwood sawdust. This was prepared from freshly cut sawn timber sourced from Rinns Såg AB Sawmill located in Torsby, Sweden. Sapwood sideboards measuring 25 x 100 x 1500 mm, were cut from the outer part of the sawlogs, while heartwood centre pieces, measuring 38 x 125 x 1500 mm, were cut from the inner part of the same sawlogs. To prepare the sawdust, the sawn timbers were initially ripped into wood strips using a bench power-driven ripping saw and the wood strips further chipped into smaller particles using a garden chipper. The resulting wood chips were then dried and milled through a 6 mm sieve to produce the sawdust (Figure 15). For the stored materials, undried wood chips were stored indoors in an open-air environment at room temperature for seven months (Figure 16) and thereafter dried and milled.



Figure 15. Preparation of sapwood and heartwood sawdust from pieces of sawn timber. (Photos by author)



Figure 16. Piles of sapwood and heartwood chips stored in an open-air environment at room temperature. (Photo by author)

3.2.3 Sawdust Preparation – Paper IV & V

Four different types of sawdust raw materials were used in papers IV and V. These included two control sawdust materials and two test materials consisting of freshly generated Scots pine (*Pinus sylvestris*) sawdust, obtained from two different specialised sawmills belonging to Bergkvist Siljan Skog AB. The sawmills are located in Mora (61.00490 N, 14.53700 E) and Blyberg (61.15120 N, 14.18150 E) in Sweden.

The test materials comprised of Mora sawdust, sourced from a sawmill that processes large diameter butt/middle sawlogs with a top diameter exceeding 22 cm under bark, and Blyberg sawdust, obtained from a

sawmill that handles small diameter top sawlogs with a bottom diameter less than 21.9 cm under bark. Mora sawdust contains a high proportion of mature wood, while Blyberg sawdust has a high proportion of juvenile wood due to the variations in positions and diameters of sawlogs from which the sawdust is generated [72]. A total of about 2000 tonnes of fresh sawdust was delivered to two pellet factories and processed into wood pellets within a month of delivery.

The control sawdust materials comprised of the regular sawdust blend employed at two different pellet factories: Solör Bioenergi Pellets AB in Älvdalen and Härjeåns Energi AB in Sveg, Sweden. The Solör Bioenergi control sawdust was composed of a volume blend ratio of 1:2:3, incorporating sawdust recovered from process shutdowns, fresh sawdust, and stored sawdust, respectively. On the other hand, the Härjeåns Energi control sawdust had a volume blend ratio of 1:2, comprising fresh and stored sawdust respectively.

3.2.4 Pure Microcrystalline Cellulose – Paper II

The cellulose used in paper II was synthetic pharmaceutical grade pure microcrystalline cellulose (MCC) powder (bp/mcc/avicel PH 101 / PH 102) purchased from Xi'an Geekee Biotech Co., Ltd from China. Microcrystalline cellulose is a purified, partially depolymerized cellulose powder synthesized by acid-hydrolysis of alpha cellulose (I β -allomorph) precursor [104]. Industrially, MCC is manufacture from wood as well as non-wood lignocellulosics. It has relatively low chemical reactivity and excellent compactibility at low pressures [105], making it amenable to compression processes such as pelletization. The synthetic pure cellulose powder was pre-treated to generate five different types of raw materials as follows:

1. Cellulose powder 100% without additives (SPC)
2. Cellulose powder plus 10% coconut oil (COC)
3. Cellulose powder plus 10% pine rosin (PRC)
4. Cellulose powder plus 10% linseed oil (LOC)
5. Cellulose powder plus 10% tall oil (TOC).

The decision to add 10% of additive oils was made due to the fact there was no other inherent extractives in cellulose powder. The synthetic

pure cellulose without any additive oil (SPC) was prepared by just conditioning cellulose powder to about 5% (w.b.) moisture content, from an initial moisture content of about 1.6%. The 5% moisture content was arrived at after carrying out a number of trial productions to ascertain the pelletability of synthetic pure cellulose powder. For the four raw materials with added additive oils (COC, PRC, LOC & TOC), 10% (based on total dry solids) of melted coconut oil, milled solid pine rosin and liquid linseed and tall oils were added to their respective weighed batches of synthetic pure cellulose powder and the mixtures conditioned to 5% (w.b.) moisture content.

3.2.5 Additive Oils

Four chemically distinct additive oils; coconut oil, pine rosin, linseed oil and tall oil were used in studies involving Scots pine sawdust (paper I) and synthetic pure cellulose powder (paper II). The selection of these additive oils was deliberate, aiming to represent various types of extractives, mainly fatty and resin acids found in wood. In the pure cellulose study, all four additive oils were used, whereas in the Scots pine study, three of these oils were used, excluding coconut oil.

3.2.5.1 Coconut Oil

Coconut oil was selected for its high content of saturated fatty acids. Coconut oil is a colourless to pale brownish yellow plant oil derived from kernels, meat, and milk of coconut plant and fruits. Generally, coconut oil is composed of about 90% saturated fatty acids and 10% unsaturated fatty acids [106]. Kung Markatta's virgin coconut oil was used. The oil is cold pressed from the coconuts and its consistency is solid at room temperature but becomes liquid when heated. The product was purchased from a local supermarket in Karlstad, Sweden.

3.2.5.2 Rosin

Rosin was selected for its high content of resin acids. Rosins are non-volatile exudates of resins obtained from resinous tree species, primarily pines [107]. Rosin is a mixture of mainly resin acids with a little amount of diterpenoid alcohols and aldehydes. The resin acid

content are mostly abietic type (abietic, levopimaric, palustric, neoabietic and dehydroabietic acids) and less pimaric type (pimaric, sandaracopimaric and isopimaric acids) [79]. Dry exudates of gum rosin obtained from the local pine forests was used.

3.2.5.3 Linseed Oil

Linseed oil was selected for its high content of unsaturated fatty acids. Linseed oil is a colourless to yellowish oil obtained from seeds of the flax plant (*Linum usitatissimum*). It is produced by pressing and/or solvent extraction of the linseed. The oil consists mainly of the esters of glycerol and five fatty acids of which three are unsaturated C18 oleic, linolenic and linoleic while two are saturated C16 palmitic and C18 stearic acids [108,109]. About 90% of the total fatty acids are unsaturated with only 10% being saturated [110]. Ready-made Biltema branded linseed oil sourced locally in Karlstad; Sweden was used.

3.2.5.4 Tall Oil

Tall oil was selected for a balanced mixture of fatty and resin acids. Tall oil is a by-product of the Kraft pulping process of softwoods. It is obtained as one of the major fractions from the distillation of crude tall oil; a viscous and sticky dark brown liquid which is a co-product of the Kraft pulping process [83]. In terms of composition, tall oil is a mixture of several components which include resin acids and other terpenoids (38 – 53 wt.%), fatty acids and triglyceride oils (38 – 53 wt.%), unsaponified (neutral) compounds (6.5 – 20 wt.%), water (0.5 – 3.0 wt.%) and ash (0.1 – 1.0 wt.%) [111].

3.3 Methods

Papers I, II, and III used the same pelletization method and off-gassing measurement technique at the laboratory scale, whereas papers IV and V employed a different setup at the industrial scale.

3.3.1 Pelletization – Lab Scale

The pellets for the laboratory experiments (papers I, II & III) were produced using an Amandus Kahl pellet press (Model 14-175) (Figure 17). This production unit is located at the Environmental and Energy Systems section at Karlstad University, Sweden. The Amandus Kahl Model 14-175 pellet press has a flat die system, power of 3 kW and 10 – 50 kg/h production capacity. Dies with channel diameter of 6 mm and press channel lengths of 15 mm and 24 mm were used. The dies were pre-heated in an oven at 103 °C for 24 hours and thereafter made to run with trial raw materials until the die temperature increased and stabilised in range of 70 – 100 °C, which is the required pelletization temperature.

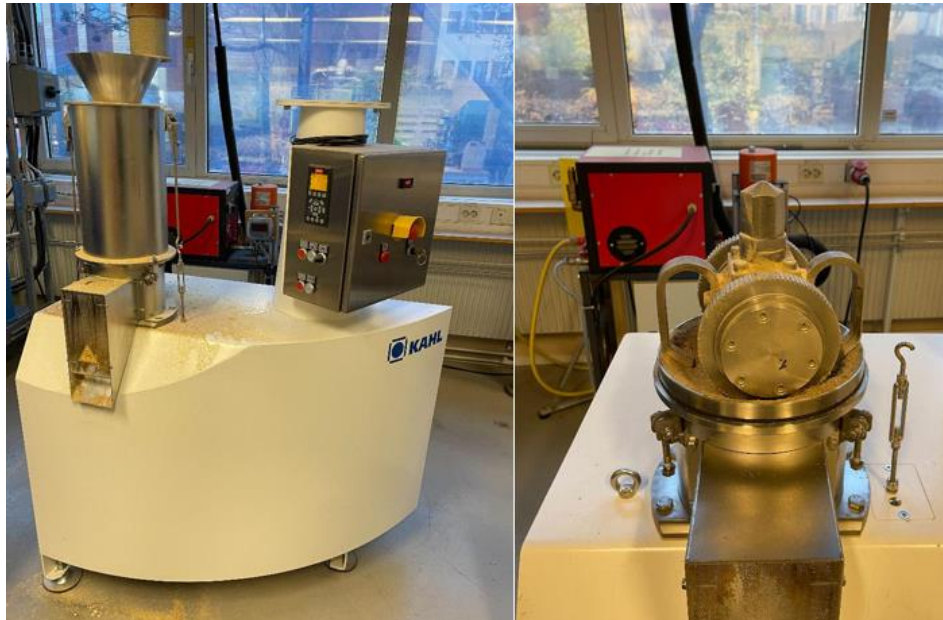


Figure 17. Amandus Kahl pellet press Model 14-175 in the pellet lab for Environmental and Energy Systems section at Karlstad University. (Photos by author)

3.3.2 Drying and Pelletization – Industrial Scale

The pellets for the industrial experiments were produced using three different processes distinguished by drying methods. These three drying methods included:

- A. *Low-temperature drying in a belt dryer (air inlet 78 °C, sawdust outlet 67 °C).* This was done at Solör Bioenergi Pellets AB in Älvdalen, Sweden. The method involved air drying wet sawdust of about 50% moisture content (MC) to the required MC in a belt dryer. The process involves placing sawdust onto a horizontally moving flat belt, while hot air is directed through the material from above.
- B. *Medium-temperature drying (air inlet 78 °C, sawdust outlet 35 °C; steam inlet 198 °C, sawdust outlet 66 °C).* This was a hybrid system of belt and drum dryers, and was also done at Solör Bioenergi Pellets AB in Älvdalen, Sweden. In this method, wet sawdust of about 50% MC was first air dried in a belt dryer to about 29% MC, and then subsequently dried to the required MC in a drum dryer. The drum is equipped with heat exchange tubes, which are heated by steam produced in an auxiliary boiler. During the drying process, sawdust is exposed to the heated surfaces, allowing the moisture within the material to evaporate.
- C. *High-temperature drying in a flash dryer (constant steam drying at 140 - 170 °C).* This was done at Härjeåns Energi AB in Sveg, Sweden. The method involved drying wet sawdust of about 50% MC to the required MC using pressurised superheated steam in a flash dryer. The dryer is composed of tubular heat exchangers. Sawdust is introduced in a closed drying system where it directly interacts with pressurized superheated steam and is conveyed through the dryer. The superheated steam acts both as a source of heat and drying medium, it evaporates and takes away the moisture in wet sawdust which becomes part of the steam. The dried sawdust and steam are separated in a cyclone and the steam is recirculated.

The drying processes were adjusted to produce three moisture levels with the target of producing pellets with varying moisture contents, as follows: high moisture content of 9%, medium moisture content of 6%, and low moisture content of 3%. At Härjeåns Energi, 0.3% of starch is routinely added to the raw material during conditioning, and this was

adopted. The pellets were produced using industrial ring die pellet presses of which the specific press conditions for each pellet mill are detailed in Table 5.

Table 5. Pellet press conditions at the two factories used to produce the wood pellets for the tests.

Pellet Factory	Material	Channel Diameter	Channel Length	Production Rate ^a	Temperature ^b
Solör Bioenergi	Control	8 mm	90 mm	3.2 t/h	98 - 126 °C
	Blyberg				95 - 140 °C
	Mora				98 - 130 °C
Härjeåns Energi	Control	8 mm	70 mm	2.9 t/h	95 - 122 °C
	Blyberg				88 - 122 °C
	Mora				87 - 121 °C

^a Average production rate/pellet press throughout the period the pellets for all the materials were being produced.

^b Temperature of the pellets upon discharge from the press, the range indicates variations in raw material moisture content, with higher temperatures being for low MC and lower temperatures for high MC.

Mora had a high proportion of mature wood, *Blyberg* had a high proportion of juvenile wood, *Control* had a high proportion of stored sawdust.

3.3.3 Off-gassing Measurement – Container Storage

The off-gassing measurement for the laboratory experiments involved storing the pellets in 18.1 L airtight plexiglass containers for papers I & II (Figure 18), and 20 L airtight polyethylene containers for paper III. Before use, the interiors of the containers were cleaned with distilled water, sterilised with ethanol and dried. The containers were filled with the pellets measuring 7 – 8 kg, corresponding to about 70% of the container's volume capacity.

The concentrations of CO, CO₂, CH₄, and residual oxygen in a gas sample were measured using a multi-instrument based on electrochemical and infrared (IR) sensors (ECOM J2KN Pro-IN gas analyser, Palgo AB, Sweden). Measurements were taken by introducing the analyser probe into the container through the sampling port positioned on the top of the container. The sampling port had an air lock valve to prevent gas loss during measurement. After insertion, the

probe was allowed to equilibrate inside the container for one to two minutes before readings were taken. When taking out the probe from the container, there was a time of about 2 seconds before the valve was closed that allowed fresh air into the container. This compensated for the volume of gas extracted by the analyser (1.9 – 2.4 L/min according to the display) and also ensured pressure equalization by allowing external air into the container. The measurements were done every 24 hours throughout the storage period, and two measurements; each from a separate container were taken for each and every category of wood pellets.



Figure 18. Wood pellets in plexiglass containers and ECOM J2KN Pro-IN gas analyser used for measuring the concentrations of CO, CO₂, CH₄ and residual oxygen. (Photo by author)

3.3.4 Self-heating and Off-gassing Measurement – Piles Storage

The self-heating and off-gassing industrial experiments (paper IV) were conducted in storage sheds at production sites (Figure 19). A total of 36 storage piles of wood pellets, each contained about 7.2 tonnes of wood pellets and measured approximately 1.5 m in height were created. The temperature inside the piles was measured using temperature data loggers (Tinytag Transit 2 – TG-4080) positioned in the middle of the piles at heights of 50 cm, 75 cm and 100 cm from the floor. The loggers

were programmed to record temperatures every one hour for the entire storage duration using Intab EasyView 11 software. Temperature changes inside the piles were computed by subtracting the initial temperature from the temperature recorded at each measurement time point, and the maximum temperature increase was taken as a measure of self-heating.



Figure 19. Piles of wood pellets for the experiments in storage sheds at Härjeåns Energi AB in Sveg (left) and at Solör Bioenergi Pellets AB in Älvdalen (right). (Photos by author)



Figure 20. A perforated 5 L polyethylene cylindrical tube positioned within a pile before being covered with wood pellets. (Photo by author)

In addition to self-heating, off-gassing from selected piles was also measured. This was achieved by inserting a perforated 5 L polyethylene cylindrical tube centrally into the pile to capture any emitted gases (Figure 20). The tube was equipped with a long pipe extending outside the pile and featured an airlock valve at its end, which served as a sampling port. Gas concentrations of CO, CO₂, and CH₄ in the tube were measured using a multi-instrument based on electrochemical and infrared (IR) sensors (ECOM J2KN Pro-IN gas analyser, Palgo AB, Sweden), following the same procedure described in section 3.3.3 above. The gas measurements were taken every 48 hours throughout the storage period.

3.3.5 Extractives and Fatty & Resin Acids

The sawdust in papers I and III was analysed for total extractives content. The total hydrophilic and lipophilic extractives were determined by separately conducting water and organic solvents extractions, in a Soxhlet apparatus. To determine the water-soluble extractives, 25 cycles of extraction were carried out for each sample, using water as the solvent, while for lipophilic extractives, a solvent mixture of petroleum ether and acetone, in a 90:10, v/v ratio was used, and the extraction was run for 12 cycles. For each material, about 3 g of sawdust samples were extracted in triplicate.

The lipophilic extracts in paper III and the additive oils in paper II were analysed for fatty and resin acids composition. This was done using a Hewlett Packard (HP6890-5973) GC/MS instrument. The instrument has an auto-sampler operating in the electron impact mode (EI 70 eV) and is equipped with a 30 m by 0.25 mm HP-5 ms capillary column coated with cross-linked (5%-phenyl)-methylpolysiloxane. The column temperature was programmed as follows: 190 °C at 12 °C/min, and then 290 °C at 5 °C/min and held for 2 minutes. Helium was the carrier gas at a constant flow rate of 1 ml/min. Full scan EI mass spectra (m/z 35 - 500) were recorded after injecting 1 µl of the samples and the peaks were identified by the NIST Mass Spectral Search Program (version 2.0). An internal standard heptadecanoic acid of known concentration was used for quantitative analysis. The relative concentrations of the identified fatty and resin acids were determined

by comparing the peak areas in each chromatogram with that of the internal standard. The fatty and resin acids determination method is described in detail in Arshadi and Gref [18].

3.3.6 Quality Properties

Important pellet quality properties including moisture content, bulk density and durability were determined. The measurements were done according to the standard methods, specified and detailed in European (EN) standard methods of determination for solid biofuels, namely EN 14774-1:2009 for moisture content, EN 15103:2010 for bulk density, and EN 15210-1:2009 for mechanical durability [81]. Moisture content determination involved the drying of at least 300 g of pellet samples in an oven at 105 ± 2 °C until a constant weight was obtained. Bulk density was determined using a standardised cylindrical container of 5 L volume. The container was tarred, filled with pellets to the brim and subjected to two impact falls from a 15 cm height. Thereafter, the tarred container together with the pellets was weighed and the bulk density calculated using the volume of the container. Durability testing was done by tumbling 500 ± 10 g of sieved pellets in a tumbling box device at 50 ± 2 rpm for 500 rotations. The resulting abraded and finely broken particles were sieved through a 3.15 mm sieve, and the remaining pellets from the initial 500 ± 10 g were weighed. Durability was calculated as a percentage of the final pellet weight relative to the initial weight.

4.0 SUMMARY OF RESULTS

This section summarises the main results from the five papers included in this thesis. Each summary provides a brief overview and highlights key results. Detailed and comprehensive results can be found in the appended full papers.

4.1 Paper I

The objective of this paper was to investigate how the quantity of extractives in sawdust raw material influences the off-gassing of Scots pine wood pellets.

The results of the linear regression analysis showed low correlations between the concentrations of off-gases (CO, CO₂, and CH₄) and the total extractive content of the raw materials, with the coefficients of determination (R²) being below 0.5 for all three off-gases (Figure 21).

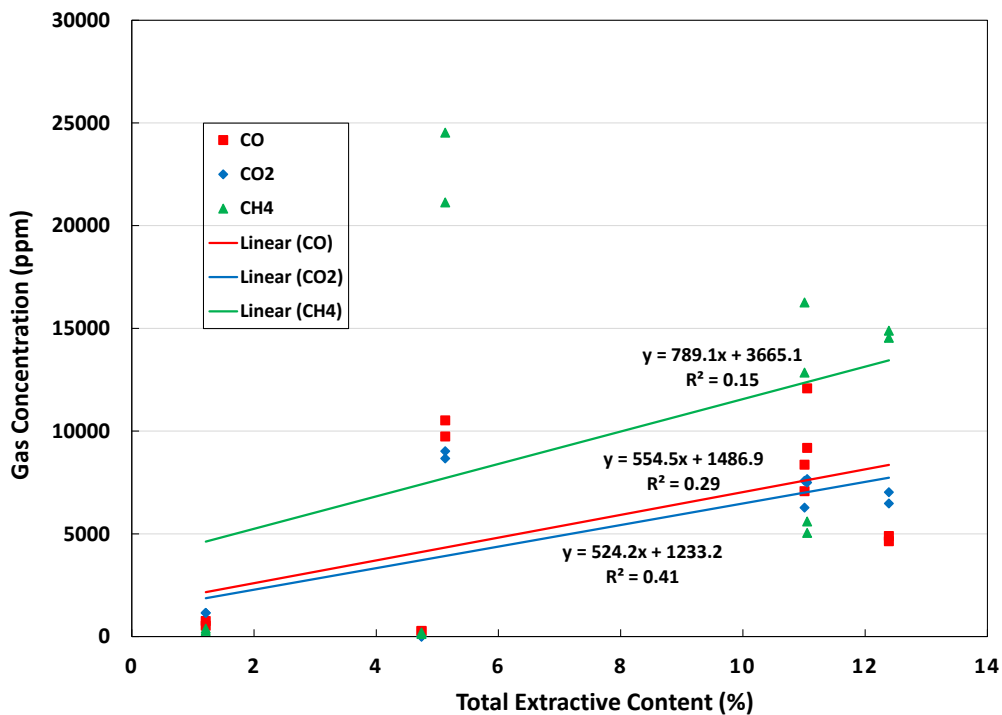


Figure 21. Linear relations and coefficients of determination between the final gas concentrations of CO, CO₂ and CH₄ in the off-gassing containers and the total amount of extractives in the sawdust raw material.

The wood pellets with added tall oil and fresh pine pellets had the highest cumulative concentrations of CO, measuring 10 630 ppm and 10 132 ppm respectively, while acetone extracted pellets (644 ppm) and stored pine pellets (272 ppm) recorded the lowest (Figure 22). Stored pine pellets did not generate any CO₂, while fresh pine pellets had the highest CO₂ concentration at 8 850 ppm, with acetone extracted pellets having the lowest at 1 155 ppm. Regarding CH₄, fresh pine pellets recorded the highest concentration (22 832 ppm), while acetone extracted and stored pine pellets had the lowest concentrations at 163 ppm and 333 ppm respectively (Figure 23). The residual O₂ was consumed in all the pellet types except for stored pine and acetone extracted pellets.

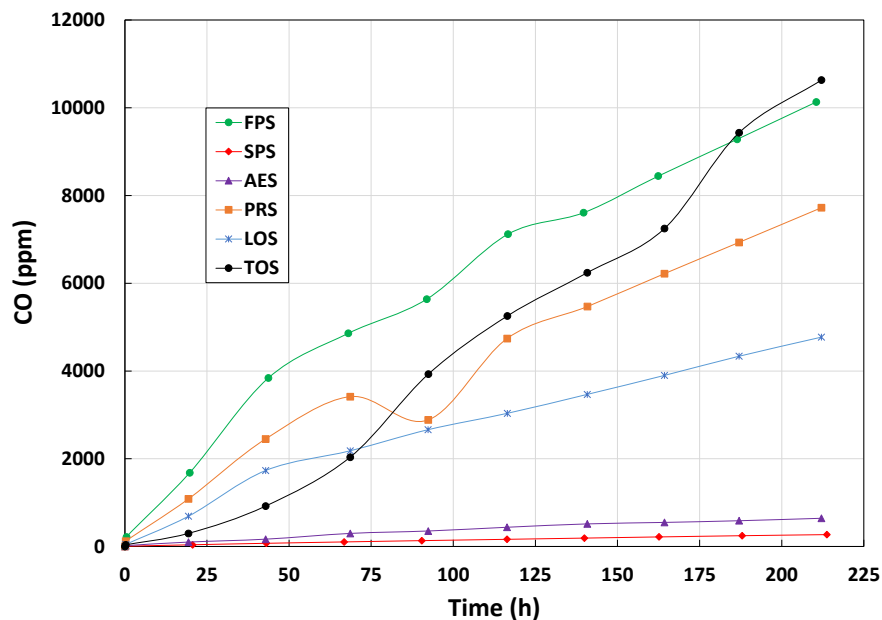


Figure 22. Cumulative mean concentrations of carbon monoxide (CO) in the containers for each of the six wood pellet types. Wood pellets made from: FPS – fresh pine sawdust, SPS – stored pine sawdust, AES – acetone extracted fresh pine sawdust, PRS – fresh pine sawdust with added pine rosin, LOS – fresh pine sawdust with added linseed oil, TOS – fresh pine sawdust with added tall oil.

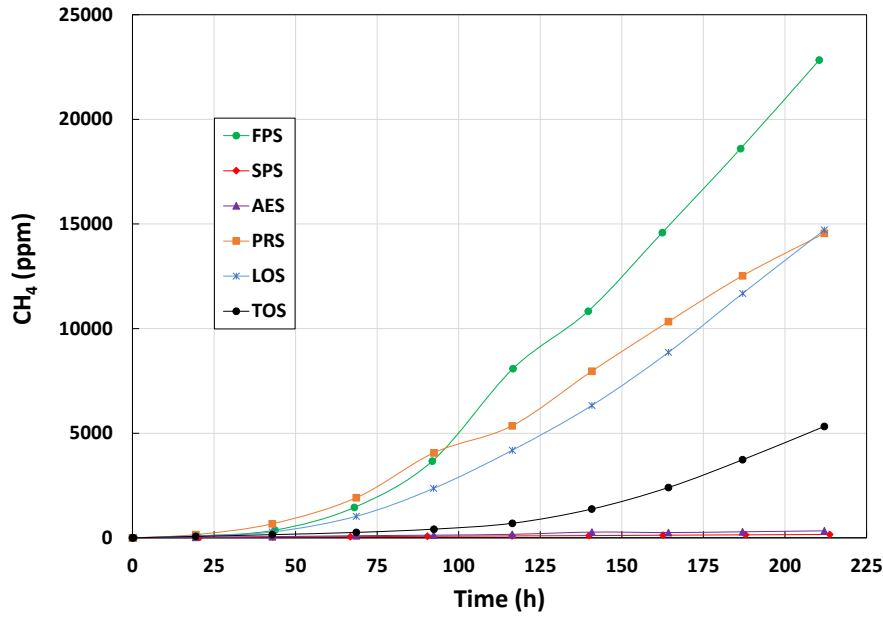


Figure 23. Cumulative mean concentrations of methane (CH_4) in the containers for each of the six wood pellet types. Wood pellets made from: FPS – fresh pine sawdust, SPS – stored pine sawdust, AES – acetone extracted fresh pine sawdust, PRS – fresh pine sawdust with added pine rosin, LOS – fresh pine sawdust with added linseed oil, TOS – fresh pine sawdust with added tall oil.

The concentrations of all the three off-gases significantly reduced for the pellets produced from stored pine sawdust and acetone extracted sawdust, compared to those produced from fresh pine sawdust and with added extractives. This showed that producing wood pellets from extractive free sawdust can substantially reduce the pellets' off-gassing propensity.

4.2 Paper II

The objective of this paper was to investigate the effect of different types of extractives on off-gassing. The four additive oils which were added to pure cellulose powder were analysed for fatty and resin acids content and the results of the identified fatty and resin acids are presented in Table 6.

The cellulose pellets with added linseed oil (LOC) had the highest cumulative concentrations of all the three off-gases: CO, CO₂, and CH₄ (refer to Figure 24 and Figure 25 for CO and CH₄ concentrations respectively). With the exception of pellets with added linseed oil, no

CO₂ was generated during the entire storage period for the other four pellet types, while methane and carbon monoxide were generated in all the five pellet samples. Pellets with added pine rosin (PRC) and tall oil (TOC) generated substantial amounts of carbon monoxide, measuring at 182 ppm and 33 ppm respectively, while the concentrations of carbon monoxide for both synthetic pure cellulose (SPC) pellets and pellets with added coconut oil (COC) remained below 10 ppm. Apart from pellets with added linseed oil, which had complete consumption of residual O₂ (Figure 26), the level of residual oxygen remained approximately at 21% for the other pellet types.

Table 6. Fatty and resin acids identified in the four additive oils by GC-MS. They are presented as a percentage of the total fatty and resin acids content for each of the additive oils.

No.	Compounds		Percentage Composition (%)			
	IUPAC Name	Common Name	Linseed	Tall Oil	Rosin	Coconut
1	Dodecanoic acid	Lauric acid	-	-	-	45.3
2	Tetradecanoic acid	Myristic acid	-	-	-	24.2
3	Hexadecanoic acid	Palmitic acid	7.6	3.4	-	30.6
4	9,12,15 linolenic acid	Linolenic acid	73.9	35.5	-	-
5	9,12 octadecanoic acid	Linoleic acid	7.6	0.8	-	-
6	Octadecanoic acid	Stearic acid	10.9	37.1	-	-
7	9-octadecanoic acid	Oleic acid	-	9.8	-	-
8	Resin acids	Pimaric acid	-	2.1	0.3	-
9		Isopimaric acid	-	6.0	2.0	-
10		Abietic acid	-	5.3	60.2	-
11		Dehydroabietic acid	-	-	37.4	-

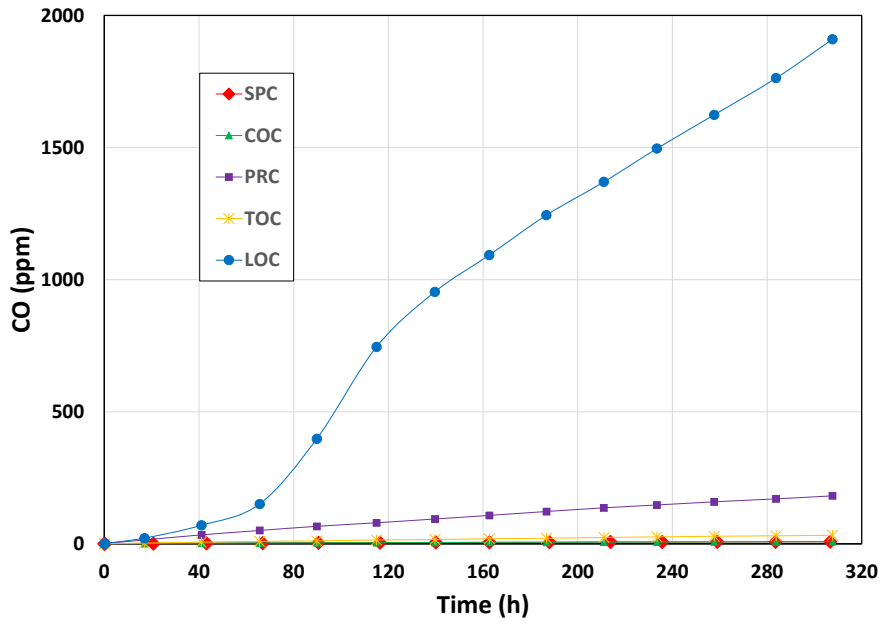


Figure 24. Cumulative mean concentrations of carbon monoxide (CO) in the containers for each of the five cellulose pellet types. Cellulose pellets made from: SPC – 100% synthetic pure cellulose, COC – cellulose plus 10% coconut oil, PRC – cellulose plus 10% pine rosin, LOC – cellulose plus 10% linseed oil & TOC – cellulose plus 10% tall oil.

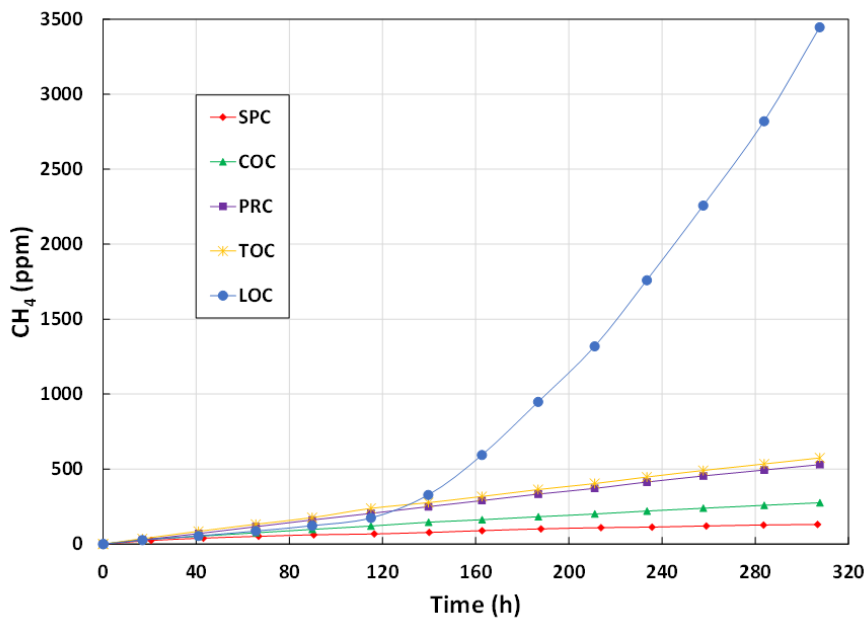


Figure 25. Cumulative mean concentrations of methane (CH₄) in the containers for each of the five cellulose pellet types. Cellulose pellets made from: SPC – 100% synthetic pure cellulose, COC – cellulose plus 10% coconut oil, PRC – cellulose plus 10% pine rosin, LOC – cellulose plus 10% linseed oil & TOC – cellulose plus 10% tall oil.

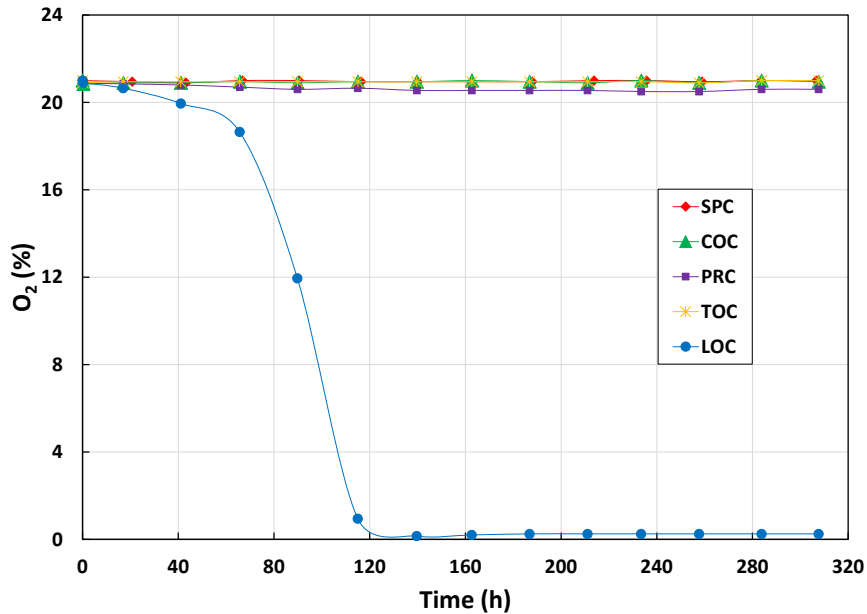


Figure 26. Residual oxygen (O_2) in the containers for each of the five cellulose pellet types. Cellulose pellets made from: SPC – 100% synthetic pure cellulose, COC – cellulose plus 10% coconut oil, PRC – cellulose plus 10% pine rosin, LOC – cellulose plus 10% linseed oil & TOC – cellulose plus 10% tall oil.

The results from this paper showed that the oxidation of unsaturated fatty acids and other triglyceride oils, which are chemically unstable, significantly contributes to the off-gassing of wood pellets. The pellets with added linseed oil had high off-gas emissions due to their high content of unsaturated fatty acids compared to the other pellets.

4.3 Paper III

The objective of this paper was to study the off-gassing tendencies of Scots pine wood pellets made from separated sapwood and heartwood sawdust. The study also investigated the effects of drying temperature, raw material storage and varying proportions of sapwood and heartwood on off-gassing of the pellets.

There was a strong linear relationship between off-gassing and the proportion of sapwood/heartwood, with correlation coefficient (R) values greater than 0.9 at $p < 0.001$ for all the off-gases (see Figure 27). An increase in sapwood content resulted in a significant rise in the off-gassing of CO (Figure 28 a), CO₂ and CH₄, along with a reduction in residual O₂ (Figure 28 b). The raw material drying temperature also

significantly influenced the off-gassing of both sapwood ($F_{(8, 26)} = 51.32, p < 0.05$) and heartwood ($F_{(8, 26)} = 334.1, p < 0.05$) pellets. Increasing the drying temperature for heartwood resulted in increased off-gassing, whereas for sapwood, the off-gassing decreased (Figure 29). Storing of sapwood raw material before pelletization reduced the off-gassing of wood pellets, whereas for heartwood, it had no significant impact. The final cumulative concentrations of CO, CO₂, and CH₄ respectively decreased from 15 988 ppm, 14 453 ppm, and 9 544 ppm in pellets made from fresh sapwood sawdust to 2 620 ppm, 1 598 ppm, and 498 ppm in pellets made from stored sapwood sawdust.

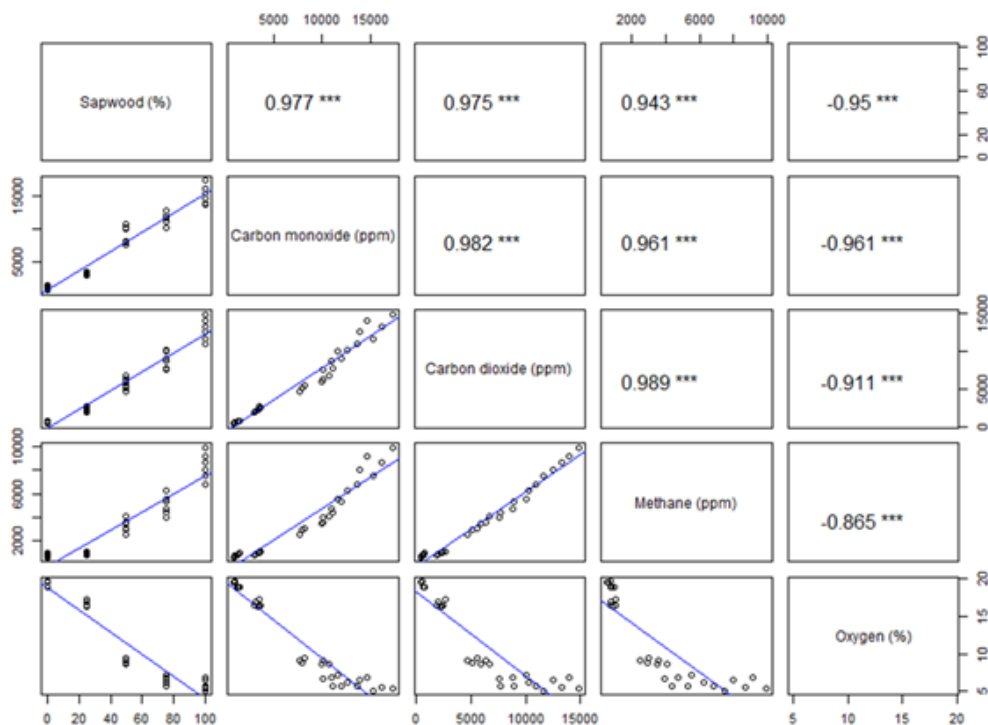


Figure 27. Correlations between the percentage sapwood content of the feedstock, and concentrations of off-gases CO, CO₂ and CH₄ and residual O₂. The scatterplots with regression lines are shown in the boxes of the lower panel, and the corresponding correlation coefficient (*R*) values in the boxes of the upper panel. The three stars following each value indicate significance at $p < 0.001$.

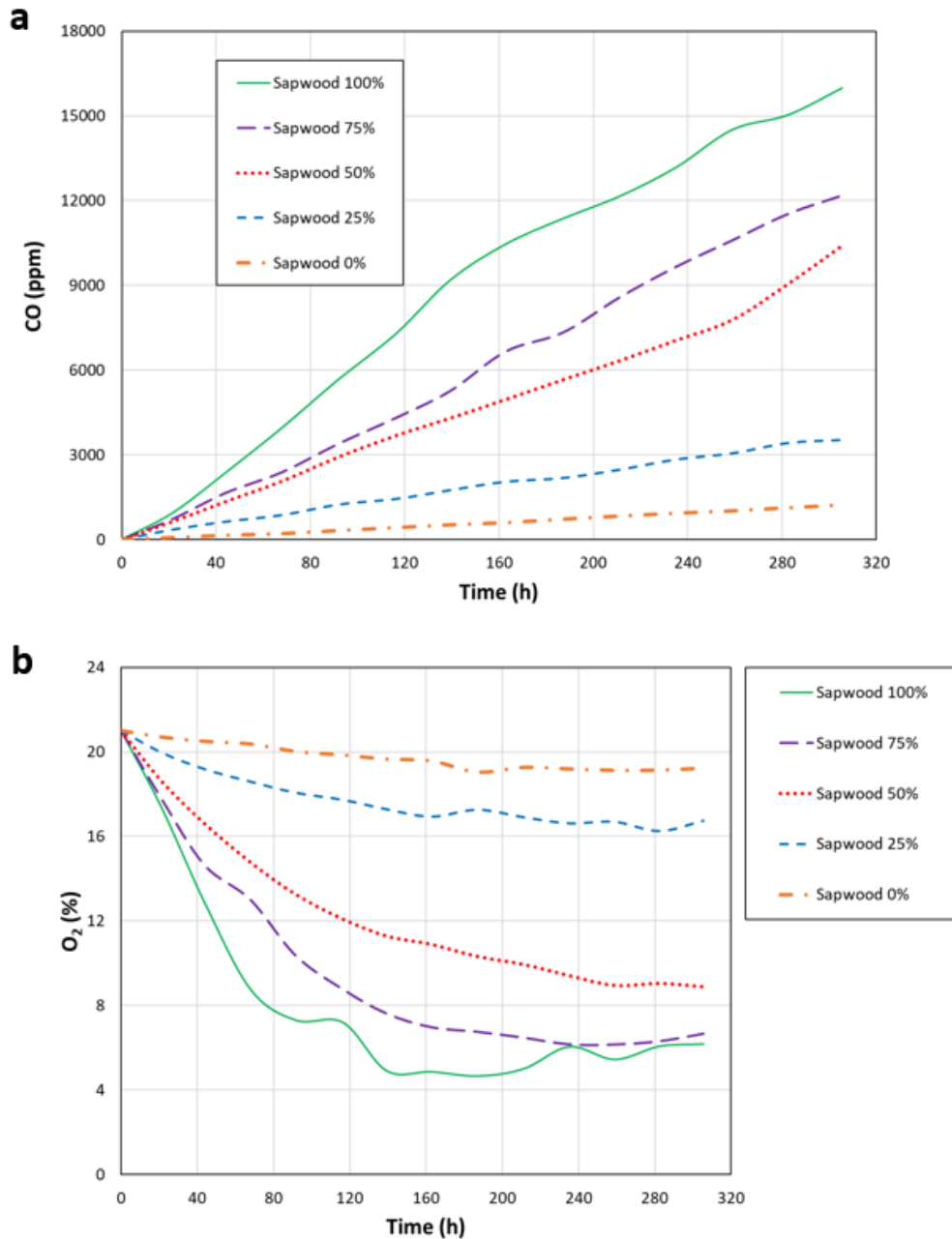


Figure 28. Cumulative mean concentrations of CO (a), and residual O₂ (b) in the containers at each time point during the 13-day storage period for Scots pine wood pellets produced from sawdust with varying (0%, 25%, 50%, 75% & 100%) mass percentage content of sapwood. The same trend was obtained for CO₂ and CH₄.

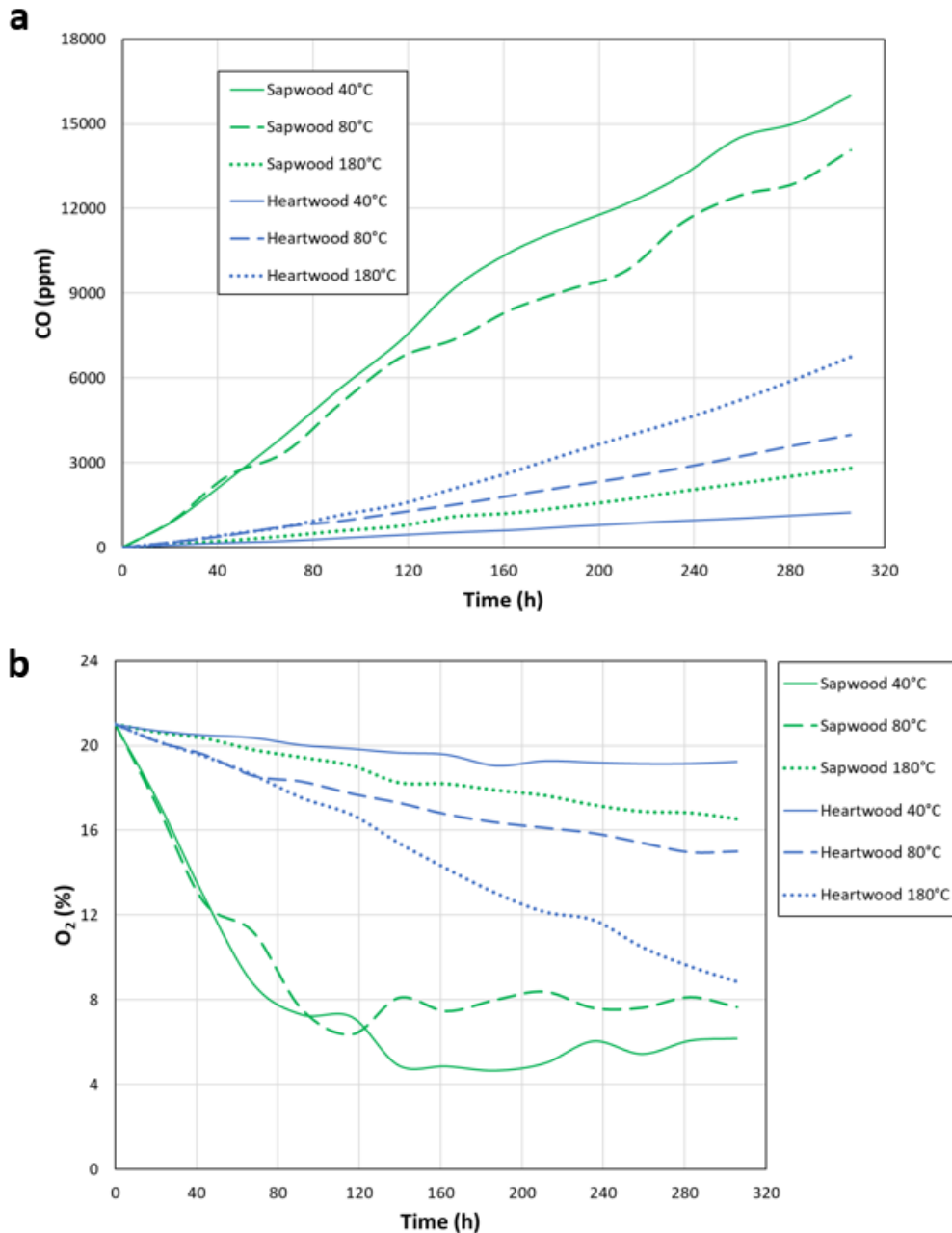


Figure 29. Cumulative mean concentrations of CO (a), and residual O₂ (b) in the containers at each time point during the 13-day storage period for Scots pine wood pellets produced from fresh sapwood (100%) and heartwood (100%) sawdust dried at different temperatures and conditions. The same trend was obtained for CO₂ and CH₄.

The results from this paper provided strong evidence of a connection between sapwood/heartwood content and off-gassing of Scots pine wood pellets. Additionally, it highlighted that raw material storage and drying temperature and conditions can play a crucial role in reducing

the off-gassing of wood pellets but their effectiveness largely depends on the composition of the feedstock.

4.4 Paper IV

The objective of this paper was to study the self-heating and off-gassing tendencies of Scots pine wood pellets made from sawdust generated from mature and juvenile wood. Additionally, the study also investigated the effects of raw material drying method and pellet moisture content on self-heating and off-gassing of the pellets.

There was a connection between self-heating and off-gassing of the pellets, as depicted in Figure 30. The results showed significant effects for both drying method ($F_{(2)} = 65.38, p < 0.05$) and type of raw material ($F_{(6)} = 61.97, p < 0.05$) (see Figure 31). Pairwise comparisons of the mean maximum temperature increase within piles for wood pellets produced using the same drying method but different raw materials showed significant differences for all paired materials under the low-temperature and medium-temperature drying methods ($p < 0.05$), while there was no significant difference for those under high-temperature drying. There were also correlations between the combined effects of the type of raw material and the moisture content of the pellets and temperature increase. The strongest correlations were observed for pellets produced using low-temperature drying ($F_{(3, 14)} = 83.52, \text{Multiple } R^2 = 0.95, p < 0.05$) (Figure 32 a) and medium-temperature drying ($F_{(3, 13)} = 62.05, \text{Multiple } R^2 = 0.93, p < 0.05$) (Figure 32 b), while the pellets produced with high-temperature drying ($F_{(3, 14)} = 8.63, \text{Multiple } R^2 = 0.65, p = 0.002$) had the lowest correlation.

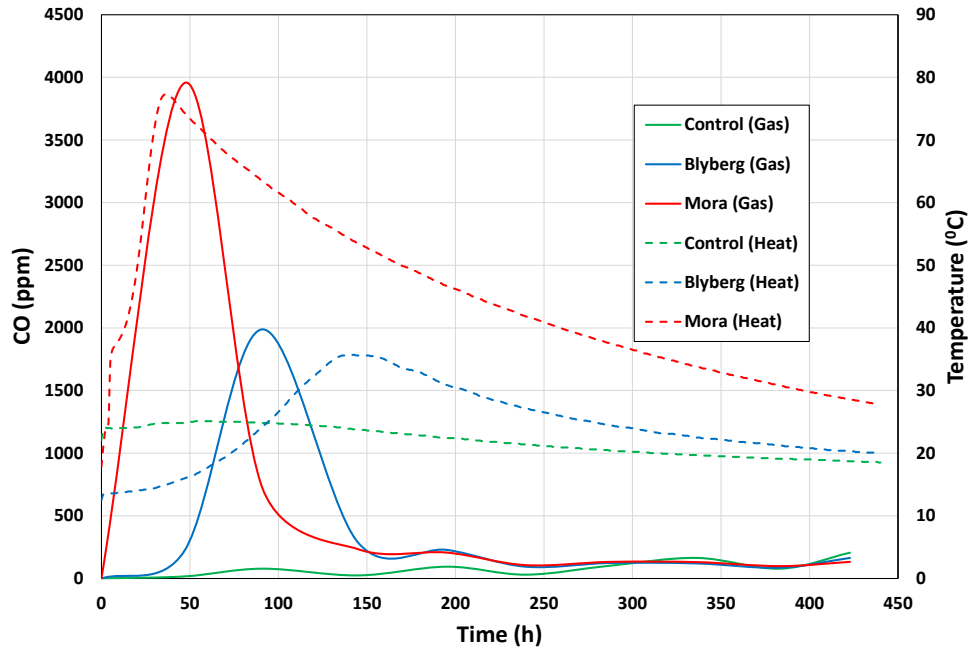


Figure 30. Concentration of carbon monoxide and temperature inside the pile at each time point during the 21-day storage period for pellets produced from fresh mature wood (Mora), fresh juvenile wood (Blyberg) and Control sawdust, under low-temperature drying in a belt dryer.

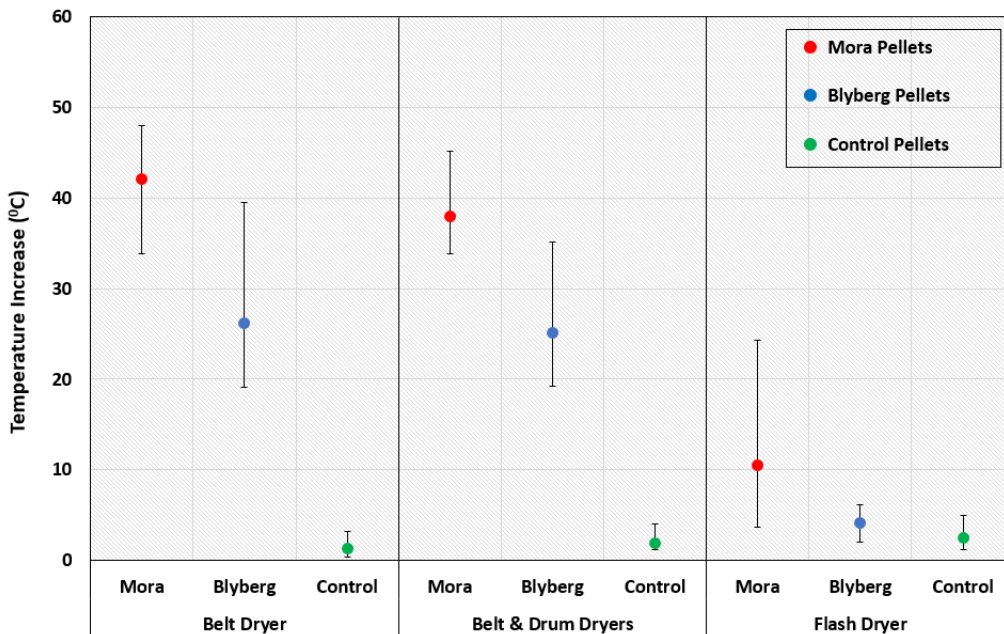


Figure 31. Mean maximum temperature increase inside the piles of wood pellets produced from fresh mature wood (Mora), fresh juvenile wood (Blyberg) and Control sawdust dried using three different methods, namely; low-temperature drying in a belt dryer, medium-temperature drying in a hybrid of belt & drum dryers, and high-temperature drying in a flash dryer. Error bars indicate the highest and lowest maximum temperature increase.

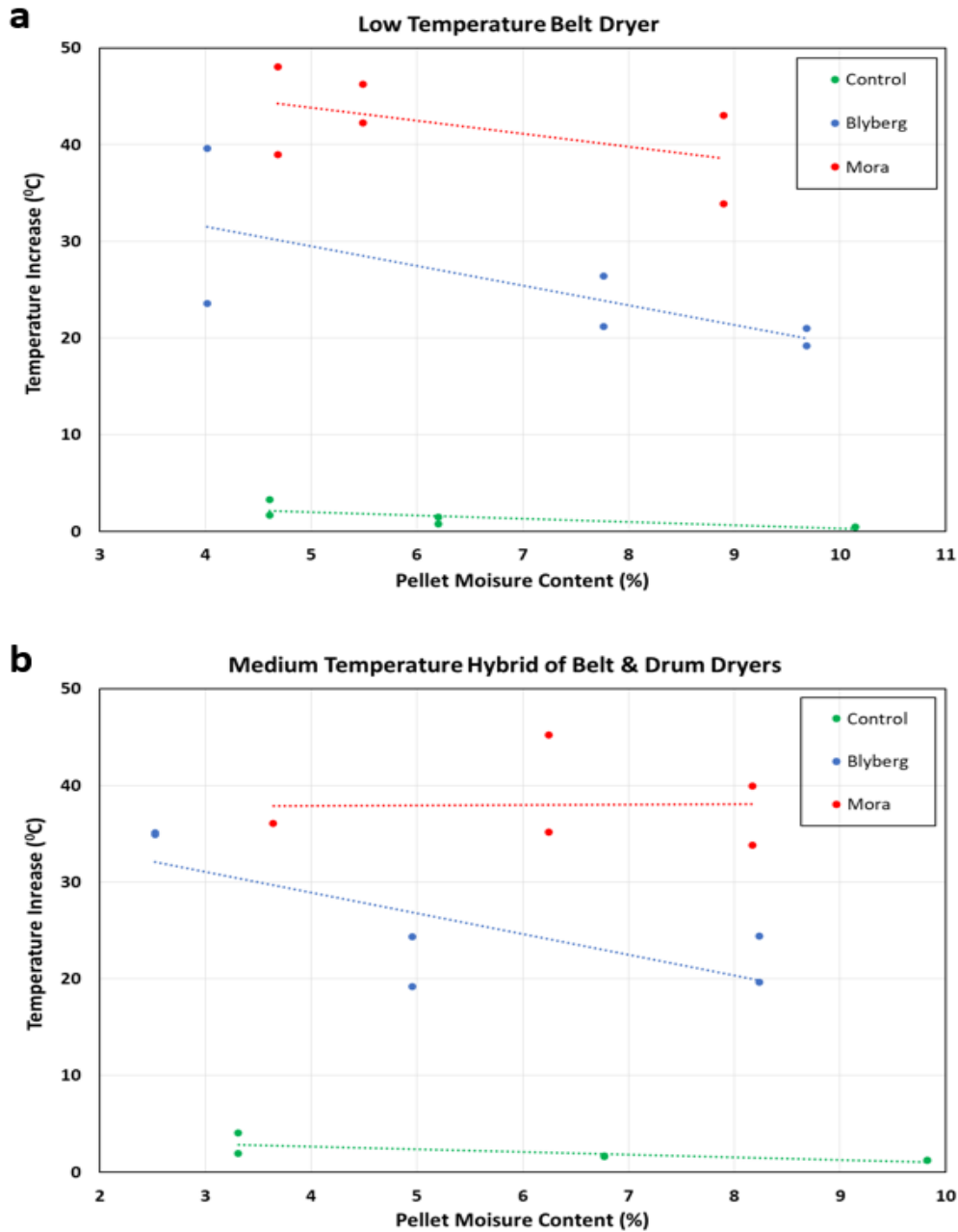


Figure 32. Maximum temperature increase inside the piles in relation to pellet moisture content for wood pellets produced from fresh mature wood (Mora), fresh juvenile wood (Blyberg), and Control sawdust, under low-temperature drying in a belt dryer (a), and medium-temperature drying in a hybrid of belt & drum dryers (b). The data for lowest moisture content of Mora pellets is missing in figure (b) because the temperature data logger malfunctioned.

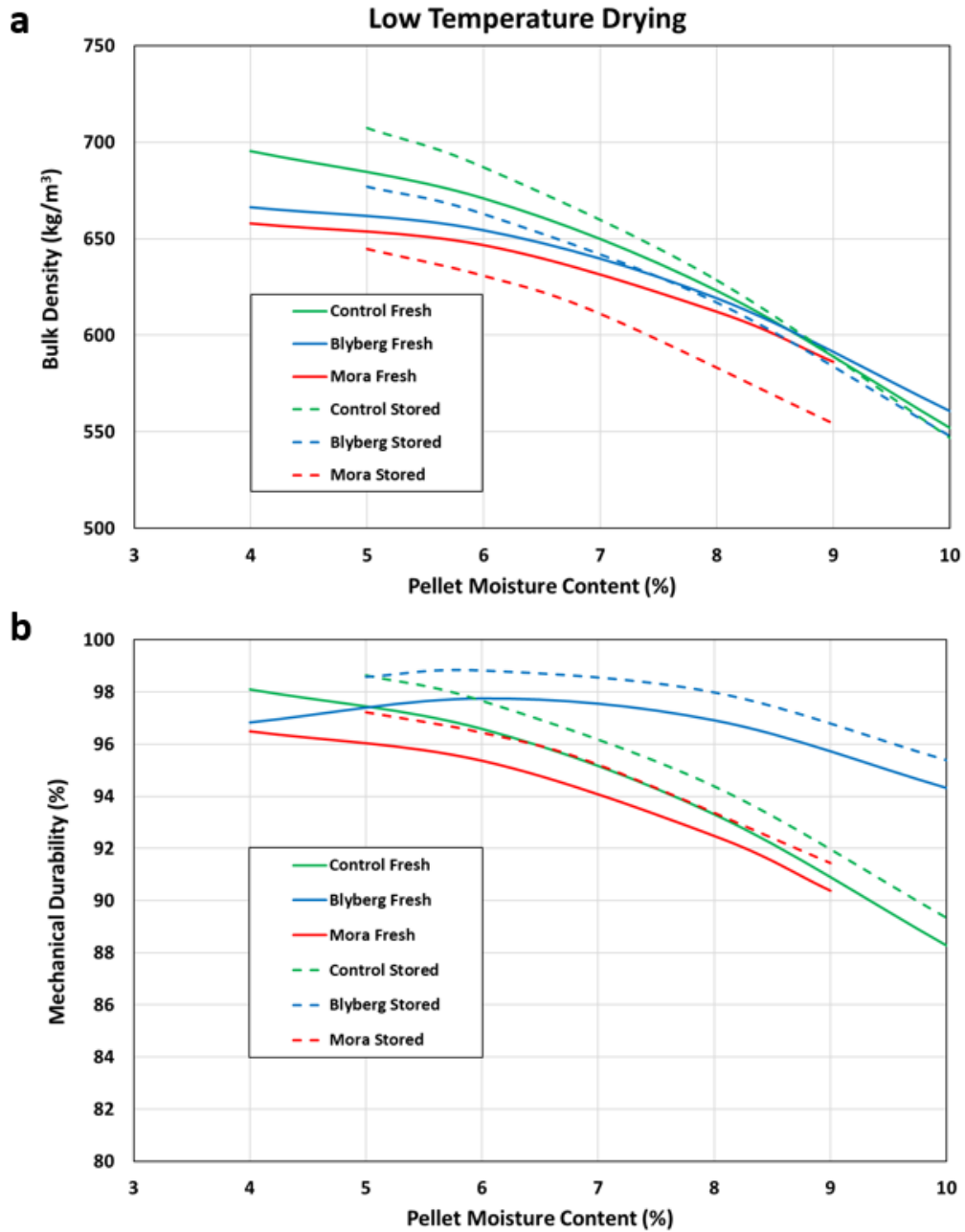
The results from this paper showed that Scots pine wood pellets produced from mature wood sawdust have a higher self-heating and off-gassing propensity compared to those produced from juvenile wood sawdust. Additionally, employing a drying method that uses steam and

has rigorous temperature settings reduces the wood pellets' self-heating and off-gassing propensity.

4.5 Paper V

The objective of this paper was to evaluate the quality properties of Scots pine wood pellets made from sawdust generated from mature and juvenile wood. The study further investigated the effects of raw material type and pellet moisture content on the pellet quality properties for three different drying processes.

The D-optimal models that were developed indicated that the main factors, raw material and pellet moisture content, had significant effects on both the bulk density and mechanical durability, with a good reproducibility and high predictability (all R^2 and Q^2 values were between 0.79 and 0.98). Figures 33 – 35 illustrate the influence of pellet moisture content on bulk density and mechanical durability across different materials and drying processes. Across all investigated drying processes, wood pellets produced from juvenile wood sawdust (Blyberg) had comparable bulk density and mechanical durability to pellets produced from the regular sawdust blend, with a high proportion of stored sawdust. However, pellets produced from mature wood sawdust (Mora) had lower bulk density and mechanical durability.



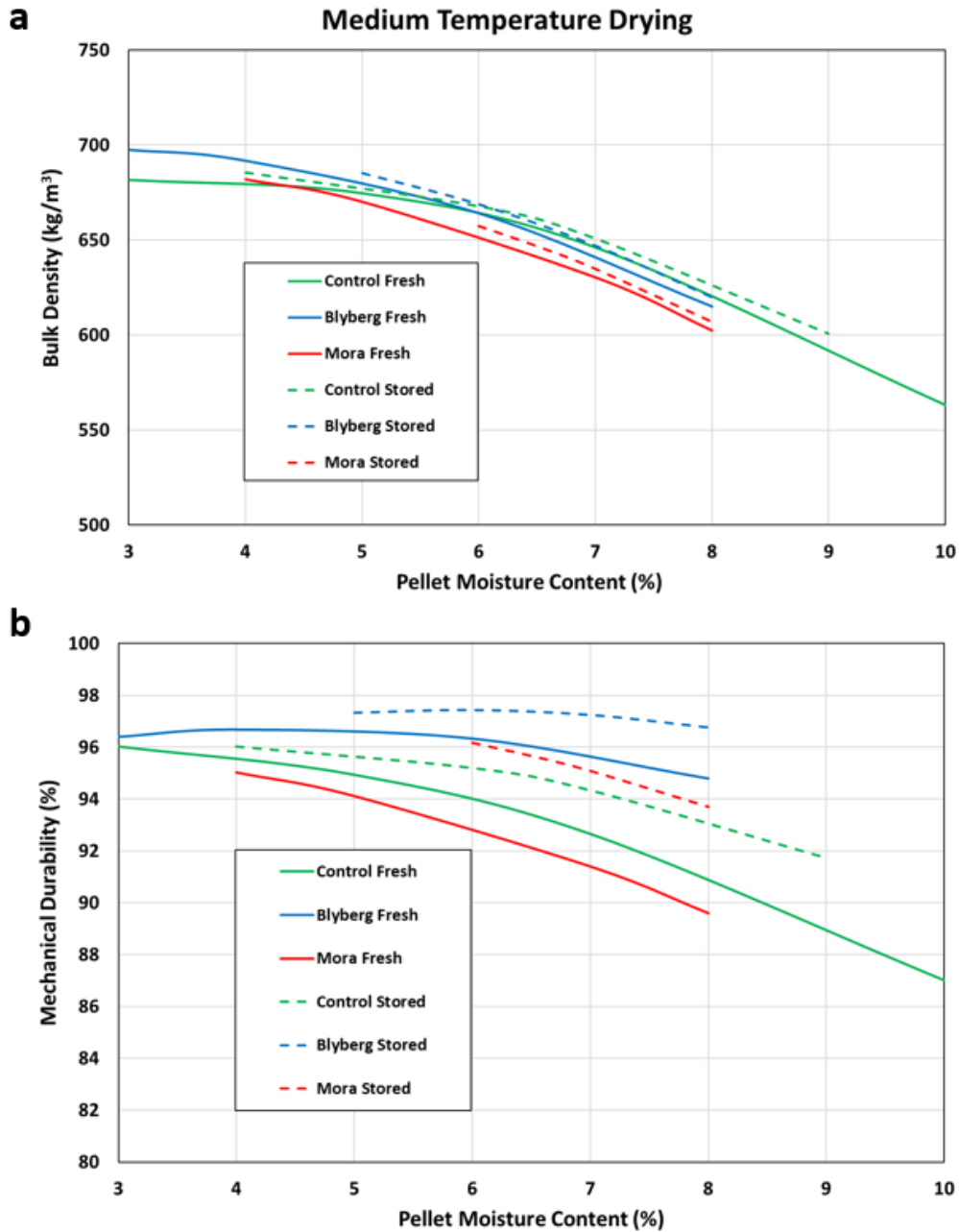


Figure 34. The bulk density (a) and mechanical durability (b) in relation to pellet moisture content for wood pellets produced from fresh mature wood (Mora), fresh juvenile wood (Blyberg), and Control sawdust, under medium-temperature drying in a hybrid of belt & drum dryers.

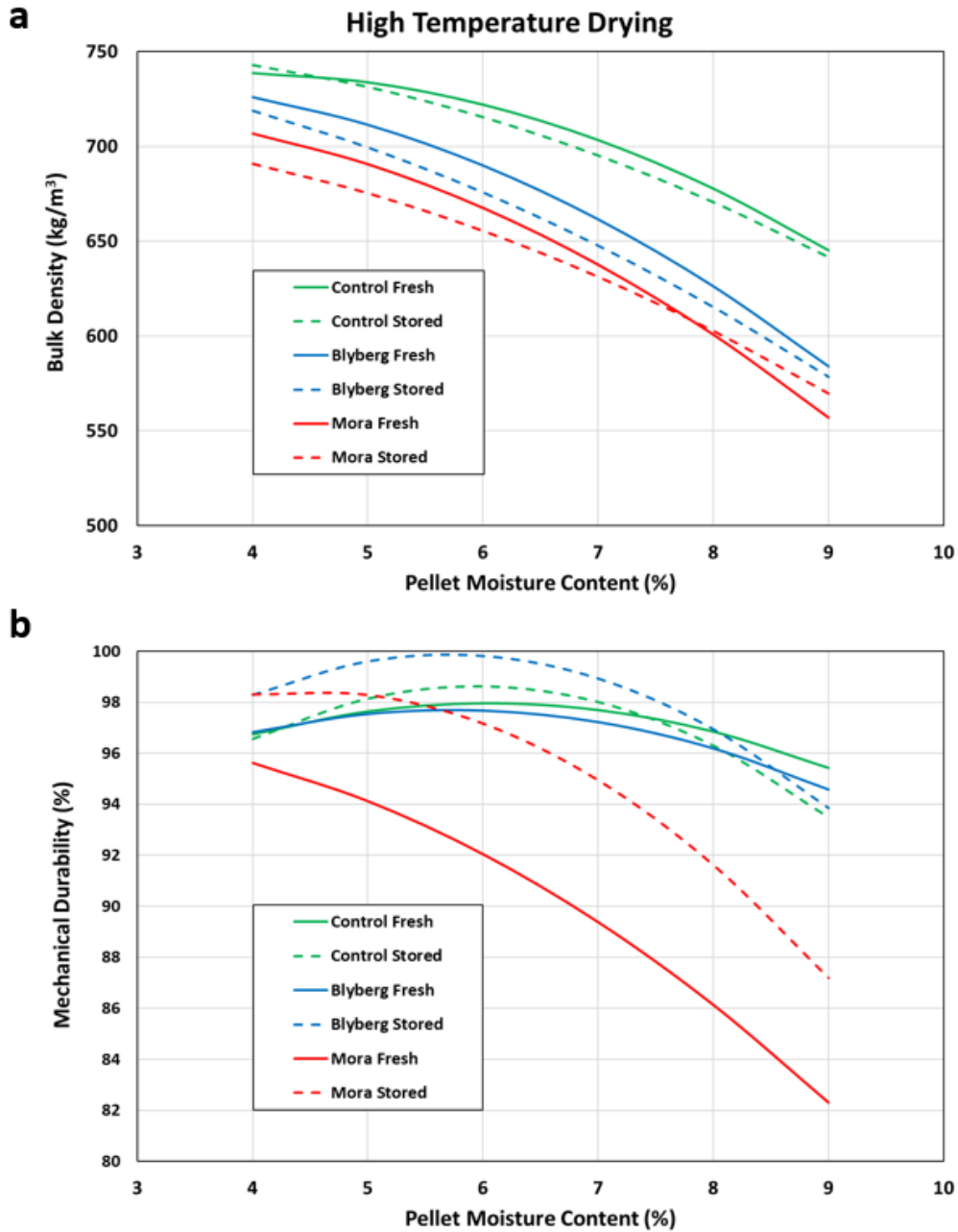


Figure 35. The bulk density (a) and mechanical durability (b) in relation to pellet moisture content for wood pellets produced from fresh mature wood (Mora), fresh juvenile wood (Blyberg), and Control sawdust, under high-temperature drying in a flash dryer.

5.0 DISCUSSION

The primary emphasis in this thesis has been on advancing our understanding of self-heating and off-gassing of wood pellets, particularly those produced from freshly generated pine sawdust. As stated in the objectives, the aim was to increase the body of knowledge in this field by investigating how wood extractives and sawdust raw materials originating from different parts of a tree impact the tendencies of wood pellets to undergo self-heating and off-gassing. This section discusses the findings from all the five papers included in this thesis. Additionally, the section evaluates the materials and methods employed in the studies by analysing their strengths and limitations.

5.1 Effect of Extractives on Off-gassing

The off-gassing of Scots pine wood pellets (paper I) and pure cellulose pellets (paper II) were investigated with regard to the effect of total extractives and various extractive types. Generally, pine sawdust pellets generated higher gas concentrations than pure cellulose pellets. For example, the concentration of carbon monoxide for pine sawdust pellets with added linseed oil was 10 630 ppm (Figure 22), while that of pure cellulose pellets with added linseed oil was 1 910 ppm (Figure 24). This was because off-gassing in pure cellulose pellets was only affected by the added oils while in pine sawdust pellets it was a combination of factors including; the inherent wood extractive, the added oils and possibly other wood chemical compounds.

The concentrations of CO and CO₂ started increasing during the first 24 h of measurements while that of CH₄ picked up later. There was a residence time of about 50 - 80 h for the Scots pine wood pellets (Figure 23) and 80 - 120 h (Figure 25) for pure cellulose pellets before the formation of CH₄ began. This showed that off-gassing of CO and CO₂ during storage of wood pellets is faster in freshly produced pellets and reduces as the pellets' storage time increases while the emission of CH₄ can continue even in wood pellets that have been stored for some time. The consumption of oxygen in both cases corresponded to the increased concentrations of CO and CO₂, pointing to the fact that

oxidative reactions were taking place. This aligns with prior research studies [29–31] which have linked wood pellet off-gassing to the oxidation of wood extractives.

The pellets produced from stored and acetone extracted pine sawdust exhibited low concentrations of CO (Figure 22) and CO₂, despite having sufficient residual oxygen. This suggests that stored and acetone-extracted pine sawdust lost the capability of fresh pine sawdust to generate CO and CO₂, as well as to consume oxygen. From this, it can be tempting to draw the conclusion that the gas emissions are directly connected to the quantity of extractives in the raw material. However, this is not generally true because adding more extractives to fresh pine sawdust did not result in higher emissions. Moreover, the linear regression analysis results (Figure 21) showed that there was no correlation between the off-gassing of CO, CO₂ and CH₄ and the total extractive contents of the raw materials. The reduced gas emissions in stored and acetone extracted pellets may be attributed to the removal of specific extractives during storage and extraction of sawdust. The triglyceride content is known to decrease during storage, while other extractive groups may remain relatively constant [112]. However, addition of three different kind of extractives to the sawdust including triglycerides and fatty acids from linseed oil and tall oil did not increase the emissions. The plausible explanation to this could be that, beyond the influence of specific extractives, gas formation may also depend on special sites within the wood, and these remained consistent across samples due to the use of the same base material (fresh pine sawdust).

The study involving pure cellulose pellets with added oils revealed that variations in the composition of fatty and resin acids in the additive oils influenced off-gas emissions. Pellets with added linseed oil exhibited high gas emissions (Figure 24) owing to its high content of unsaturated fatty acids, notably 73.9% linolenic and 7.6% linoleic acids (Table 6). Despite containing around 45% unsaturated linolenic and oleic acids, pellets with added tall oil showed low gas emissions. This indicated that reducing the unsaturated fatty acids content of the raw material can significantly mitigate the off-gassing of carbon oxides and methane. Previous studies by Arshadi et al [29] and Attard et al [33] similarly found unsaturated fatty acids to have high impact on off-gassing.

Therefore, considering that the concentration of residual oxygen was decreasing (Figure 26) as the concentration of carbon oxides increased in the pellets with added linseed oil, it can be concluded that the formation of carbon oxides was due to α -oxidation of the unsaturated fatty acids linolenic and linoleic. However, the pellets with added rosin had low gas emissions despite containing 100% resin acids and the possibility of oxidation of abietic acid to dehydroabietic acid [31]. There was little to no emission of the carbon oxides in the pellets with added coconut oil and tall oil because their main composition; saturated fatty acids are highly stable [56].

Methane emission did not start until the oxygen was depleted. It was also noted that significant amount of methane was only formed in pellets that had high carbon dioxide and carbon monoxide concentrations. This indicated that methane formation is linked to the formation of the carbon oxides and reduction of residual oxygen. However, it is difficult to explain the chemical reaction mechanisms responsible for the formation of methane in conditions under which the pellets were stored, and to the author's knowledge, there are no known abiotic mechanisms that generate methane from organics having similar composition to the studied extractives. On the other hand, there are biotic explanations for methane formation [113,114], and microorganisms and living parenchyma cells, could be special gas forming sites in wood pellets discussed above.

5.2 Effects of Raw Materials & Pre-treatments on Self-heating & Off-gassing

Building upon the findings from papers I and II, separating sawdust based on variations in extractives composition appeared to offer a promising and practical solution to mitigate self-heating and off-gassing in wood pellets. Therefore, in the subsequent studies (papers III and IV), sawdust was separated based on sapwood and heartwood, as well as mature and juvenile wood compositions.

The results from the industrial production and piles storage trials showed a connection between self-heating and off-gassing (paper IV). The storage piles with high temperatures also had high concentrations

of off-gases (Figure 30). Sedlmayer et al. [26] in their methods correlation analysis, similarly observed a direct correlation between self-heating, determined by isothermal calorimetry, and off-gassing, measured using offline and glass flask methods. This suggests that self-heating and off-gassing in wood pellets either occur concurrently with a common mechanistic explanation, or are influenced by common factors.

The self-heating and off-gassing of biomass in general is reported to be a results of either physical transition reactions, biological oxidation, chemical oxidation or a combination of these mechanisms [10,14]. However, most studies [26,29,30,33] primarily attribute the self-heating and off-gassing of wood pellets to chemical oxidation of wood extractives, particularly fatty acids. The biological oxidation is often overlooked due to the assumption that the pellets are relatively dry (with a moisture content of 5 – 10%) and that microorganisms cannot survive the pelletization temperature of around 100 °C. However, the findings from the study in paper III suggested the possible involvement of a biological mechanism in the self-heating and off-gassing of wood pellets.

The off-gassing results for the pellets produced from separated sapwood and heartwood sawdust (paper III) indicated a strong linear correlation between off-gassing and sapwood/heartwood composition, with correlation coefficient (R) values greater than 0.9 for all the three off-gases (Figure 27). Specifically, increasing the sapwood content of the raw material increased off-gassing of the wood pellets. Fresh wood pellets produced from 100% sapwood sawdust produced the highest concentrations of CO (Figure 28 a), CO₂ and CH₄ under consumption of oxygen (Figure 28 b), and the effects were diminishing with higher drying temperature (Figure 29 a-b). This was despite sapwood having a lower total extractives and fatty and resin acids content than heartwood. The total lipophilic extractives in sapwood and heartwood were found to be in the range of 2.6 – 4.2% and 5.1 – 6.8%, respectively. The amounts of fatty acids decreased with an increase in drying temperature while the resin acids increased in both sapwood and heartwood. Therefore, the decrease in off-gassing of sapwood pellets due to increased drying temperature may be attributed to the reduction

in the total fatty acids in the raw material. However, increasing the drying temperature for heartwood resulted in increased off-gassing of the wood pellets, even though the fatty acid content of the raw material had decreased. This showed that other mechanisms than just the chemical oxidation of fatty acids contribute to off-gassing of wood pellets.

The self-heating and off-gassing study of mature and juvenile wood pellets equally showed significant variations in maximum temperature increase (Figure 31) and gas concentrations (Figure 30) among different materials. The type of raw material used, particularly wood maturity influenced the self-heating and off-gassing propensity of the pellets. Additionally, the moisture content of the pellets and the drying method employed were also found to have an influence. Generally, across all materials, the temperature within storage piles decreased as pellet moisture content increased (Figure 32). However, the difference drying methods had a stronger effect (Figure 31). The pellets produced from sawdust dried at low temperature in a belt dryer, and at medium temperature in a hybrid of belt & drum dryers exhibited high levels of self-heating. In contrast, steam drying sawdust at high temperature and pressure in a flash dryer resulted in a significant reduction in self-heating for all materials. The piles containing wood pellets made from fresh mature wood sawdust (Mora) had the highest temperatures and gas concentrations. Temperatures ranging from 70 to 80 °C, representing an increase of about 50 °C, were recorded. Additionally, pellets placed on top of these piles disintegrated, likely due to increased self-heating (Figure 36). This deterioration can negatively impact pellet quality properties and result in dry matter loss [22]. On the other hand, the pellets produced from fresh juvenile wood sawdust (Blyberg) showed significant reductions in both the maximum temperature increase and gas concentrations compared to those produced from fresh mature wood sawdust.

The results obtained from the study of juvenile and mature wood pellets closely corresponded with the results of the study involving sapwood and heartwood pellet. The increased self-heating and off-gassing in fresh mature wood pellets can be attributed to its higher proportion of sapwood containing living parenchyma cells [73], or the

increased nutrient storage in mature sapwood compared to juvenile sapwood. The findings in both studies fit well with a biological explanation for self-heating and off-gassing. Regarding the emission of CO₂ and CH₄ and the consumption of O₂, it is likely that cells, either microbes or more probable surviving parenchyma cells, initially aerobically metabolize stored starch/fats (found in the sapwood), producing CO₂, and subsequently undergo anaerobic methane fermentation [115]. The formation of high concentrations of CO is however puzzling, but there are reports of biological processes forming CO in for instance cell signalling [116]. One possibility is that formation of this toxic gas is a stress reaction of the wood cells with a purpose to harm invading organisms. The formation of CO as a stress reaction has been reported in animals [117], and also in plants [118].



Figure 36. Storage piles for wood pellets made from fresh mature wood sawdust (*Mora*) and low temperature drying in a belt dryer, exhibited high self-heating tendencies leading to the disintegration of the pellets on top of the piles. (Photo by author)

Another puzzling observation, as discussed earlier, was the increase in off-gassing of heartwood pellets with increasing drying temperature. This can be interpreted in two ways, one is that another abiotic reaction is responsible for the gas formation, and the elevated drying temperature in some way stimulates this reaction, possibly by redistributing some extractives. Another explanation is that the high

temperature during drying inactivates antimicrobial chemicals in the heartwood, such effects are reported for antibiotics [119]. The decreased effect of high temperature drying on materials with high sapwood content aligns with the biological explanation of self-heating and off-gassing. Essentially, high temperature simply killed a large fraction of the living parenchyma cells, hence leading to less heat and gas generation. The same principle applies to the storage of raw material, which led to significant reduction in self-heating and off-gassing of the control samples in paper IV, as well as off-gassing of sapwood pellets in paper III. Over time, the parenchyma cells naturally die due to nutrient depletion.

Although there appeared to be an increase in the concentration of off-gases in heartwood pellets made from stored sawdust compared to those produced from fresh sawdust, the difference was not statistically significant. Furthermore, there were no significant differences in off-gas concentrations between stored sapwood and fresh heartwood pellets. This suggests that the storage of raw material had mitigated the off-gassing of sapwood pellets to the same extent as that of heartwood pellets produced from fresh sawdust. Therefore, the effects of raw material storage may vary depending on the composition of the biomass raw material.

5.3 Effects of Raw Materials & Pre-treatments on Pellet Quality Properties

The quality properties of wood pellets are influenced by various factors, including press parameters like die channel length-to-diameter (L/D) ratio, die temperature and pressure, as well as feedstock-related variables such as moisture content, particle size, additives, and biomass types and mixes [42,78,91,120,121]. Therefore, the separation and selection of sawdust raw materials, along with adjustments to pre-treatment methods to mitigate self-heating and off-gassing, can impact on the quality properties of the produced wood pellets.

Bulk density and durability are two most important mechanical properties to consider for pellet storage and transportation. High bulk density gives high energy content per unit volume, while high

mechanical durability enables the pellets to have high resilience against breakage [84]. For all the materials and drying methods, the bulk density decreased as pellet moisture content increased within the range of 3% to 10% (Figures 33a – 35a), while mechanical durability improved with increasing pellet moisture content up to an optimal level of about 6%, after which it began to decrease (Figures 33b – 35b). This may be explained by the fact that moisture lowers the friction occurring when the feedstock is compacted and pressed through the die channels and consequently also the back pressure [122]. High die temperature temperatures and lower moisture content has been known to favour high bulk density [84].

The pellets produced from Mora (mature wood) sawdust had lower bulk density and mechanical durability compared to those produced from the control sawdust (mainly stored sawdust) and Blyberg (juvenile wood) sawdust. This can be attributed to the high content of lipophilic extractives in Mora sawdust. Previous studies have shown that the lipophilic extractive content of the raw material significantly affects the bulk density and durability of pine pellets, storage of sawdust leads to a reduction in lipophilic extractives, thereby enhancing the bulk density and durability of the resulting pellets [41].

5.4 Discussion of Materials

Scots pine (*Pinus sylvestris*) sawdust was selected for three self-heating and off-gassing studies (papers I, III, and IV) because among the European produced pellets, Scots pine wood pellets are the most susceptible to self-heating and off-gassing [35]. Moreover, Scots pine and Norway spruce (*Picea abies*) are the dominating tree species in Swedish forests with each respectively contributing 38% and 40% to the total wood volume [124]. Therefore, the primary raw material for wood pellet production in Sweden comprises sawdust and other residues derived from processing pine and spruce trees.

Paper II utilized pure cellulose so as to eliminate the possible reactions and involvement of other wood chemical compounds such as hemicelluloses and lignin in the off-gassing process. Hemicelluloses, in particular, are known for their chemical and thermal instability. Even

at relatively low temperatures, the saccharide related molecules within hemicelluloses can undergo various oxidative reactions [75]. Pelletization being a thermal process is likely to degrade some hemicelluloses and, in the process, form compounds that may have an effect on off-gassing. Therefore, by using only pure cellulose and added extractives, the study aimed to isolate and understand the specific influence of different types of extractives on off-gassing, given that cellulose is quite stable both chemically and thermally.

5.5 Discussion of Methods

A number of studies on self-heating and off-gassing of wood pellets have employed heat and gas measuring techniques such as isothermal calorimetry that require preheating and the use of a small pellet quantities [19,34,123]. This is due to the challenge of conducting such experiments on large volumes of pellets in storage piles or industrial silos where the heat and gases quickly diffuses or escape as soon as they are formed. To overcome this challenge, the method employed in papers I, II, and III (Figure 18) involved storing the pellets in closed containers. This effectively prevented the escape of formed gases, maintained consistent storage environmental conditions (temperature and humidity), and enabled real-time measurement of off-gassing. However, considering the nature of the study and the pre-treatments applied to the raw materials, gas measurements were limited to just two replicates per sample as a larger quantity of pellets (approximately 7 – 8 kg per test) was needed. Despite this limitation, the samples were representative because for every sample of pellets, 14 – 16 kg of about 20 kg that was produced was subjected to off-gassing measurements.

The piles storage method (Figure 19Figure 20) offered an advantage in that it replicated the actual conditions found in industrial settings where wood pellets are stored in large volumes. This facilitated the study of the self-heating and off-gassing phenomena under conditions that more closely mimic real-world scenarios, thereby providing more meaningful and applicable results. However, the method had the same limitation regarding the number of samples as the container storage method. Additionally, the gas trapping tubes employed for off-gassing measurements were not very effective.

The use of 30 L capped containers to extract the sawdust in paper I (Figure 14a) proved to be effective. The total extractive content decreased from 5.1% in fresh sawdust to 1.2% in extracted sawdust, and there was a noticeable change in colour of the sawdust from brownish to whitish. The selection of acetone as a solvent was intentional due to its safer handling properties, especially considering the large quantity of sawdust involved in the extraction process. While the industrial feasibility of extracting sawdust with acetone may pose handling and economic challenges, the method provided valuable information on the impact of producing wood pellets from extractive free sawdust, particularly concerning off-gassing effects.

6.0 CONCLUSIONS

The following conclusions were made based on the collective results obtained across all independent studies conducted in this thesis

1. There is a connection between self-heating and off-gassing of wood pellets. As heat generation increases, the concentration of emitted gases also increases, indicating that both processes are influenced by shared underlying mechanisms.
2. There were two main conclusions regarding the effect of extractives on off-gassing of wood pellets.
 - a. The total amount of extractives in the raw material has little effect on off-gassing of wood pellets.
 - b. Off-gassing of wood pellets is largely influenced by specific types of extractives, particularly unsaturated fatty acids. Lowering the unsaturated fatty acids content of the raw material can substantially decrease off-gassing of the wood pellets.
3. The composition of sapwood and heartwood has an effect on the propensity for self-heating and off-gassing in Scots pine wood pellets. A higher sapwood content results in increased self-heating and off-gassing. Furthermore, Scots pine wood pellets produced from mature wood sawdust exhibit a greater propensity for self-heating and off-gassing compared to those made from juvenile wood sawdust.
4. The self-heating and off-gassing of wood pellets appear to arise from a combination of abiotic chemical oxidation of unsaturated fatty acids and biological processes. The noticeable difference in off-gassing observed between sapwood and heartwood pellets, underscores the involvement of biological mechanisms.
5. The formation of methane during storage of wood pellets is dependent on anaerobic conditions, whereas formation of

carbon oxides can occur both under aerobic and anaerobic conditions.

6. The storing of sawdust raw material and drying temperature can play a crucial role in minimizing the risks of self-heating and off-gassing of wood pellets. However, the effectiveness of these measures largely depends on the composition of the feedstock. Generally, employing a high-temperature steam drying method significantly reduces the likelihood of wood pellets undergoing self-heating and off-gassing.

7.0 CONTRIBUTIONS & FUTURE PERSPECTIVES

The results from this thesis have generated additional knowledge towards our understanding of self-heating and off-gassing in wood pellets. Particularly, the identification of the connection between these phenomena and the sapwood/heartwood content is a novel contribution. The findings are also significant for the wood pellet production industry. Notably, the observed effects of drying temperature and raw material types highlights the need for careful consideration in the selection of both parameters to mitigate the risk of self-heating and off-gassing during wood pellet storage and transportation.

Storing of fresh sawdust for a period of time prior to pellet production remains the most effective method for minimizing self-heating and off-gassing of wood pellets. However, sorting and separating the raw materials at the source can facilitate the development of storage schedules tailored to specific raw materials. This approach has the potential to substantially reduce the overall storage time required for raw materials. Therefore, in the context of sawdust supply for pellets production in Sweden, there is need to investigate the optimal storage durations for spruce and pine sawdust obtained from different sources, such as Blyberg and Mora sawdust. Based on findings from both the self-heating and off-gassing measurements, as well as the pellet quality properties, we advise pellet producers in Sweden to consider processing pine sawdust from small diameter timbers (Blyberg) directly, and prioritise storing of sawdust from large diameter timbers (Mora) before pellet production.

This study only focused on the self-heating and off-gassing of Scots pine wood pellets. However, given the growing demand and utilization of pellets, it is expected that a broader range of biomass types will be used for production. Pellets made from non-wood biomass may exhibit differing self-heating and off-gassing tendencies compared to wood pellets. Therefore, there is need for further research to investigate self-heating and off-gassing of various biomass pellets.

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Mitigating Off-Gassing and Self-Heating in Fuel Wood Pellets Storage

The global wood pellets production increased from about 18 million tonnes in 2012 to about 46 million tonnes in 2022. Wood pellets have become a preferred solid biomass fuel for heat and power generation due to their standardized nature, known properties, and consistent quality. However, pellets produced from fresh sawdust sometimes undergo self-heating and off-gassing and this poses a challenge for their transportation and storage. The aim of this thesis was to increase the body of knowledge towards understanding the mechanisms underlying the self-heating and off-gassing of wood pellets, and offer solutions for producing wood pellets with reduced tendencies for self-heating and off-gassing using freshly generated sawdust.

The results showed that a biological process, in combination with the chemical oxidation of unsaturated fatty acids lay behind the self-heating and off-gassing of wood pellets. While storing of fresh sawdust for a period of time prior to pellet production remains the most effective method for mitigating self-heating and off-gassing, sorting and separating the raw materials at source can facilitate the development of storage schedules tailored to specific raw materials, thereby reducing on the raw material storage time.

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