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Faculté de génie
Département de génie chimique et de génie biotechnologique

OPTIMISATION DES PROCÉDÉ DE COMBUSTION POUR LA VALORISATION DES RÉSIDUS SOLIDES INDUSTRIELS EN ENERGIE

OPTIMIZING COMBUSTION PROCESSES FOR THE VALORIZATION OF INDUSTRIAL SOLID WASTE INTO ENERGY

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Rafael ELOY DE SOUZA

Sherbrooke (Québec) Canada
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JURY MEMBERS

_____

Bruna REGO DE VASCONCELOS

Directrice de recherche

_____

Inès ESMA ACHOURI

Rapporteur

_____

Eduardo LINS DE BARROS NETO

Évaluateur

_____

Juan Alberto CHAVEZ RUIZ

Évaluateur

Résumé

Le changement climatique a intensifié les phénomènes naturels à travers la planète, incitant la communauté scientifique à avertir des impacts potentiellement irréversibles. Cela a poussé le milieu académique et l'industrie à rechercher des alternatives pour atténuer les effets du changement climatique. Les stratégies de transition énergétique et de gestion des déchets solides sont devenues essentielles pour un développement durable. La valorisation énergétique par combustion offre une approche prometteuse pour convertir les résidus en chaleur ou en électricité utile. Cette étude vise à explorer la faisabilité technique de la valorisation énergétique de deux types de résidus industriels : les résidus de papier provenant d'une industrie de stratifiés haute pression (HPL) et les résidus de cannabis. Les deux types de résidus ont été granulés et soumis à une combustion dans un réacteur à lit fixe sous des conditions contrôlées.

Dans la première étude, les résidus de papier ont été catégorisés en groupes phénoliques, non-phénoliques et mixtes. Les résultats ont montré que les résidus de papier phénolique, lorsqu'ils sont combinés avec des résidus non-phénoliques, ont amélioré la température d'inflammation et la libération de chaleur, en particulier à une température de combustion de 300°C et un coefficient d'air de 3,0. Malgré la teneur élevée en cendres, les granulés mixtes ont démontré une compétitivité dans la production d'énergie.

La deuxième partie de l'étude a examiné le potentiel de valorisation énergétique des résidus de cannabis. Un prétraitement par explosion de vapeur a été utilisé pour évaluer son impact sur les propriétés des granulés et le comportement à la combustion. Les granulés de cannabis ont été brûlés à 290°C, avec trois coefficients d'air différents (α=1,0, α=1,15, α=1,3). Bien que le prétraitement par explosion de vapeur ait amélioré la valeur calorifique, la teneur en cendres et la durabilité des granulés, il a eu un effet négatif sur la combustion, conduisant à une combustion incomplète. Parmi toutes les réactions, la meilleure qualité de combustion pour les granulés de cannabis a été obtenue avec un coefficient d'air de α=1,15 pour les granulés sans prétraitement par explosion de vapeur, résultant en une conversion d'oxygène plus élevée et des émissions de CO_2 accrues.

Les deux études mettent en évidence le potentiel de valorisation énergétique de deux types de résidus industriels. Un contrôle approprié des paramètres de combustion est essentiel pour optimiser l'efficacité de la combustion et garantir une gestion durable des déchets ainsi que la production d'énergie.

Mots-clés: résidus industriels, combustion, granulés, explosion de vapeur, valorisation énergétique.

Abstract

Climate change has intensified natural phenomena across the planet, prompting warnings from the scientific community about the potential for irreversible impacts. This has driven academia and industry to seek alternatives to mitigate climate change effects. Strategies for shifting energy profiles and managing solid waste have become critical for sustainable development. Energy recovery through combustion offers a promising approach to converting waste into useful heat or electricity. This study aims to explore the technical feasibility of energy recovery from two distinct industrial residues: paper residues from a High-Pressure Laminate (HPL) industry and cannabis residues. Both residues were pelletized and subjected to combustion in a fixed-bed reactor under controlled conditions. In the first study, paper residues were categorized into phenolic, non-phenolic, and mixed groups. The results showed that phenolic paper residues, when combined with non-phenolic residues, enhanced the ignition temperature and heat release, particularly at a combustion temperature of 300°C and an air coefficient of 3.0. Despite the high ash content, the mixed pellets demonstrated competitiveness in energy production. The second part of the study investigated the energy recovery potential of cannabis residues. Steam explosion pretreatment was used to assess its impact on pellet properties and combustion behavior. The cannabis pellets were combusted at 290°C, with three different air coefficients (α=1.0, α=1.15, α=1.3). While steam explosion pretreatment improved the calorific value, ash content, and pellet durability, it adversely affected combustion, leading to incomplete combustion. Among all the reactions, the best combustion quality for cannabis pellets was achieved with an air coefficient of α=1.15 for pellets without steam explosion pretreatment, resulting in higher oxygen conversion and CO_2 emissions. Both studies highlight the potential for energy recovery from two industrial residues. Proper control of combustion parameters is essential to optimize combustion efficiency and ensure sustainable waste management and energy production.

Keywords: industrial waste, combustion, pellets, steam explosion, energy recovery.

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1. INTRODUCTION

1.1. Context

Global leaders are concerned about reducing greenhouse gas (GHG) emissions due to the harmful effects these gases have brought to the atmosphere, causing climate change. These gases have been released into the atmosphere for many years and, in large quantities, cause the Earth's surface warming. Climate change is already intensely discussed, and the possibility of mitigating the damage that GHG emissions have caused to the planet has been widely studied. Science has already warned the community the dangers that global warming will have on us that changes must be done now to protect the planet and secure the future as well as to build a strong and resilient economy for the next generations, harnessing the power of a cleaner future (Government of Canada, 2021).

The temperature in Canada is rising in a pace two times faster than the global average, and in the north of Canada it is three times faster. Reducing GHG emissions will be an essential step in Canada's future trajectory (Government of Canada, 2022). The country has committed to reducing emissions across the entire economy to reach an emissions reduction target of 40 to 45% below 2005 levels by 2030 and making they actions to lead the country to achieve net-zero emissions by 2050 (Government of Canada, 2024a). The province of Quebec is committing to reduce up to 37.5% of emissions at levels below those recorded in 1990 by 2030. In addition, the province announced in 2015 that it intends to become carbon neutral by implementing a policy of reducing GHG emissions by 80-95% below 1990 levels by 2050 (MELCCFP, 2024).

According to the national inventory report, total national greenhouse emissions were 708 megatons (Mt) of carbon dioxide equivalent (Mt CO_{2eq}) in 2022 (Government of Canada, 2024a). The energy sector (consisting of stationary combustion sources, transport and fugitive sources) emitted 82% of Canada’s total GHG emissions (Government of Canada, 2024a). Quebec's GHG emissions represented 11.6% of Canadian emissions, which reached 670.4 Mt CO_{2eq}. (MELCCFP, 2023). The transport sector is the leading emitter of GHGs, contributing with 42.6% of Quebec's emissions (MELCCFP, 2023). The industry sector ranks second in

GHG emissions, estimated at 32.3% of total emissions. In this sector, 52.6% of emissions come from industrial processes and the use of products, 46.5% come from combustion, and 0.9% are fugitive emissions (MELCCFP, 2023).

As a part of a strategy to achieve the GHG emissions reduction target, Canada created the 2030 Plan for a Green Economy that helps industries develop and adopt clean technology and work on improving industries' carbon footprint while ensuring their competitiveness and more efficient uses of energy. The government is encouraging existing businesses to invest in reducing their energy consumption through efficiency and heat recovery measures and replacing fossil fuels with renewable forms of energy (MELCCFP, 2024).

GHG emissions are starting to represent high costs to the industry due to the carbon credits system. The Government of Canada has seen carbon pricing as a global opportunity and recognized it as the most cost-effective policy to cut emissions (Carbon Pricing Leadership Coalition, 2022). Since 2019, the carbon market price has been US$20 per ton and has increased by US$10 per ton per year until 2022, when it reached US$50 per ton. After 2022, federal fossil fuel tariffs will increase by $15/ton CO_2 eq annually until fuel charge rates reflect a carbon price of $170/t CO_2 eq in 2030 (Government of Canada, 2022).

In Quebec, this action was also taken. In 2014, Quebec linked its system to that of California under the Western Climate Initiative (Government of Quebec, 2022a). According to the Government of Quebec (2022b), the minimum purchase price will also increase. Table 1.1 below shows an example of the potential evolution of the annual minimum price in California until 2030 for units of the present vintage (Government of Quebec, 2022b).

Table 1.1 – Potential evolution of the joint annual minimum price in US dollars (US$) until 2030. Adapted from: (Government of Quebec, 2022b).

2022	**2023**	**2024**	**2025**	**2026**	**2027**	**2028**	**2029**	**2030**
$18.95	$20.28	$21.70	$23.22	$24.85	$26.59	$28.45	$30.44	$32.57

The government of Quebec has also set numbers for carbon emissions that have decreased over the years. It means that it will be harder and more expensive to buy carbon credits in the next

few years. The carbon purchase ceiling will drastically reduce in the coming years, from 151 $MtCO_2$ in 2023 to 44 $MtCO_2$ by 2030.

The Quebec government has also taken measures to reduce and manage waste dumped in landfills. Fees payable for waste materials disposal are adjusted each year according to the rate of change in Canadian consumer price indices. In 2022, royalties payable was US$24.32/ton. In the same year, the Minister for the Environment and the Fight Against Climate Change proposed increasing the fees for the disposal of waste materials to reduce the amount of materials sent to landfills. The current waste material disposal fee is $32 for each metric ton plus an annual indexation of $2 for subsequent years (Government of Quebec, 2024).

These measures are taken to reduce the disposal of waste materials in landfills, encourage the green economy in Quebec and make the recovery and recycling of these materials more competitive (Government of Quebec, 2022c). Waste valorization implies enhancing the value of waste materials by converting them into useful products, energy, or materials. Recovery of waste is one of the main advantages of the valorization because it reduces the use of primary sources and fosters the circular economy by putting the waste back into the system. These measures reduce the waste sent to landfills or incinerators and minimize environmental impacts resulting from disposal. This assists in cutting down on greenhouse gas emissions (Magazzino et al., 2020).

Even though the paper is related in the literature with low calorific value, the phenol presented in the composition of these residues has a high calorific value that makes the residues competitive to be used to generate heat. The project investigates the feasibility of using these residual materials for energy recovery.

Another industry that has seen exponential growth is the cannabis industry. Since the legalization of cannabis in October 2018, for recreational use, the Canadian cannabis market has experienced rapid growth. Along with this growth, a significant increase in the production of cannabis residues is noticed. Cannabis production generates green residues such as stalks, stems, leaves, and roots not used in the final product. These plant parts are often discarded after harvesting the flowers and leaves for processing. According to Kraszkiewicz et al. (2019), 70-75% of the output process is green residue. This residue has a great potential to be recovered as

energy due to its high energy yield of approximately 100 GJ/ha/y (Brar et al., 2024). Moreover, steam explosion has been a technique suggested in the literature as a pre-treatment for biomass before pelletization to improve bonding characteristics, hydrophobicity, dimensional stability, durability, and high heating value (HHV) of wood pellets (Alizadeh et al., 2022).

The technologies most described for energy recovery from wastes are combustion, gasification, and pyrolysis, where combustion technology standing out as the most widely used and effective process for producing energy and heat from biomass, with the advantage of easy adaptability to established energy conversion technologies (Briones-Hidrovo et al., 2021; Kpalo et al., 2020). An evaluation of residual forest biomass energy recovery through combustion and gasification confirmed that the combustion process returned 17% more energy. The study suggests that this advantage could increase up to 40% (Briones-Hidrovo et al., 2021; Carotenuto et al., 2022).

1.2. Research question and objectives

The scientific answers sought through this project were based on the following research question: *Is the combustion of residues from the high-pressure laminate and cannabis industry suitable for energy recovery? What is the impact of steam explosion pretreatment on cannabis pellets' properties and combustion?* To answer these questions, we aimed to evaluate the technical feasibility of using these solid residues for energy recovery by combustion. To achieve the goal of this study, the specific objectives were defined as described below:

- The residues were characterized by physical and chemical properties, including moisture content, elemental composition, calorific value, ash content, and thermogravimetric analysis.
- Production of pellets and characterization according to the properties specified by the CANplus certification.
- Construction and optimization of a laboratory-scale fixed-bed combustion system.
- Determination of the combustion operational parameters (temperature, air coefficient).

- Investigation of the profile of gas emissions during the combustion.
- Investigate the impact of steam explosion pretreatment on the pellets and combustion of cannabis residues.

1.3. Organization of the document

The study in this project is for the energy recovery of two types of industrial residues via combustion. The organization of this document was structured as follows: Chapter 1 contextualizes the current scenario of climate change that has been drawing attention to the impact of anthropogenic actions on the planet's climate through greenhouse gas emissions and their main contributors, landfill disposal, as well as the objectives of this work. Chapter 2 presents a literature review introducing the general aspects of greenhouse gas emissions, the current scenario of solid waste situation in Canada, and also the general aspects of the characteristics of the two types of waste studied in this work (High-pressure laminate and cannabis), the main thermochemical transformation pathways studied in the valorization of solid waste. Then, the pelletization strategy is used as an alternative for valorizing these residues. The next chapter, Chapter 3, was dedicated to the experimental results of the valorization of solid waste from the high-pressure laminates industry through combustion, with the objective of energy recovery and reduction of landfill disposal. Chapter 4 discusses the energy recovery of cannabis green solid waste for energy recovery by combustion and verifies the impact of pretreatment by steam explosion on the properties of pellets and on the combustion behavior and gas emissions. The Chapters 3 and 4 are presented in the form of scientific papers that were developed in the course of this work and submitted for publication in peer-reviewed journals. The final chapter was dedicated to the final conclusions obtained in this work.

2. STATE OF THE ART

2.1. Greenhouse gas emissions (GHG) current scenario

The concentration of GHGs has increased steadily since 1750 due to human activities. Each of the past four decades has been warmer than any decade since 1850. For the decade from 2011 to 2020, the globally averaged surface temperature was 1.09°C higher compared with the standard period from 1850-1900; more significant increases occurred over land and ocean (IPCC, 2021). These changes are already affecting the clime on the Earth, altering weather and climate, and this has brought extreme events to the planet such as heatwaves, heavy precipitation, droughts, and tropical cyclones (IPCC, 2021).

Faced with this crisis's scenario about the damage of anthropogenic actions in the climate change on the planet, ambitious measures for a significant global reduction and transition to a low carbon economy have been debated and applied worldwide. At least 35% of global emissions must be eliminated by 2030, 41–72% by 2050, and 78–118% by 2100, compared with the level in 2010. Taking these measurements are necessary to avoid the global temperature goes over 2ºC on average, considering pre-industrial levels and targets set by the Paris Agreement (Davis et al., 2018; Yokoi et al., 2022).

Burning fossil fuel sources has considerably contributed to GHG emissions, and it is a concern in major world countries (Gurney et al., 2022). The energy sector in Canada is the most contributor to CO_2 release. In 2022, the sector emitted 306 Mt or 82% of Canada's total GHG emissions. This corresponds to the Stationary Combustion Sources (306 Mt or 43%), Transport (196 Mt or 28%) and Fugitive Sources (75 Mt or 11) (National Inventory, 2024).

Canada is taking action to achieve GHG reduction. The country has a plan called the Pan-Canadian Framework on Clean Growth and Climate Change (2016), also the Strengthened Climate Plan (2020), Canada's enhanced 2030 target (2021), and the enactment of the Canadian Net-Zero Emissions Accountability Act (2021) to support the reduction of emission. Canada approached to reach a reduction of 40-45% below 2005 levels by 2030 and become net-zero emissions by 2050 (National Inventory, 2024).

The province of Quebec emitted, in 2021, 77.6 Mt_{eq}. CO_2 in GHG emissions. It represented 11.6 % of emissions in the country. Between 2020 and 2021, GHG emissions increased by 5.0% (MELCCFP, 2023). The Sector that most contributed to GHG emissions was the transport sector, with 33 Mt_{eq}. CO_2 or 42.6% of the emissions, followed by the industry sector with 25 Mt_{eq}. CO_2, or 32.3 % of the total emissions. The agriculture sector has 10.4% of total emissions. Residential, commercial, and institutional sector 9.1% of total emissions and the waste sector correspond to 5.2% of total emissions (MELCCFP, 2023).

For the province of Quebec, we observed that the Transport and Energy sectors are the biggest contributors to emissions. Together, they account for approximately 75% of the province's emissions. Therefore, changes made in these two sectors will significantly impact gas emissions reduction. The province of Quebec also has its own reduction target for the reduction of GHG (Y. Yao et al., 2023) emissions with the “Plan pour une économie verte 2030” (PEV 2030), which is Quebec's framework policy on electrification and climate change, and it is this plan that will help Quebec reduce its GHG emissions by 37.5% below the 1990 threshold by 2030. It also puts Quebec on a carbon-neutral path to 2050 (MELCCFP, 2024).

2.2. Solid waste current scenario

With economic development and the growing global population, waste generation has also significantly increased, and it is sent to landfills because it is cheaper (Yao et al., 2020). If not properly managed, this waste can cause serious damage to the environment and human health, in addition to contributing to global warming due to greenhouse gas emissions (Aibar-Guzmán et al., 2023). For this reason, managing this waste has become essential today. Waste management options include reusing, repairing, refurbishing, remanufacturing, repurposing, recycling, and composting. These actions are key components of a circular economy, reducing the environmental impact of waste. The concept of a circular economy focuses on using materials and resources for as long as possible, and after their full use, they can be diverted from landfills and reused in some way (Environment and Climate Change Canada, 2023).

In 2020, Canada generated 36 million tonnes of waste, with 72.5% ending up in landfills and only 27.5% being diverted (Environment and Climate Change Canada, 2023). The residential

sources were responsible for 52% of diverted solid waste and 42% of disposed solid waste. In both cases, the amount of waste increased from 2002 to 2020, where solid waste diverted increased by 85%, and waste disposed of increased by 29% (Environment and Climate Change Canada, 2023).

The non-residential sector, including industrial, commercial, institutional, construction, renovation, and demolition sectors, contributed to 43% of the diverted waste and 58% of the waste sent to landfills (Environment and Climate Change Canada, 2023).

Reducing and managing waste is a responsibility shared by the federal, provincial, territorial, and municipal governments in Canada. To address this problem, the Canadian government aims to reduce waste sent to disposal by 30% by 2030 compared to the 2014 baseline (Environment and Climate Change Canada, 2023).

In response to these challenges, there has been a push to develop waste-to-energy technologies to reduce waste volume and lessen environmental impacts (Chen, 2018).

2.3. High-pressure Laminates (HPL) production and waste

High-pressure laminate (HPL) consists of design surfacing material used for commercial and residential to provide an aesthetic appearance in interior design and architectural projects. The manufacturing process of these laminates involves using different papers and additives such as dyes, phenolic and melamine resins. Santos et al. (2021) proposed the manufacturing process for HPL. First, layers of kraft paper are impregnated in a phenolic resin bath (1st bath). Then, it follows a first drying phase, where the resin content attained in the paper is between 40% and 42%. Subsequently, the sheets are subjected to a second bath where resin content is between 54% and 56%, continuing the drying process in the same oven at the same temperature (Figure 2.1).

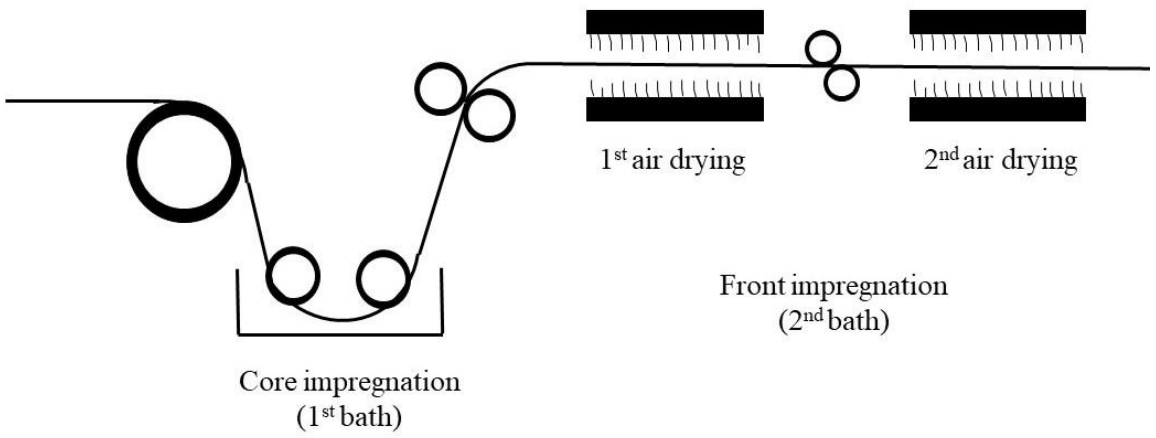

Figure 2.1 - Impregnation of phenolic resin in the kraft papers for HPL production (Santos et al., 2021).

Then, a core layer for the HPL is elaborated with kraft paper sheets impregnated with phenolic resin. A decorative paper is superimposed on the core layer, followed by a melamine paper (overlay), as shown in Figure 2.2. After this assembly, the laminate is pressed at temperature and pressure. The high-pressure process consists of simultaneous application of temperature ≥ 120°C and high-pressure specific pressure ≥ 5 MPa (Martins et al., 2015).

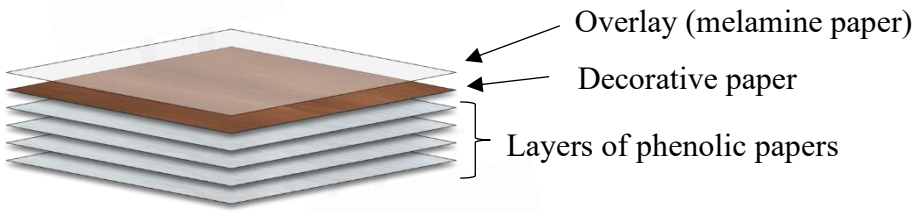

Figure 2.2 -. HPL composition. Adapted from (Haynes, 2017)

The waste generated from the HPL production processes is mainly paper and paper impregnated with resins used to produce the laminates. As the purpose of the study is to recover energy from

these materials, understanding their chemical composition and thermal degradation is a key knowledge of the process.

2.3.1. Chemical composition and thermal degradation of the constituents of the residues of HPL

Paper is made from raw materials containing cellulose fiber. Cellulose is a natural linear polymer composed of repeating units of β-(1→4)-D-glucopyranose with a high molecular weight (M. He et al., 2018; Roy et al., 2009). Due to its abundance, being a renewable and biodegradable polymer, cellulose is a promising raw material for producing papermaking, food, printing, cosmetics, oil well drilling, pharmaceuticals, etc. (Varshney & Naithani, 2011).

Despite being made of cellulosic compounds, papers are different from each other by chemical composition, primarily because of their production process. In Table 2.1, we can see the different chemical compositions of different papers and compare them with wood.

Table 2.1 – Comparison of the chemical content of cardboard, paper, and wood. Adapted from (Agarwal et al., 2014a).

Material	Cellulose (%)	Hemicellulose (%)	Lignin (%)	Other (%)	HHV (kJ/kg)	Reference
Cardboard	59.7	13.8	14.2	12.3	14.48	(Grammelis et al., 2009; Yáñez et al., 2004)
Office paper	64.7	13.0	0.93	21 .4	13.82	(Y. Chen et al., 2004; Grammelis et al., 2009)
Newsprint	48.3	18.1	22.1	11.5	15.87	(Y. Chen et al., 2004; Grammelis et al., 2009)
Pinewood	49.8	20.8	26.7	2.7	21.6	(Müller-Hagedorn et al., 2003; Viana et al., 2018)

Due to the paper's diversity of applications, its properties and characteristics will depend on its final application. During the paper production step, additives are added to the pulp. An example is the functional additives, which are included in the composition of the paper, such as fillers,

resins, dyes, and dry strength agents, that modify its properties (Peña & Aguilera, 2003). Table 2.2 shows the percentage of organic and inorganic composition of different paper types.

Table 2.2 – Organic and inorganic composition of different types of paper. Adapted from: (Goyal, 2022)

Type/Grade of Paper	**Organic**	**Inorganic**
1. Newsprint	>95%	<5%
2. Cardboard	>95%	<5%
3. Printing and Writing (P&W), copying & book paper	70-100%	0-30%
4. Tissue	>98%	<2%

The presence of these additives (inorganic material) in the composition of the paper has been a challenge for its reuse to generate heat since the presence of non-fiber additives, such as kaolin (Al_2O_3, SiO_2) and calcium carbonate ($CaCO_3$), increases the amount of ash generated in the process.

Ash deposition is a problem in combustion systems, as it is associated with fouling, slagging, equipment corrosion, and reduced energy efficiency**.** *Fouling* is a type of deposit formed in the convective zones of combustion systems, where the temperature is lower. These deposits can also be caused by the condensation of some volatile elements and by the impact of ash present in the flow of gases and on the surfaces of the exchangers. When compared to slagging, it has less sticking power. *Slagging* is formed in the combustion chamber zone where the temperature is higher. It is related to the smelt and sintering of elements on the walls of exchangers, which, due to its high adhesion power, can add other elements (Nunes, 2016).

The deposition of ash is affected by the chemical and physical processes such as ash chemical composition, distribution of mineral matter in ash, ash fusion temperatures, furnace temperature, ash particle temperature, the surface temperature of heat-exchanger tubes, tube materials, the flow field in the furnace, and ash transport mechanisms (Wieland et al., 2012).

The formation of deposits depends on the fuel and the system's temperature. For example, in coal combustion plants, the deposits are formed at high temperatures above 900°C, showing many amorphous phases, such as anhydrite, hematite, and silicates. In biomass combustion systems, the vaporization and condensation of elements composed of alkali metals, particularly

potassium, plays an essential role in the deposition process since it is present in the biomass in different forms, such as organometallics and salts, while silicon occurs primarily as hydrated silica grains (Horák et al., 2019).

According to G. Chen et al. (2022), the main methods used to minimize slagging and fouling in combustion have been washing with water, optimization of the operating parameters, blending different feedstocks, and screening additives. Among these methods, additives have been an economically more efficient option in minimizing the formation of slagging and fouling, as they act in modifying the properties of the melting point of the ash. According to the same authors, the common additives used to reduce the impact of slagging and fouling are aluminosilicates, kaolin, SiO_2, Al_2O_3, TiO_2, $(NH_4)_2SO_4$, $Al_2(SO_4)_3$, $Fe_2(SO_4)_3$, $(NH_4)_3PO_4$, H_3PO_4, $CaCO_3$, CaO, zeolites, etc.

G. Chen et al. (2022) studied different additives on the migration and transformation patterns of alkali/alkaline earth metals and chlorine during MSW incineration. The ash content of the sample was 22.45%, and its composition was Fe_2O_3, SiO_2, CaO, Al_2O_3, Na_2O, MgO, K_2O, Cl, SO_3. He used the following additives: $Al_2(NO_3)_3$, Ba $(NO_3)_2$, NH_4 H_2PO_4, Ca $(NO_3)2$, and Mg $(NO_3)_2$, which were diluted in deionized water. He found that Mg $(NO_3)_2$ > Ca $(NO_3)_2$ > Al $(NO_3)_3$, following this order had a positive impact on the ash fusion temperature. For instance, Mg $(NO_3)_2$ notably increased the initial deformation temperature, softening temperature, and flow temperature of ash from 1180ºC, 1190ºC, and 1240ºC to 1220ºC, 1230ºC, and 1260ºC, respectively.

In combustion equipment, this ash can affect the lifetime and operation, increase operating cost and carbon dioxide emission, deteriorate combustion behavior with higher combustion temperature, increase the production of nitrogen oxides and carbon monoxide, reduce heat transfers and cause corrosion (Radačovská et al., 2017). Other disadvantages of directly combusting paper residues are their irregular shape, their low energy density (Table 2.2) as well as transportation issues (Machinery, 2014).

Solving residual paper to generate heat into densified materials, such as pellets and briquettes, is a good option because it creates a regular shape for the papers, increases its energy density, and is easier to transport. When compared to other lignocellulosic biomasses, they can be easily

compacted and are likely to have lower nitrogen content than crop residues, which would lead to lower NO_x emission (Xiu et al., 2018).

The chemical compositions of papers will also affect the thermal decomposition and oxidation of these materials (Agarwal et al., 2014a). The thermal decomposition of paper is given as a function of time and heating rate. The gases involved in the decomposition of paper consist of H_2O, CO_2 and CO (Agarwal et al., 2014a).

Soares et al. (1995) studied the thermostability of a kraft paper sample in oxidizing and inert atmospheres. It was observed that air conditions seem to accelerate the thermal degradation of kraft paper, with the principal mass loss between 317 and 329ºC and maximum loss at 324ºC. Oxygen did not significantly affect the char yield, and it was rapidly oxidized over a narrow temperature range of 380 to 420°C, with a peak curve at 403°C.

Agarwal et al. (2014), studying cardboard pyrolysis and char oxidation, observed that the char formed at a temperature of 440ºC was more reactive than the char formed at temperatures up to 800ºC. However, it had a lower peak heat release rate when compared to char formed at higher temperatures. According to the authors, despite these differences, both chars had effective heat of combustion values of -32,860 kJ/kg and -34,070 kJ/kg, respectively. For the thermal oxidation, there was an initial peak of mass loss due to the pyrolysis of the hemicellulose and cellulose, corresponding to the most significant mass loss and heat release rate at 335ºC. The adequate heat of combustion for the thermal oxidation of cardboard was determined to be -16,200 kJ/kg.

Singh & Namjoshi (2021) compared the thermal decomposition in an N_2 atmosphere of newspapers, cardboard and glossy paper and observed a maximum decomposition in newspapers and minimum decomposition in glossy paper, probably due to the presence of more clay in this type of paper. The peak temperature of a newspaper was 353.7ºC (57% of mass loss), and that of glossy paper was 341.1ºC (33% of mass loss), and the decomposition of this paper was relatively slow. The cardboard performed between these values with a peak temperature of about 335ºC and mass loss of 53%.

According to Galiwango & A.Gabbar (2022), studying co-pyrolysis of municipal paper residues observed a shift in thermal analysis (TA) curves towards the higher temperature side with

increased heating rates. According to the same author, the curve shift on the right-hand side was due to the rise in the thermal energy in the pyrolysis chamber. In the thermogravimetric analysis (TG), the sample showed a decreased mass loss with increased temperature and short reaction time due to an increased heating rate. In the same study, the author observed that higher activation energy was necessary for polypropylene residues than for paper. For example, polypropylene needed 187.25 kJ/mol in a "coats-redfern" model, and paper needed 159.25 kJ/mol. It was assumed that the aliphatic compound and aromatic rings in the sample increased the energy needed to break inherent chemical bonds. Then, the study observed in the same model that, in a mixture of residues (paper and polypropylene), lower activation energy was necessary (143.74 kJ/mol), than in the homogeneous residues, showing a positive energy indicator for pyrolysis of mixed polymer-lignocellulosic residues found in many municipal solid residues. Polypropylene is a material with aromatic rings in your chemical structure, like the residues present in this work with phenol-formaldehyde.

Phenol formaldehyde resin has been widely used since its first formulation in the 1900s as a replacement for shellac and is one of the earliest polymers adapted for use in industry (Purohit & Orzel, 1988; Zhou et al., 2022). Due to its strong acid resistance, good thermal resistance, and immaculate mechanical properties, it has been used as an anti-corrosion and flame-retardant material and as an adhesive (Mougel et al., 2019).

The thermal degradation behavior of resins depends on the atmosphere (inert or oxidizing) and the temperature. Bobrowski et al. (2018) show that low molecular compounds are released in the initial stage of resin degradation without a thermal oxidation process. Then, the decomposition and bridging of methylene groups make release phenol, and cresol homologue. In the next and final step, total degradation of phenol structure takes place.

According to Bouajila et al. (2003), in their studies of the degradation of resins in an oxidizing atmosphere, the degradation began around 380°C. The most significant mass loss (≈ 70%) occurred between 400 and 500°C, and the total mass loss was at 600°C. In temperatures below 300°C, mass losses are considerably low, proving that the resin exhibits good thermal stability up to this temperature in any atmosphere (inert or oxidizing). The author also observed that degradation in the air mainly produces low molecular weight compounds.

In thermal degradation of phenolic compounds, most of the gases produced are CO, CO_2, CH_4, and H_2O (Bouajila et al., 2003). Smaller quantities of H_2, formaldehyde and other volatile organic chemicals (including phenol, methyl phenols, and dimethylphenols) also appear to be produced. Even though in fewer quantities (>0.1% by weight), additional products during the combustion process can be produced, probably due to the addition of blowing agents and fillers to the phenolics (Purohit & Orzel, 1988).

Torres-Herrador et al. (2021), in their studies on the pyrolysis of phenolic resins, they have observed several permanent gases such as H_2, CO, CO_2, H_2O, CH_4, C_2H_4, C_2H_6, NH_3, C_2H_3N, and CH_2O. Phenol and similar compounds were the major products. Large molecular weight compounds were also observed, in most cases diphenols, naphthalenes and three fused rings (anthracene).

Regarding the volatile compounds produced in an oxidizing atmosphere, water is released between room temperature and 530°C, and the amount released depends on the crosslinking cycle of the resin. The water is released in two steps. The first, between room temperature and 130°C, corresponds to small amounts of chemically bound or adsorbed water. The second stage, which takes place between 130 and 530°C, is the exit of the compounds from the condensation residue, between 130°C and 180°C, followed by the degradation of polyoxymethylenes, oligomeremiacetals and the phenolic network at temperatures higher than 180°C (Bouajila et al., 2003).

Torres-Herrador et al. (2021) also found similar results in the pyrolysis products. At temperatures below 550ºC, water and phenols are the most relevant products. Between 550ºC and 700ºC, CO, CO_2, and H_2 gain importance. At temperatures higher than 750ºC, H_2 became the most important product.

Loss of formaldehyde occurs in small quantities between 80-200°C and increases between 200-310°C. Methanol is released between 60 and 310°C. The formation of carbon monoxide occurs between 200 and 600°C, and the formation increases between the temperatures of 320 and 600°C due to the decomposition of the phenolic network. Carbon dioxide is formed between 140 and 660°C with a rapid increase from 350ºC up to 600ºC (Bouajila et al., 2003).

(Bobrowski et al., 2018) presented a hypothesis of the mechanism of the thermal-oxidation process in the initial phase (Figure 2.3).

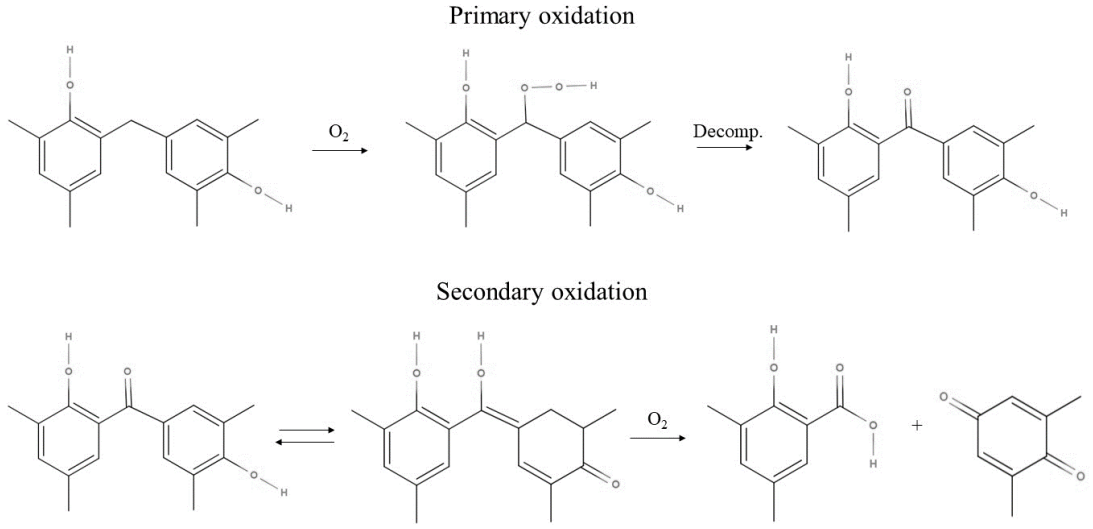

Figure 2.3 - Thermal degradation of phenolic resin at a low temperature due to thermo-oxidation. Adapted from (Bobrowski et al., 2018).

In high temperatures, the resin can act as a source of oxygen for the oxidation process. The oxygen may be a source to form carbonyl bonds. The initial stage of the degradation releases low molecular compounds without a thermo-oxidation process. Then, an emission of phenol, cresol, and homologue occurs due to the decomposition and bridging of methylene groups. Peroxide intermediates may be produced (Bobrowski et al., 2018). Considering the chemistry of this system, it was deduced that two further chemical processes are possible to form carbonyl-containing fragments further. The first is the cleavage to acid fragments, and the second is the rearrangement to quinoid-type structures. Previous research has shown that sterically hindered phenols can be oxidized in quinone-type structures. It has also been reported that quinone-type structures are not formed if the hydrogens are located at the α position of the aromatic ring. Once the carbonyl structure is formed from the methylene group, the conditions of steric hindrance and absence of α hydrogens are fulfilled, supporting the quinoid intermediates in the secondary oxidative processes. In the secondary oxidation, phenolic hydroxyl decreases, and acid hydroxyl appears. (Conley & Bieron, 1963; Hardrict et al., 2003; Bobrowski et al., 2018).

2.4. Cannabis waste industry

The cannabis industry is a valuable source of biomass resources for implementing sustainable second-generation biorefineries. The waste generated from the utilization process of the *Cannabis sativa* L. plant is considered to have low added value but can play a key role in bioenergy production (Moscariello et al., 2021).

The global cannabis market has grown rapidly and is expected to reach a worldwide valuation of 40.6 billion dollars in 2024. Canada was the second country to legalize marijuana, with an expected growth of CA$ 6.5 billion by 2025 and CA$ 10.5 billion by 2030, growing at a Compound Annual Growth Rate (CAGR) of 16% between 2020-2030 (Cannabis Business Plan, 2024). With increased production, there is also an increase in waste generated by production. A typical cannabis production cycle can vary from 3 to 5 months, starting from cutting clones to harvest, with indoor production being faster than outdoor production (Upland Agricultural, 2019).

In cannabis production, solid waste is generated, and they are categorized as green, non-green, or hazardous waste (Upland Agricultural, 2019). The green plant material includes the cannabis flowers, trim, leaves, stalks, and roots (Upland Agricultural, 2019). Once the plant is ready for harvest, it is entirely removed from the growing area. Individual plant branches are removed from the stalks, and flowers are cut from the branches and dried (Upland Agricultural, 2019). Producers have indicated a large amount of green waste generated over the cannabis crop cycle. As the demand and annual production of cannabis increase, it is estimated that 70-75% of the output is process residue, amounting to approximately 10.5 t/ha of green waste (Kraszkiewicz et al., 2019). This waste has been incinerated onsite or in offsite disposal, which contributes to global warming (Butsic & Brenner, 2016).

In terms of composition, the stem of cannabis contains two types of fiber: short inner fibers known as hurd and outer fibers known as bast. They differ in composition (Gomez et al., 2021). Although they have equivalent diameters in the structure of the hemp stem, hemp bast is rich in cellulose, while hurds contain less cellulose and more lignin (Gomez et al., 2021). When compared to other important industrial biomasses, hemp has a higher cellulose content and lower lignin content than corn stover and sorghum bagasse, for example (Zhao et al., 2020).

Table 2.3 compares cannabis with other biomasses, showing that hemp has high cellulose content, suggesting a good energy density with a comparable high heating value (HHV) and competitive ash content with the other biomass feedstock (Parvez et al., 2021). When used as solid fuel, hemp performs better when harvested between February and April when the moisture content is lower. Its energy yield is high, around 100GJ/ha/y (Brar et al., 2024; Parvez et al., 2021).

Table 2.3- Chemical composition, HHV and ash content of biomass feedstock. Adapted from (Parvez et al., 2021) and (Brar et al., 2024).

Biomass	**Cellulose (%)**	**Hemicellulose (%)**	**Lignin (%)**	**HHV (MJ/kg)**	**Ash content (%)**
Hemp	57-77	9-14	5-9	18-19	1.5-8.5
Spruce wood	46.7	29	26.2	19-20	0.5-2.5
Miscanthus	50	25	12	18-19	2.0-7.0
Willow	44.4	31	25.5	19.5	1.3
Reed canary grass	35.9	29.6	5.0	17-19	3.0-8.5

The federal government controls cannabis waste management in Canada, and the waste management strategies include incineration, where the solid parts of cannabis, such as branches, leaves, and bush, are destroyed; composting, which can be done onsite or transported to authorized locations; and aerobic or anaerobic digestion, which is considered more advantageous as it does not include transportation costs (Brar et al., 2024).

The legislation in Canada, described in the Cannabis Act, which was established in October 2018 for the control of cannabis sales and production, states that, according to Schedule 2 to the Act, all parts of the plant that do not present significant levels of phytocannabinoids are exempt from the application of the Act. These parts include "non-viable seeds; mature stalks without any leaf, flower, seed, or branch; fibre; and roots". According to the regulation, these parts do not pose a risk to public health and public safety (Government of Canada, 2024b). Given this, these parts can be used in technologies such as energy recovery.

2.5. Use of waste as an energy source

In recent years, interest in replacing fossil fuels with lower carbon-intensity options has increased (Tauro et al., 2018). Biofuels have been studied widely to reduce dependence on fossil fuels and have been a promising alternative to mitigate climate change (Jeswani et al., 2020).

Biofuel is made from biomass. Biomass can be defined in different ways and in different fields of application. It refers to any non-fossil biological material, such as crops, crop residues, plants, algae, animal residues, food residues, and yard residues, as appropriate biomass feedstock (Abdoli et al., 2018).

Different methods of conversion technology are used for biomass for biofuel generation, and they can be divided into two groups: thermochemical conversion and biochemical conversion (Figure 2.4) (Perera et al., 2021).

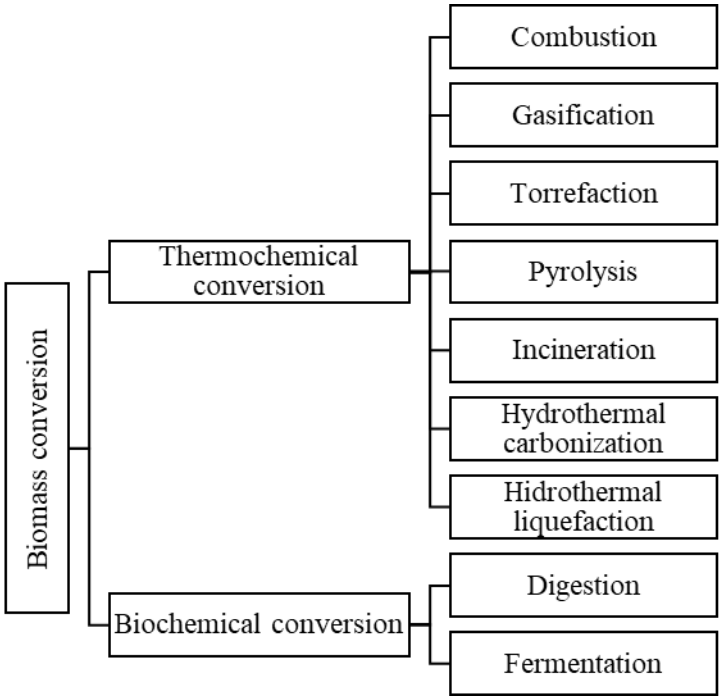

Figure 2.4 - Biomass thermochemical and biochemical conversion process. Adapted from (Perera et al., 2021)

Combustion of biomass is a thermochemical conversion process that degrades biomass into low molecular products through physico-chemical reactions. These successive reactions happen in

the presence of fuel, air and heat. When all these three requirements are available, in a correct proportion, the combustion is self-sustaining (L. J. R. Nunes et al., 2014; Perera et al., 2021; Williams et al., 2012). Esteban et al. (2014) have shown in their study the environmental benefits of using biomass instead heating oil as fuel for residential heating boilers. For some techniques, such as combustion of the biomass, an additional process or pretreatment might be required to improve biofuels' quality. Its direct use may be less effective due to the physical and chemical properties of each biomass, such as moisture, hydrophilic nature, low calorific value, poorer grindability, heavy metals, etc. (Bach & Skreiberg, 2016; Perera et al., 2021).

For example, biomass absorbs nutrients for its development during growth, including some metals. These metals, associated with some hydrocarbon bonds, may influence different aspects of the thermochemical conversion process, such as the calorific value, the corrosiveness of the flue gas, the stickiness of the ash, etc. (Mandø, 2013; Marangwanda et al., 2020). Ash deposition is a problem in solid biomass combustion. Once the accretion of inorganic matter on superheater tubes and boiler walls reduces efficiency, leads to downtime and prolongs maintenance periods in biomass-fueled power plants. High amount of ash contents causes slagging, fouling and corrosion (Plankenbühler et al., 2019).

2.5.1. Use of technologies for waste valorization for energy recovery

The efficiency and economics of waste valorization will depend on its composition, moisture content, and contaminants (Bhatt et al., 2021). The most commonly used technologies for the thermal conversion of solid waste are pyrolysis, gasification, and combustion (Bhatt et al., 2021; Kpalo et al., 2020).

Pyrolysis is a process that involves the thermal decomposition of material in the absence of oxygen, producing primarily biochar, bio-oil, and syngas (Lu et al., 2020). Hasan et al. (2021) studied converting municipal solid waste by pyrolysis, and they found an overall production yield of around 43% bio-oil, 27% biochar, and 25% syngas. When applied to papers, the bio-oil yield was 52% at 400ºC, but the bio-oil quality was very poor, with many hydrocarbon compounds (Hasan et al., 2021). Li et al. (2020) have mentioned the unsatisfactory quality of fuels from non-catalytic pyrolysis of municipal solid waste. Su et al. (2022) showed that co-

pyrolysis of microalgae and biomass enhances overall pyrolysis performance and bio-oil properties due to synergetic effects between the materials. Considering the emission, the authors mentioned that the system should be equipped with an emissions control system to make the process environmentally friendly (Hasan et al., 2021).

Quan et al. (2022) considered combustion and gasification reliable and feasible methods in response to concerns about landfilling. Gasification is a conversion process alternative to combustion that converts solid residues into syngas (CO, CO_2, H_2 and CH_4) through a reaction between the solid residue and the gasification agent (air, oxygen or steam water) at high temperatures (700ºC-900ºC) (Briones-Hidrovo et al., 2021). The yield and composition of syngas depend on the gasification agent. (Carotenuto et al., 2022). The syngas obtained from the process must be cleaned before use in subsequent applications, such as hydrogen production, methanol synthesis, liquid-fuel synthesis, and power generation (Shahabuddin & Alam, 2022).

Coal has been the dominant fuel used to produce syngas, accounting for 84%, while biomass represents just 1% (Shahabuddin & Alam, 2022). According to Carotenuto et al. (2022) the gasification process regarding energy and environmental performance has not been extensively studied in the literature. (Briones-Hidrovo et al., 2021) summarized biomass-based gasification case studies and concluded that gasification will vary widely depending on geographic location, biomass type, final product, and electrical efficiencies. Paengjuntuek et al. (2015) used rice straw in gasification to produce electricity as the final product and found 59% efficiency. However, it had a high emission of 864g CO_{2eq}/KWh_e when compared, for example, to wood chips, which had 30% electrical efficiency and 90g CO_{2eq}/KWh_e (Siegl et al., 2011).

Nguyen et al. (2013) comparing gasification and the combustion of straw as feedstock, it was found that gasification seems to be a more environmentally friendly option due to the disadvantage of gases emitted during the combustion process. Siegl et al. (2011) made the comparison of combustion and gasification of wood chips for electricity production, and gasification performed better in just 2 out of 11 impact categories in a life cycle assessment of their work.

In the study of Briones-Hidrovo et al. (2021) they evaluated the assessment and energy recovery of residual forest biomass by combustion and gasification, and gasification performed better in 5 out of 8 impact categories; however, the combustion process returned 17% more energy than

gasification. It shows the importance of different aspects in the thermal valorization of a feedstock, taking into consideration the feedstock properties when applied for energy recovery, such as the feedstock composition, technology and process parameters to generate heat or electricity.

Another technology for energy valorization is combustion. Combustion is the most widely used technology for producing heat and electricity from biomass. It presents advantages over other technologies because it is well-known and has a well-established infrastructure. In addition to being highly efficient in converting biomass into energy, it can accept a variety of feedstocks, and the production of energy and heat is done through direct use (Kpalo et al., 2020).

Combustion is an exothermic chemical reaction that involves the complete oxidation of the fuel, accompanied by heat release to the external environment. Combustion is considered completed when the entire carbon chain of the fuel is oxidized, and products such as water and carbon dioxide are formed (Eq. 2.1, Eq. 2.2) (Turns, 2014).

$$C + O_2 \rightarrow CO_2 \qquad \Delta H^O_{298} - 283\ kJ/mol \qquad \text{(Eq. 2.1)}$$

$$H + \frac{1}{2}\ O_2 \rightarrow H_20 \qquad \Delta H^O_{298} - 242\ kJ/mol \qquad \text{(Eq. 2.2)}$$

However, when combustion is incomplete, it is said that the oxygen supply was insufficient to consume all the fuel and CO is formed (Eq. 2.3) (Turns, 2014).

$$C + \frac{1}{2}\ O_2 \rightarrow CO \qquad \Delta H^O_{298} - 111\ kJ/mol \qquad \text{(Eq. 2.3)}$$

The overall equation for a combustion reaction in an air atmosphere is given by the equation below (Eq 2.4):

$$C_{x_1}H_{x_2}O_{x_3}N_{x_4}S_{x_5} + \eta.(3.76N_2 + O_2) \rightarrow aCO_2 + bH_2O + cO_2 + dN_2 + +eCO + fCH_4 + fNO_x + gSO_x \qquad \text{(Eq. 2.4)}$$

where η is the theoretical and stoichiometric amount of air for the combustion of the material (Mandø, 2013) .

Moreover, there is a chemical reaction involving carbon dioxide and carbon that plays a key role in the combustion process, especially in systems where solid carbon is burned. This reaction is called Boudouard reaction (Eq.2.5), which can contribute to the formation of CO in the system (Chien & Chuang, 2011).

$C + CO_2 \rightleftharpoons 2CO \qquad \Delta H_{298}^{O} - 172.5\ kJ/mol$ (Eq. 2.5)

Other possible reactions that may occur at the surface of the carbon particle, according to Dorokhov et al. (2021) are:

$2C + O_2 \rightarrow 2CO \qquad \Delta H_{298}^{O} - 221.0\ kJ/mol$ (Eq. 2.6)

$C + H_2O \rightarrow CO + H_2 \quad \Delta H_{298}^{O}\ 131.3\ kJ/mol$ (Eq. 2.7)

$C + 2H_2 \rightarrow CH_4 \qquad \Delta H_{298}^{O} - 74.8\ kJ/mol$ (Eq. 2.8)

Any conventional fuel requires a stoichiometric amount of oxygen according to its chemical composition to theoretically achieve complete combustion. We say that the theoretical volume of air is the amount necessary for the total combustion of the fuel (Marangwanda et al., 2020).

When this air supply parameter is equal to one (α =1), it means the exact stoichiometric air supply needed for the complete combustion (Abdoli et al., 2018). However, it is necessary to administer a volume of air higher than the stochiometric value for complete combustion of the fuel due to the difficulty of guaranteeing a perfect mixture of the air with the fuel and the limited time of permanence of this in the combustion chamber (Caposciutti et al., 2018; Huang et al., 2021). This amount of excess air is called the coefficient of excess air, which in this work will be represented by the Greek letter α.

According to Silva Lora & F. Happ, (2002) the optimum value of excess air is calculated as a function of the type and characteristics of the fuel. In general, in modern combustion systems, the air coefficient (α) takes the following values:

Solid fuels: 1.15 - 1.30 / Liquid fuels: 1.05 - 1.10 / Gaseous fuels: 1.02 - 1.05

It is very important to find the optimal α values since a value beyond the necessary may represent excessive electrical energy consumption in the fans due to the excess volume of air and gases

moved by these fans. In addition, the excess air is heated, which constitutes an additional heat loss. An insufficient amount of air leads to incomplete combustion and the formation of incomplete combustion products, such as CO, CH_4, H_2 (Silva Lora & F. Happ, 2002).

The emission of polluting gases must be considered when implementing combustion technology since the combustion of solid residues can lead to the emissions of contaminants. Contaminants are all the gases and materials emitted by the combustion chamber that alter or generate disturbances to the natural balance of the environment (Signoretti, 2008). Nitrogen oxides, carbon monoxide, organic compounds (unburned or partially burned hydrocarbons) and sulphur oxides are the four main chemical classes of pollutant species considered in combustion processes (Mandø, 2013).

CO and hydrocarbons arise from the incomplete combustion of fuel due to inadequate air mixing or the fact that the fuel is rich in hydrocarbons. Both can be oxidized to carbon dioxide and water with sufficient residence time and air (Signoretti, 2008). Malaťák et al. (2017) have found that under conditions of low excess air coefficient ($\alpha<2$) on the combustion of Timothy grass, incomplete combustion occurred, and high concentrations of carbon monoxide (CO) were found. Conversely, with a higher excess air coefficient ($\alpha>3$), a rising concentration of carbon monoxide emissions occurred. The authors attributed these results to the flue gas's low temperature and premature cooling.

Xiu et al. (2018) burning paper briquettes in a small-scale heating stove also found high concentrations of CO emission in combustion. The authors attributed this result to the bulk density of the paper briquette used since the briquettes maintained their shapes during combustion, which could restrict air penetration and result in higher CO emissions. A faster burning rate and higher combustion temperature of residue paper briquettes also could favor the formation of NO_x.

The nitrogen in the air or the one contained in the fuel can be converted into pollutants such as NO, NO_2, N_2O, NH_3 and HCN. The pollutant species formed depended mainly on the combustion zone's temperature and fuel/oxygen ratio. NO and NO_2 are the most emitted nitrogen components in the combustion process. Typically, the amounts of NO formed are higher than those of NO_2. NO_2 tends to occur in the region with rapid cooling, for example, in

the mixture of hot combustion gases with the air inlet. N_2O, on the other hand, can be formed by a large number of reactions in combustion gases, but it reacts quickly with H and OH radicals to form N_2 (Asghar et al., 2021).

It is possible to reduce NO_x emissions by acting on the factors that determine its formation, particularly on the maximum temperature and oxygen concentration in the ovens where the combustion process occurs. According to Asghar et al. (2021) the leading technologies for controlling NO_x emissions are described in Table 2.4

Table 2.4 – Mainly corrective technologies for controlling NO_x emissions. Adapted from Asghar et al. (2021).

Type of method	Name of the method	Characteristic	Efficiency	Cost
Corrective	Selective catalytic reduction	Reduction of NO_x to N_2 by ammonia injection using catalysts	70-90%	High cost in investment
	Non-selective catalytic reduction	Reduction of NO_x to N_2 by ammonia injection without the use of catalysts	30-80%	Low capital investment

Also, due to the presence of phenolic compounds in some solid residues in this work, the combustion of this material can generate polycyclic aromatic hydrocarbon (PAH) compounds that can contaminate the atmospheric air (Bouajila et al., 2003). In thermal degradation studies of phenolic compounds, most of the gases produced are CO, CO_2, CH_4, and H_2O (Bouajila et al., 2003). Smaller quantities of H_2, formaldehyde and other volatile organic chemicals (including phenol, methyl phenols, and dimethylphenols) also appear to be produced. Torres-Herrador et al. (2021), in their studies on the pyrolysis of phenolic resins, have observed several permanent gases such as H_2, CO, CO_2, H_2O, CH_4, C_2H_4, C_2H_6, NH_3, C_2H_3N, and CH_2O. Phenol and similar compounds were the major products. Large molecular weight compounds were also observed, in most cases diphenols, naphthalene and three fused rings (anthracene) (Torres-Herrador et al., 2021).

Z. Zhang et al. (2021) cites four methods for controlling PAHs in the flue gas: adsorption, catalytic oxidation, photochemical degradation, and low-temperature plasma method. Among these methods, catalytic oxidation is the most widely used.

- Adsorption method – in this method, they use adsorbents to adsorb organic pollutants in the flue gas. Pollutants are diffused into these adsorbents, and adsorption can be carried out in fixed beds, fluidized beds and other types of reactors. Some examples of absorbents that can be used are activated carbon, zeolite, and alumina. However, this method usually produces waste adsorbents and secondary pollution and requires further treatment of adsorbents.

- Catalytic oxidation method – for the oxidation of the PAHs, catalyst preparation is the crucial process to oxidize PAHs into carbon dioxide and water. The most common supports used in the catalytic oxidation of organic compounds are Al_2O_3, zeolites, and granular activated carbons, and active components are metals, including noble metals (Pt, Pd, Rh, and Au) and base metals (Mn, Co, Cu, Fe, and Ni) (Lin et al., 2011).

- Photochemical degradation method – for this method, PAHs and derivatives can be degraded by ultraviolet irradiation due to increasing the light intensity, water vapour concentration and temperature which can accelerate the photochemical degradation reaction rate and improve the degradation efficiency of PHAs.

- Low-temperature plasma method - Free radicals (N^+, N_2^+, O^+, O_2^+, and NO^+) ionized by ionizing radiation in flue gas can react with PAHs.

Despite gas emission being a concern in the combustion of some feedstock, mainly those with some contamination, as some of the feedstock in this study, the use of thermal conversion technologies for waste management might be a good alternative to reduce landfill disposal, increase waste recycling and achieve energy recovery. Moreover, it can contribute to reducing environmental risks and support the model of the circular economy that has been the focus of having more environmentally friendly processes in the economy (Nanda & Berruti, 2021).

2.6. Pellet production

The combustion process accepts the direct burning of fuels; however, with today's technologies, the direct combustion of these materials is not economically interesting due to the low bulk density, high transportation costs and high mass loss during transportation (Abdoli et al., 2018).

According to Abdoli et al. (2018) pellets are defined as densified cylindrical material produced by compression and it is considered a traded commodity, used for producing heating for residential appliances and electricity generation in large power plants.

There are some specifications to follow to guarantee that pellets have good properties for their use. In the world, several countries have their own standard. In Canada, there is CANplus certification, which provides the framework of wood pellet properties given to producers to guarantee the quality of wood pellets to the end consumers. CANplus currently employs the identical set of parameters as the ENplus certification, according to Table 2.5 (Canadian Biomass, 2013).

Table 2.5 – Key specifications of graded wood pellets based on the CAN/CSA-ISO 17225 Part 2 Standard (Canadian Biomass, 2013).

Property Class	**Unit**	**Grade A1***	**Grade A2***	**Grade B***
Diameter, D	mm	6 ± 1 or 8 ± 1	6 ± 1 or 8 ± 1	6 ± 1 or 8 ± 1
Length**, L	Mm	3.15 ≤ L ≤ 40	3.15 ≤ L ≤ 40	3.15 ≤ L ≤ 40
Moisture, M	% of weight	≤ 10	≤ 10	≤ 10
Ash, A	% of weight	≤ 0.7	≤ 1.2	≤ 2.0
Durability, DU	% of weight	≥ 97.5	≥ 97.5	≥ 96.5
Fines Content, F	% of weight	≤ 1	≤ 1	≤ 1
High Calorific Value, Q	MJ/kg	≥ 18.6	≥ 18.6	≥ 18.6

*Suitable for residential and commercial applications.
**Maximum length of wood pellets shall be ≤ 45 mm. Number of pellets longer than 40 mm can be 5% weight. Further restrictions may be stipulated by de supplier of the combustion equipment regarding ash characteristics of the pellets, such as ash melting temperature, to minimize damage to equipment.
CAN*plus* (A1): is the top quality used in private domestic boilers or stoves.
CAN*plus* (B): is the grade used for industrial pellets.

Table 2.4 shows the pellet properties required for CANplus. Grade A is used for residential and commercial applications, and Grade B is used for industrial applications. These parameters are essential in the pellet's quality. The diameter and length give the pellets a better structure so that they do not break too easily and will not release fines, which would lead to a loss in weight of material for combustion. Also, fines burn faster, and they can be an explosion hazard for material storage and transport (Abdoli et al., 2018)

Moisture content is a parameter that can help in the material's densification and the pellets' durability. Increasing the moisture content of feedstock and optimizing this parameter can contribute to increasing the strength and forming a denser and more stable pellet. If the moisture content is in excess, the combustion can be affected because there will be a waste of heat for vaporizing the moisture content present in the pellet before starting the combustion, affecting efficiency. Vaporization uses energy released from the combustion to lower the temperature in the combustion chamber, which slows down the combustion process as the material needs heat to evaporate the water and subsequently heat the water vapour (Abdoli et al., 2018). These results were found by Biswas et al. (2014) in the combustion of a single Pine wood pellet at two different moistures, (1% and 12%), and the high moisture content required a longer combustion time.

Ash is also limited for quality combustion as they can form clinkers, cause uneven combustion of pellets, and increase stove maintenance. This factor is affected by the chemical composition of the material, especially calcium, magnesium, potassium, and sodium (Plankenbühler et al., 2019; Radačovská et al., 2017). Ash content was related to a lower burning rate in the experiments of Sun et al. (2016) in the combustion of municipal solid waste in a fixed-bed reactor and a higher emission of CO in the study of Xiu et al. (2018) on burning waste paper briquettes.

3. EVALUATION OF TECHNICAL FEASIBILITY OF INDUSTRIAL PAPER WASTE PELLETS FOR ENERGY RECOVERY BY COMBUSTION

3.1. Avant-propos

Auteurs et affiliations:

Rafael Eloy de Souza: PhD Candidate, Université de Sherbrooke, Faculté de génie, Département de génie chimique et génie biotechnologique.

Eduardo Lins de Barros Neto: Professor titular, Universidade Federal do Rio Grande do Norte (Brésil), Département de génie chimique.

Jean-Michel Lavoie: Professor titular, Université de Sherbrooke, Faculté de génie, Département de génie chimique et génie biotechnologique.

Bruna Rego de Vasconcelos: Professor agrégrée, Université de Sherbrooke, Faculté de génie, Département de génie chimique et génie biotechnologique.

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Titre en français: Évaluation de la faisabilité technique des pellets de résidus industriels de papier pour la valorisation énergétique par combustion.

Contribution au document: This article explores the technical feasibility of valorizing solid waste from the high-pressure laminate manufacturing industry through energy recovery via combustion. The study addresses two critical sectors —energy and waste—when the world is focused on solutions to mitigate climate impacts. The search for alternatives for reducing dependence on non-renewable resources and improving waste management practices has garnered significant attention in recent years. This work investigates the potential of solid waste

valorization as an alternative for energy generation, analyzing the combustion parameters relevant to the application of these residues.

Résumé français: Le traitement des résidus solides et leur mise en décharge restent des défis pour le développement d'une société durable. La valorisation énergétique des résidus pourrait être une option pour la production de chaleur ou d'électricité par combustion. Cette étude a examiné la faisabilité technique de la valorisation énergétique des résidus industriels de papier provenant de l'industrie des stratifiés haute pression (HPL) par combustion contrôlée dans un réacteur à lit fixe afin d'identifier les conditions optimales pour maximiser la récupération d'énergie. Les papiers ont été classés en trois groupes : phénoliques, non phénoliques et mélange, et ils ont été granulé avant la combustion. Le mélange de pellets a été priorisé dans cette recherche avec l'idée d'une utilisation totale des résidus. Les pellets répondaient aux exigences du standard CANplus, à l'exception de la teneur en cendres. La résine phénolique a contribué à augmenter la teneur énergétique des pellets de papier. L'efficacité de la combustion a été maximisée à une température de combustion de 300°C et un coefficient d'air de 3,0 avec le dégagement de chaleur le plus élevé. Le mélange de papier phénolique avec du papier non phénolique a positivement influencé la température d'ignition des pellets, entraînant un dégagement de chaleur plus élevé. Des coefficients d'air plus élevés ont entraîné une diminution des niveaux de CO et une augmentation des niveaux de CO_2 ; cependant, des émissions de CO ont été observées lors de la combustion des trois groupes de résidus de papier. Dans l'ensemble, les résultats ont suggéré que l'utilisation des résidus de papier phénolique peut être techniquement viable pour la valorisation énergétique par combustion, et qu'elle peut être compétitive avec certaines sources de biomasse, comme le bois, pour la récupération d'énergie.

Mots-clés : Résidus de papier ; valorisation énergétique; combustion; participation de partenaire industriel

3.2. Abstract

Solid waste treatment and landfilling are still challenges to developing a sustainable society. Energy recovery from waste could be an option for heat or electricity production through combustion. This study investigated the technical feasibility of energy recovery of industrial paper residues from a High-Pressure Laminate (HPL) industry through controlled combustion in a fixed-bed reactor to identify optimal conditions to maximize energy recovery. The papers were categorized into three groups: Phenolic, non-phenolic, and mixture, and they were pelletized before combustion. The pellet mixture was prioritized in this research with the idea of total use of the residues. Pellets met CANplus requirements except for ash content. Phenolic resin contributed to enhancing the energy content in the paper pellets. Combustion efficiency was maximized on a combustion temperature of 300°C and an air coefficient of 3.0 with the highest heat released. The mixture of the phenolic paper with non-phenolic paper positively affected the ignition temperature of the pellets, resulting in a higher heat release. Higher air coefficients resulted in lower CO and higher CO_2 levels; however, CO emission was presented in the combustion of the three groups of residues paper. Overall, the results suggested that using phenolic paper residues can be technically feasible for energy recovery by combustion, being competitive with some biomass sources, like wood, for energy recovery.

Keywords: paper residues; energy recovery; combustion; industrial partner.

3.3. Introduction

Urbanization and economic development are globally rising and have significantly altered the global energy scenario and waste production (Kosai & Unesaki, 2020; Shah et al., 2021). Fossil fuel has represented more than 80% of the energy source in homes and industries and has considerably contributed to greenhouse gas (GHG) emissions (Gurney et al., 2022; Samrot et al., 2023). In 2022, Canada's total GHG emissions were 708Mt of CO_{2eq}, and the energy sector emitted 577 Mt CO_2 eq or 82% of the total emissions (Government of Canada, 2024a).

Greenhouse gas emission is also linked to a poor waste management that can lead to environmental damage and affect the climate (Aibar-Guzmán et al., 2023). For waste

management, landfilling is still the least expensive option for dealing with waste (Y. Yao et al., 2023). In Canada, most of the solid waste ends up in landfills. In 2020, 36 million tonnes of waste were generated, with only 27.5% being diverted and the remaining 72.5% sent to landfills (Environment and Climate Change Canada, 2023). The non-residential sector, including industrial, commercial, and institutional areas, as well as construction, renovation, and demolition sectors, accounted for 43% of diverted waste and 58% of solid waste sent to landfills. In the light of this, the Canadian government aims to reduce landfilling by 30% under the 2014 level by 2030 (Environment and Climate Change Canada, 2023).

To reduce landfill disposal, efforts to find a way to decrease this number have focused on developing and applying waste-to-energy technologies. These technologies could help not only reduce landfill disposal but also it could help to minimize and mitigate negative environmental impacts (Y. C. Chen, 2018). This concept is aligned with the concept of circular economy, which is focused on minimizing environmental impact by reusing and recycling, reducing waste from products and services (Aibar-Guzmán et al., 2023; Khajuria et al., 2022).

The circular economy positively impacts these processes because when the waste is destined for landfills, it often incurs disposal costs, giving it zero or even negative value; however, the organic portion of this waste holds energy content that has the potential to be converted into fuel, heat, and electricity, which are valuable products (Y. Yao et al., 2023). The success of this transformation process is based on several factors, such as the characteristics of waste in terms of heating value, moisture content, contaminant concentration, and biogenic content (Y. Yao et al., 2023). Besides that, the availability of waste, government incentives, and the type of technology used for conversion are crucial for the success of this process. (Y. Yao et al., 2023).

A reliable and feasible alternative to landfilling is the thermochemical conversion of solid waste by combustion and gasification (Quan et al., 2022). The combustion technology is well established and allows energy recovery, significantly reducing the volume of solid residues that must be disposed of in landfills, while gasification is more complex and allows the reduction of volume disposed as well and can co-gasify different kinds of waste (Arena et al., 2015). Rezaei et al. (2020) has studied the pelletization of combustible fractions of municipal solid waste as a strategy for energy recovery.

One example of industrial solid waste disposal in landfills is the residual papers from Paper-based laminates (HPL) companies. Paper-based laminates are multipurpose, high-strength, value-added surface materials widely used in the construction and furniture sectors (Němec et al., 2023). A typical HPL comprises layers of kraft paper impregnated with phenolic resin, decorative paper to give a design appearance to the laminate, and a top overlay paper composed of melamine-formaldehyde resin (Martins et al., 2015). Although paper can be recycled, laminates composed of synthetic resins pose significant recycling challenges. The different materials used to make these laminates are hard to separate, making them not easily recyclable. So, thermochemical conversion would be an option to reuse this material.

Despite paper being made of cellulosic compounds, they are different from each other by chemical composition, primarily because of their production process and their final use (Murphy et al., 2021). During paper production, additives, such as fillers, resins, dyes, and dry-strength agents, are added to the pulp to modify its properties, affecting the amount of inorganic content on the paper that depends on the paper's final application (Goyal, 2022).

In combustion equipment, this inorganic content can affect the lifetime and operation, increase operating costs, increase the production of nitrogen oxides and carbon monoxide, reduce heat transfers and cause corrosion (Radačovská et al., 2017). Other disadvantages of directly combusting paper residues are their irregular shape, their low energy density and transportation issues (Machinery, 2014). For using residual paper to generate heat, converting them into densified materials, such as pellets and briquettes, is a good option because it creates a regular shape, increases its energy density, and makes it easier to transport (Xiu et al., 2018). Moreover, the presence of phenolic resin in the papers that come from the HPL process can be advantageous for energy recovery as it gives a higher calorific value to the paper (N. Zhang et al., 2020).

Considering these different types of waste paper that are disposed in landfills and the high potential of energy content that these residues may present, this study aims to investigate the efficiency of energy recovery from paper waste through controlled combustion at different temperatures and air supply conditions to identify optimal conditions to maximize energy recovery from industrial paper residues.

3.4. Material and methods

Feedstock

The residual paper used in this study comes from a company that produces High-Pressure Laminate (HPL). The production of these HPLs leads to the generation of different solid residues of paper, such as paper impregnated with phenol formaldehyde resin, melamine paper, non-phenolic papers and a mixture of all residues from the process together, forming a mixture of phenolic paper and non-phenolic paper, as well as other wastes, such as dust coming from the cleaning process of the production area.

We divided all these residues into three groups: phenolic paper - composed of the residues impregnated with any resin, so melamine paper was included in this group; non-phenolic papers- which are all the residues that do not have resin in it; and, the phenolic mixture - that is the mixture of residues that comes from the production process of the HPL's, that include papers from the other groups and dust. As the objective of this work was to focus on the use of 100% of residues, we aimed in produce pellets mixturing all the residues.

Pellets production

All paper samples were shredded using a micro-cut shredder to achieve standardized particle dimensions of 2.0 x 11.9 mm as a pretreatment. This project defined the pellet's properties according to CANplus certification requirements. In Canada, CANplus is a certification that provides the framework for a system certification of Wood Pellet producers and traders that guarantees the wood pellet quality (Canadian Biomass, 2013). It is not a mandatory seal to sell pellets.

For each group of residues, pellets were produced in batches in a pellet mill PM605 model laboratory scale (Buskirk Engineering) with a production capacity of up to 150 lb per hour (approximately 68 kg per hour) equipped with a variable-speed electric drive. A heating tape

that was controlled by a BSAT time percentage dial was wrapped with thermal insulation around the die to control the temperature during pellet production. To control the temperature, a pocket thermal camera Fluke PTi120, was used, and the die temperature was adjusted at 70°C, with an operational range of 70°C to 100°C due to the frictional heat generated by the roller-die interaction. The die featured a hole diameter of 6.35 mm and a length of 25.4 mm. After production, the pellets were cooled down at ambient temperature and sieved through a 3.15 mm diameter sieve.

The proportion of the mixture pellets were defined as 70% of phenolic dust, 15% of phenolic paper, and 15% of non-phenolic paper.

Pellets characterization

The Pellets were characterized at the Biomass Technology Laboratory (BTL) to evaluate their Physical, Thermal, and Chemical properties.

Moisture and ash content

The moisture content was determined according to the ISO 18134-1:2015 standard (International Organization for Standardization, 2015d) by drying the samples in an oven at 105°C ± 3°C to a constant weight. The ash content was taken by incinerating the samples in a muffle furnace at 575°C ± 25°C, following the ISO 18122:2015 standard (International Organization for Standardization, 2015a).

Pellets dimension

To measure the dimensions of the pellets, a digital micrometer with a precision of 0.001 mm was used, and the measurement was conducted according to the ISO 17829:2015 standard (International Organization for Standardization, 2015b).

Calorific value

The pellets' calorific value was determined by burning the samples in a Parr Instruments model 6400 bomb calorimeter (Moline, IL, USA) following ISO 18125:2017 standard (International Organization for Standardization, 2017).

Elemental Analysis

Elemental analysis was conducted in a Flash 2000 Organic Elemental Analyzer (Thermo Fisher Scientific) to determine the C (carbon), H (hydrogen), O (oxygen), N (nitrogen), and S (sulfur) content of the samples at BTL. This analysis was fundamental to calculating the air used in combustion.

Thermogravimetric Analysis

The papers' thermogravimetric analysis (TGA) was conducted using a TGA instrument (TA Instruments Q 500, New Castle, DE, USA). The TGA was carried out in an air atmosphere, and the operating temperature ranged from room temperature to 1000°C at a ramp rate of 20°C/min in the BTL. The TGA was used to determine the combustion temperature, which was chosen at the highest temperature in which a mass loss was observed.

Mechanical Durability

The durability test was determined by weighing two samples of 100 grams each and tumbling for 2 minutes at 50 rpm. The samples were then sieved using a 3.15 mm diameter sieve, as described in ISO 17831-1:2015 (International Organization for Standardization, 2015c). The durability was determined according to Equation 3.1.

$$Durability\ (\%) = \frac{m_2}{m_1}\ x\ 100 \qquad \text{(Eq. 3.1)}$$

Where m_1 represents the weight of the sieved pellet before the tumbling procedure, in grams (g), and m_2 is the mass of the sieved pellet after the tumbling process, in grams (g).

Durability is measured to evaluate the pellet's tendency to rupture. Low durability can result in a higher rupture rate, leading to the production of fine particles.

Fines content

Fines content in pellet production is defined as any particle with dimensions smaller than 3.15 mm attached to the pellets. A high concentration of fine particles can indicate storage and handling problems and pose health and environmental risks. The fine content of the pellets was determined according to ISO 18846:2016 (International Organization for Standardization, 2016).

Inductively coupled plasma mass spectrometry (ICP-MS)

Inductively coupled plasma mass spectrometry (ICP-MS) analysis was performed with Agilent 7800 ICP-MS (Agilent Technologies) to quantify the metals present in samples with high ash content. The metals analyzed were alkali, alkaline Earth, and transition metals.

Combustion tests

The combustion system used in the experiments was a fixed-bed reactor, which included a compressed air supply system, a stainless-steel combustion reactor with a diameter of 2 inches and a length of 48 inches, and a gas cooling system connected to an exhaust and gas sampling

system. Manual rotameters controlled the airflow at the inlet. Before starting the reaction, a digital flow meter was used at the end of the system to ensure the correct mass flow of air during the reactions. This digital flow is not attached to the system. Both air and pellets were fed from the top of the reactor. The CO_2 was used just for safety purposes. The temperature inside the reactor was measured by thermocouples placed in three different zones to control the temperature in the reactor. When the combustion system reached the desired temperature, 30 g of pellets were introduced from the top to zone 3, and the reaction started to be counted. The reaction occurred in the zone 3. The air was flowing continually during the reaction. Figure 3.1 shows the P&ID of the fixed-bed reactor.

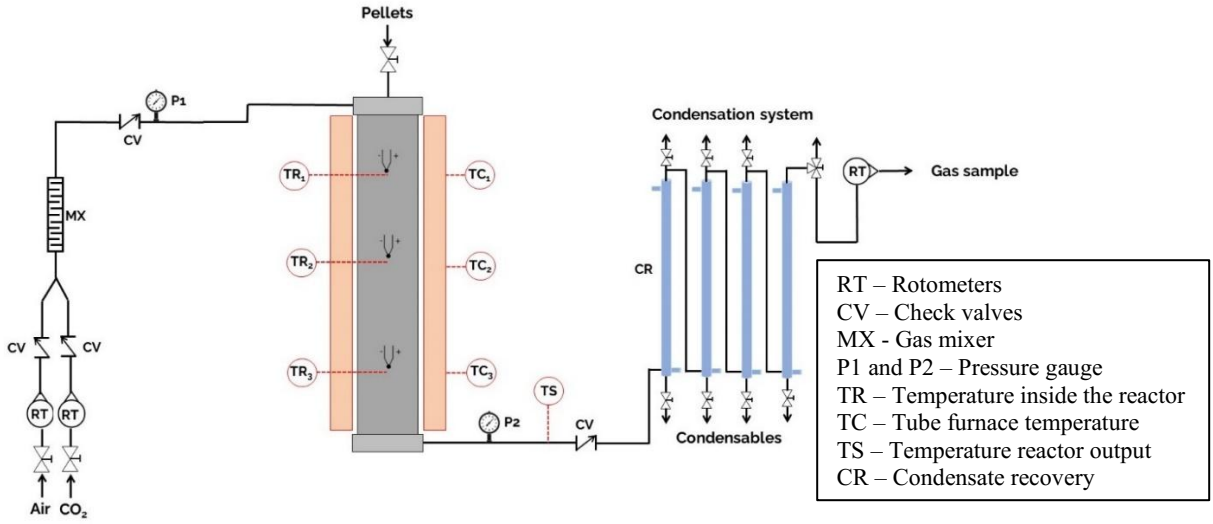

Figure 3.1 - P&ID of the semi-pilot fixed-bed set up for biomass combustion.

The gases generated during combustion passed through a cooling system to separate the condensable and non-condensable gases. This system consisted of a glycerol solution recirculated at a temperature of -10°C in a condenser system. The gases were collected at the cooling system outlet, through a valve, and were subsequently identified using gas chromatography. Gas chromatography (GC) used was the model Scion 456 GC (Bruker). The GC had two detectors of thermal conductivity (TCD) with two columns to separate the gases: Molsieve 13X, 80/100 mesh, 1.5 m x 1/8" IS and Hayesep N, 80/100 mesh, 0.5 m x 1/8" IS, and one thermal conductivity detector of flame ionization (FID) used for more prolonged chain hydrocarbon detection and separation with a BR-1, 10 m x 0.15mm, 2 µm column. In the

analyses, the measured gases were O_2, CO_2, and CO. The pressure in the system was monitored using a manometer.

Equation 3.2 was applied to determine the air supply for the combustion of the pellets based on the elemental analysis as described by (Silva et al., 2018).

$$C_{x_1}H_{x_2}O_{x_3}N_{x_4}S_{x_5} + \eta.(3.76N_2 + O_2) \rightarrow aCO_2 + bH_2O + cO_2 + dN_2 + +eCO + fCH_4 + fNO_x + gSO_x \quad \text{(Eq. 3.2)}$$

The excess of air was calculated according to equation 3.3.

$$\alpha = \frac{V_a}{V_a^o} \quad \text{(Eq. 3.3)}$$

Where V_a is the real volume of air supply for the combustion; V_a^o is the stochiometric air supply volume, and α is the air coefficient.

The emission and temperature registration tests were performed on alpha 1.0. The values used for alpha were 1.0, 1.5, 2.0, and 3.0 to study the variation of alpha applied to the mixture pellets.

ESP Arduino monitored the temperature during the combustion with a measurement accuracy of 99.6%.

3.5. Results and discussion

Pellet production and characterization

Table 3.1 presents the composition of three groups of paper residues obtained by elemental analysis, moisture content, ash content, and high heating value (HHV).

Table 3.1 Results of elemental analysis, moisture content, ash content, and calorific value of residues.

Pellets	Elemental analysis (%)					Moisture Content	Ash Content	HHV
	C	H	O	N	S	(%)	(%)	(MJ/kg)
Phenolic mixture	46.02	5.79	41.48	1.10	0.02	5.66	6.38	19.18
Phenolic paper	43.64	5.64	43.88	2.52	0.00	1.89	2.23	20.07
Non-phenolic paper	46.51	5.88	40.00	1.42	0.00	2.53	3.61	15.52
Phenolic dust	45.45	5.80	41.95	0.44	0.00	7.92	10.08	17.70

The carbon content is relatively close in the three groups of residues. The phenolic mixture was 46.02%, the phenolic paper was 43.64%, and the non-phenolic paper was 46.51%. Hydrogen varied slightly, with 5.79% for phenolic mixture, 5.64% for phenolic paper, and 5.88% obtained in non-phenolic paper. Oxygen content varied, with the highest content in phenolic paper at 43.88%, 41.48% for the phenolic mixture, and non-phenolic paper at 40.00%. The highest nitrogen content, 2.52%, was found in the phenolic paper, then 1.10% in the phenolic mixture, and 1.42% in the non-phenolic paper. The presence of the N in the samples is because dyes are added to the resin to provide different colors to the papers, and the N is probably part of the composition of these dyes. Another source of N is melamine paper, which has N in its chemical structure. Sulphur was detected in 0.02% of the phenolic mixture.

Moisture content varies more between samples. The phenolic dust had the highest moisture content, 7.92%, while the phenolic paper had the lowest, 2.23%. The non-phenolic paper presents 2.53%. The mixture pellets presented a moisture content of 5.66%. This can be explained since most compositions of the pellets are made of phenolic dust. As dust has a small

particle size and paper is considered a hygroscopic material, it would be easier for this material to absorb water or humidity, contributing to a higher moisture content in the sample. These values are similar to those found in the literature in studies using different types of paper waste. Pérez et al. (2019) found moisture content values of 5.4 % ± 0.7 for waste paperboard and 5.0 % ±1.4 for waste kraft paper. Brummer et al. (2014) studied several types of waste paper, such as offset paper, recycled paper, recycled paper-printed, cardboard, and filter paper. The moisture content results ranged from 3.07% to 4.64%.

Ash content also shows some variation. Phenolic dust presented a high ash content, with 10.08%, while the phenolic mixture presented an ash content of 6.38%. The phenolic paper showed 2.23% for the ash content, and the non-phenolic paper was 3.61%. Pérez et al. (2019) found an ash content for paperboard samples of 4.0% and 11.8% for kraft paper. Brummer et al. (2014) got 14.02% of ash content for cardboard and 24.06% for recycled paper. Rahman (2020) studied pellets made from paper waste (paper towels and cardboard boxes) and found an ash content of 2.9% for paper towels and 5.3% for cardboard. Inorganic content on papers depends on the paper's final application. According to Goyal (2022), for cardboard, the inorganic content (ash) is usually <5%, while for tissue paper, it is <2%, and in printing and writing paper, the content varies between 0-30%. These non-fiber additives, such as kaolin (Al_2O_3, SiO_2) and calcium carbonate ($CaCO_3$), increase the ash content on these papers. This justifies the high ash content in decorative paper because it is used to design the laminate and fits on the printing and writing paper category. This type of paper receives a high amount of fillers to decrease imperfections on the paper surface for printing on it. The phenolic mixture presented high ash content due to the material being a mixture of all the residues produced in the industry, which makes the material heterogeneous with a mixture of all the properties of the other papers, including the phenolic dust, which is composed of impurities originating from the factory cleaning process, as explained earlier in this document.

An inductively coupled plasma mass spectrometry (ICP-MS) test was performed to evaluate the presence of transition metals in the phenolic mixture due to its high ash content. The results are shown in Table 3.2. The high calcium concentration in the phenolic mixture stands out.

Ash deposition is a problem in combustion systems, as it is associated with fouling, slagging, equipment corrosion, and reduced energy efficiency. The vaporization and condensation of elements composed of alkali metals, is direct related in the deposition problems in combustion systems, mainly potassium (Horák et al., 2019). The high presence of Al and Ca could lead to ash deposits in grate-firing boilers and formatting CaO, and Al_2O_3 (Z. Tang et al., 2016).

Table 3.2 - ICP-MS analyses of the phenolic mixture.

Element	Phenolic mixture (ppm)
Ca	14671
Al	2917
Na	1273
Fe	767
Mg	621
K	166
Mn	29.8
Zn	22.2
Cu	13.1
Cr	3.74
Pb	3.74
Ni	1.96
Co	0.71
Cd	0.062

According to G. Chen et al. (2022) the main methods used to minimize the problems with ash in combustion have been washing with water to solubilize ash incrustation and screening potential additives. Among these methods, the use of additives has been an economically more efficient option in minimizing the negative impact of the boilers.

The calorific value represents the energy content of the residues. The high heating value of the samples varied. The highest HHV is phenolic paper pellets, 20.07 MJ/kg, followed by pellets mixture, 19.18 MJ/kg. The non-phenolic paper pellets have the lowest HHV, 15.52 MJ/kg. Table 3.3 compares the calorific value of phenolic resin, cardboard, paper, wood, natural gas, and the mixture pellets in this study.

Table 3.3 - Comparison of calorific value of phenolic resin, cardboard, paper, and wood. Adapted from (Agarwal et al., 2014b).

Material	HHV (MJ/kg)	Reference
Phenolic resin	31.00	(N. Zhang et al., 2020)
Cardboard	14.48	(Grammelis et al., 2009)
Office paper	13.82	(Grammelis et al., 2009)
Newsprint	15.87	(Grammelis et al., 2009)
Pinewood	21.60	(Viana et al., 2018)
Methane	34.12*	(Canada Energy, 2024)
Mixture pellets	19.18	In this study

* in MJ/m^3

According to Table 3.3, phenolic resin is a material with a high calorific value (31.00 MJ/kg) when compared to the different types of paper and wood. Paper is a material with a low calorific value, making it less attractive for energy recovery; however, the presence of the phenolic resin in the papers in this study makes it more attractive for energy recovery, increasing the calorific value of the paper. Compared to pine wood, a feedstock widely used for combustion to produce heat, the phenolic paper could be competitive in terms of caloric value. Also, it presents energy content is comparable to methane, which is a non-renewable source mostly used as an energy source. This shows that the presence of the phenolic resin on the papers contributes positively to increasing their calorific value.

Considering an annual residues production estimate of around 5 tonnes for an HPL industry and considering the calorific power presented by the mixture pellet of 19.18MJ/kg, compared to the calorific power of natural gas at $34.12MJ/m^3$, this industry could replace the use of natural gas around 60% with the combustion of mixed pellets.

Figure 3.2 shows the blend for the mixture of pellets and pellets ready. Then, the properties of the mixture pellet were compared to the CANplus requirements, as shown in Table 3.4.

Figure 3.2 - The mixture of paper residues before and after pelletization.

The pellets made with the mixture of the residues attended the CANplus specifications, except for the ash content, which was 6.4%. The mixture pellet presented a moisture content of 5.7%. For pellet production, moisture content is a very important property as it increases the fluidity of the sample through the holes in the die of the pelletizing (García et al., 2019). Although it has the moisture content required by the CANplus, when the mixture was added to go through the pelletization process, the blend was moistened to 20% by spraying water to maintain the fluidity of the mixture in the die. According to Chew et al. (2018), low sample humidity can lead to blockage of the die. After the process, pellets presented 21% of moisture content. For stabilization of the pellet structure, they were stored in an open room with a temperature of 21ºC and a humidity of 40%. During this process, pellets lose moisture content to the environment until it is in equilibrium with the environment.

Table 3.4 - Comparison of properties of pellets mixture with CANplus specifications.

Property	**Unit**	**Grade B***	**Pellet mixture**
Diameter	mm	6 ± 1 or 8 ± 1	6.15 ± 0.03
Length	mm	$3.15 \leq L \leq 40$	25.03 ± 1.27
Moisture	% mass	≤ 10	5.7 ± 0.7
Ash	% mass	≤ 2.0	6.4 ± 1.1
Durability	% mass	≥ 96.5	98.7 ± 0.05
Fines	% mass	≤ 1.0	0.99 ± 0.12
HHV	MJ/kg	≥ 18.6	19.2

*CAN*plus* (B): is the grade used for industrial pellets.

Another aspect addressed is moisture content's influence on pellets' durability. After the pelletizing process, the moisture content and durability of the pellets were measured for 15 days until both properties stabilized in the last three measurements. On the first day, when the pellets were produced, they presented a moisture content of 21.2% and durability of 96.2%, which is already a value very close to CANplus requirements. The pellets lost moisture over the next few days until they stabilized at 5.6% on the fifteenth day. It was observed that the durability of the pellets increased as the moisture content decreased. On the fifth day after storage, durability reached 98.3% and moisture content 10.5%. Durability stabilized over the next ten days, reaching 98.7%, while humidity decreased to 5.6%, as shown in Figure 3.3. So, durability responded inversely to moisture content.

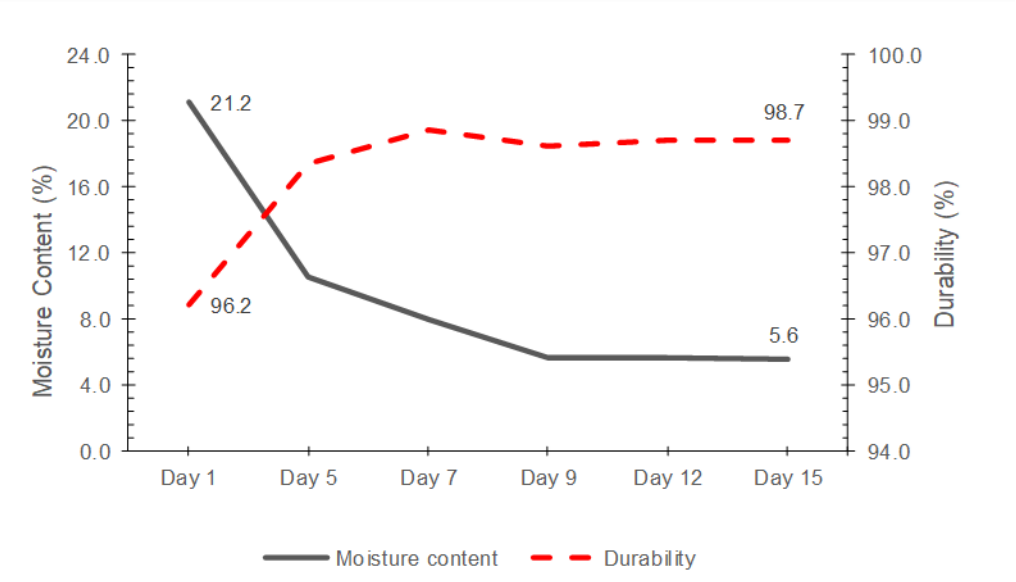

Figure 3.3 - Durability and humidity of the pellets produced as a function of storage time.

According to Rueda et al. (2022) higher moisture content induced low durability in lignin pellets. The higher the moisture content, the lower the durability of the pellets, with durability of 94.8% for pellets with 8% moisture content and 88.9% durability for 38.83% moisture content (Rueda et al., 2022). Lee et al. (2021) exposing commercial softwood pellets to 95% relative humidity, the pellet's durability dropped from 99% to below 80% after 10h of exposition. (Lavergne et al., 2021) found a negative correlation between moisture content and pellet

durability for Scott pine made by forest residues, while for willow and poplar, pellet durability increased with higher moisture content. It was not found to have a significant effect of moisture content on pellet durability for the other biomass residues in their study. García et al. (2019) produced pellets of residual biomass with the best durability (99.4%) for pellets with a moisture content of 16.6%. Senila et al. (2020) studying pellets from vineyard waste found the highest durability (>97.5%) in pellets with a moisture content of 10-12% and the lowest durability (<65%) in a moisture content of 6-8%.

In the study of Rezaei et al. (2020), pellets from municipal solid waste (paper, cardboard, wood, food waste, and soft and hard plastic) were made, and they concluded that the pellets' composition affected their durability. Paper content increased pellets durability (92%) in pellets with 50% of paper and 30% of plastic, against pellets with 40% plastic and 30% paper content (50%). Rahman (2020) produced pellets from paper towels and reported that the pellets' durability increased with moisture content. At 45% moisture content, the durability value reached almost 92%, compared with 65% durability at 15% moisture content.

Figure 3.4 shows the TGA curve for (a) Non-phenolic paper pellets, (b) Phenolic paper pellets, and (C) Mixture pellets in an air atmosphere. There are two curves: the percentage of weight loss (TG), which measures the change in mass of the sample as a function of temperature, and the first derivative of weight change (DTG), which represents the weight loss events. The peak in the DTG curve corresponds to a significant weight loss that can be attributed to the oxidation of the material under air conditions. This peak was used to define the initial temperature for the combustion of the pellets.

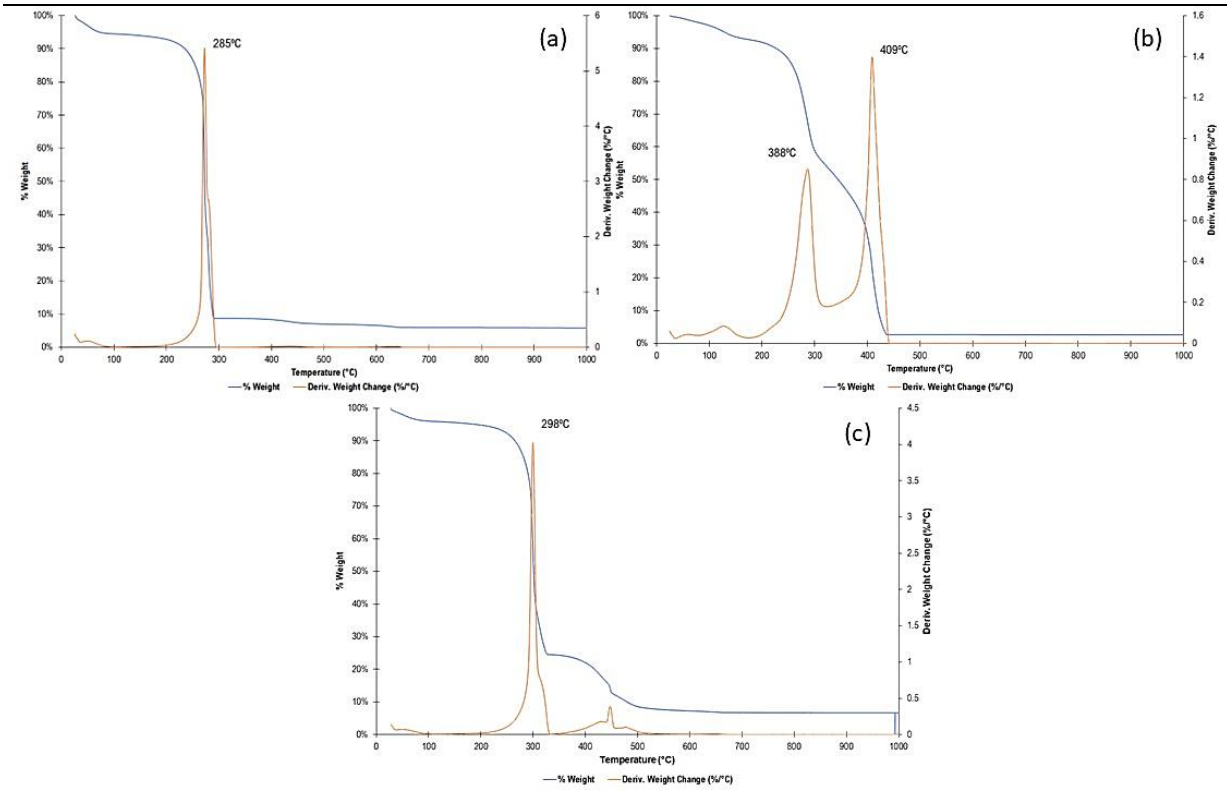

Figure 3.4 - TGA of pellets under an air atmosphere at 20°C/min. (a) Non-phenolic paper pellets; (b) Phenolic paper pellets; (C) Mixture pellets.

The TGA curve for a non-phenolic paper pellet in an air atmosphere is shown in Figure 3.4 (a). The weight loss curve shows a significant decrease in weight at about 285°C, and this temperature is pointed out with a peak in the derivative weight change curve, indicating a significant decomposition rate at this temperature for the non-phenolic paper pellets. As paper is made mostly of cellulose, we assume that this peak corresponds to the thermal decomposition of the cellulose that form the paper. These results are lower than the results studied by Agarwal et al. (2014b) in the thermal oxidation of cardboard, which presented a peak of mass loss at 335.4°C at 20°C/min and Soares et al. (1995) studied the thermostability of a kraft paper sample in oxidizing and inert atmospheres and reported that air conditions seem to accelerate the thermal degradation of kraft paper, with the principal mass loss between 317 and 329°C and maximum loss at 324°C.

Figure 3.4 (b) illustrates the TGA analysis of the phenolic pellets. Two major weight loss events are seen for the phenolic paper pellets. First, it starts with the rupture of methylene bridges.

Then, the first significant weight loss is observed at about 388°C and matches well with a peak of the derivative curve. This peak represents primarily the degradation of low molecular weight compounds such as the phenol compounds. The second weight loss of note occurs at 409°C. These temperatures thus represent the significant steps in decomposition for the phenolic paper pellets and imply a significantly more complex decomposition process than non-phenolic paper pellets. It can represent the decomposition of mostly complex molecules like compounds from the BTEX group (Benzene, Toluene, Ethylbenzene, and Xylene) (Bobrowski et al., 2018). Bouajila et al. (2003) in their studies of the degradation of phenolic resins in an oxidizing atmosphere, they found that the degradation process started at 380°C and had a first most significant peak of mass loss at 451.4°C and a second one around 475°C with total mass loss at 600°C. In the study of Bobrowski et al. (2018) the thermal decomposition of phenol resin at 10°C/min also revealed two stages of decomposition: the first peak at 412°C with 35% of mass loss and the second one at 440°C with 45% of mass loss. The difference in the early peak in this study might be explained by the fact that resin has papers in its constitution.

Figure 3.4 (c) shows the TGA curve of the mixture pellets. There is a significant weight loss at around 298°C, located by a peak in the derivative weight change curve that indicates the main decomposition temperature of the mixture pellets.

As the pellets composition of the mixture is a blend of the different groups, this might contribute to (1) facilitating the thermal degradation of phenolic resin present in the pellets due to the dilution of phenolic content, (2) pellet mixture has a composition less complex than phenolic-paper pellets, resulting in an earlier peak of mass loss (3) shifting the peak of significant mass loss to the right, when compared to the non-phenolic paper, because the phenolic content might provide a protection effect, retarding the decomposition of the non-phenolic paper.

Combustion

Influence of the temperature

Temperature is important in the combustion evaluation process because it is related to the thermos degradation of the material and, consequently, the total energy release of the material

(Xu et al., 2020). The initial combustion temperature was defined by the most significant peak of mass loss on thermogravimetric analysis. Then, the combustion temperature was studied in the fixed-bed reactor. The group of mixture pellets was chosen for this reaction because we aim for the energy recovery of all residues.

The objective was to verify if the initial combustion temperature of 300°C, defined by the thermogravimetric analysis, would be sufficient to release all the heat from the pellets. The delta temperature (ΔT) of the combustion was used to verify the influence of the initial temperature on heat production. The pellets were subjected to combustion temperatures of 300°C, 350°C, and 400°C in α = 1.0.

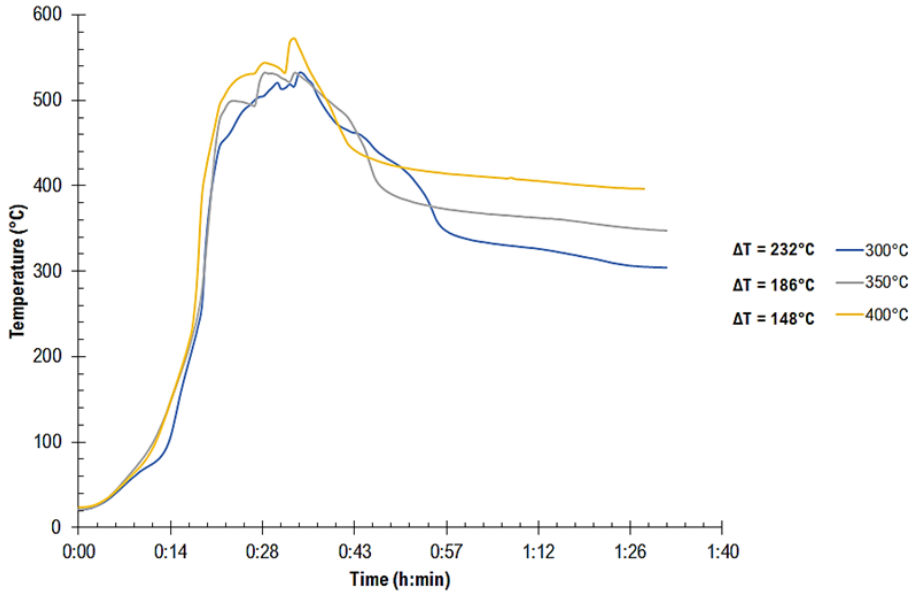

Figure 3.5 - Influence of temperature on the combustion of mixture pellets at 300°C, 350°C, 400°C, α= 1.0.

The results obtained for the ΔT while the pellets combust at different initial temperatures of 300°C, 350°C, and 400°C are shown in Figure 3.5. At the temperature of 300°C, the pellets generated heat of 532°C, resulting in a ΔT of 232°C. Further, increasing the initial temperature to 350°C decreased the ΔT to 186°C. Then, at an initial temperature of 400°C, the ΔT decreased to 148°C. These results indicate that raising the combustion temperature did not increase heat production or a higher ΔT for the pellets. It is accordingly concluded that 300°C is the optimum

temperature for the combustion of these pellets, thus justifying the methodology used in this research.

A mixture of the different groups of paper residue may have lowered the ignition temperature of the pellets. This increase in temperature provides sufficient energy for the degradation of the resin present in the pellets. This phenomenon could explain why increasing the initial combustion temperature did not positively correlate with heat production. It is important because these thermal properties are crucial for optimizing the combustion of the pellets and maximizing energy output.

Previous reports have indicated that the efficient combustion of wood biomass and agricultural solid waste ranged between 224°C and 260°C (Yan et al., 2019). Bouajila et al. (2003) and Agarwal et al. (2014b) report that the degradation of resins in an oxidizing atmosphere begins at approximately 380°C, and total mass loss is achieved at 600°C, and kraft paper presented a peak of mass loss at 335°C, respectively, as mentioned earlier in this paper.

Influence of the composition

The chemical compositions of papers affect these materials' thermal decomposition and oxidation (Agarwal et al., 2014a). Figure 3.6 shows the thermal decomposition and heat production by the three groups of paper residues during the combustion process, carried out with an air coefficient of $\alpha = 1.0$ at a combustion temperature of 300°C.

From the combustion graph of the three types of pellets (Figure 3.6), we can obtain information for the utilization of the material according to the purpose of energy recovery.

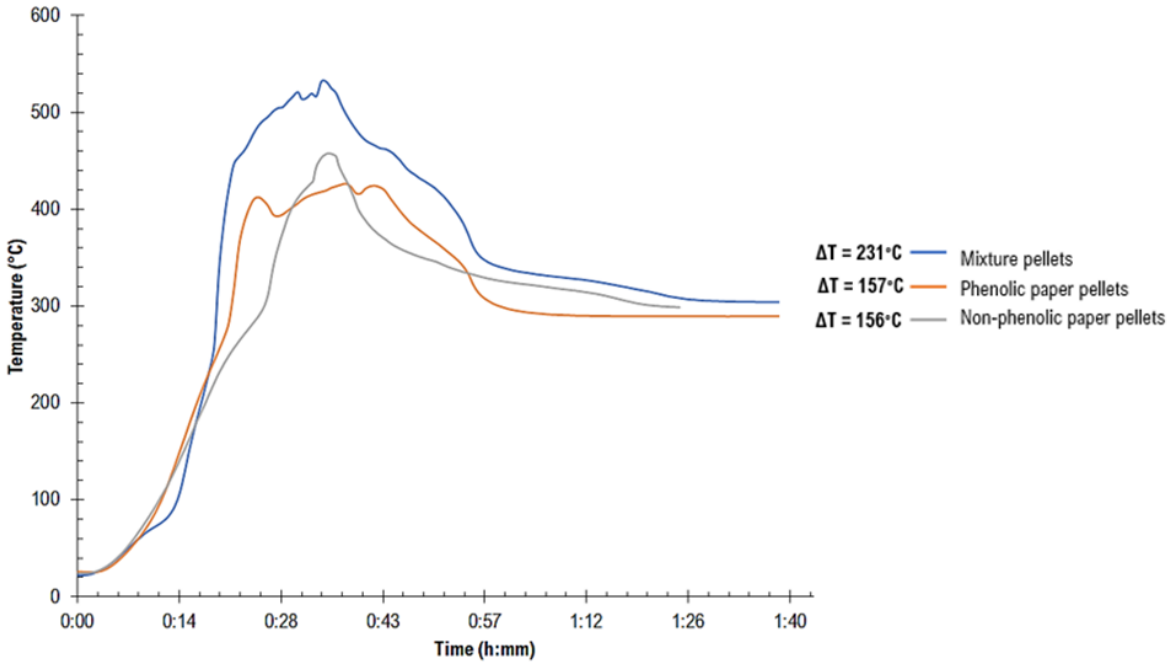

Figure 3.6 - Influence of composition of the pellets on the combustion at 300°C, α= 1.

Non-phenolic paper pellets burned faster when compared to the phenolic paper pellets, reaching a peak temperature of 456°C and exhibiting a temperature increase (ΔT) of 156°C during combustion. Xiu et al. (2018) reported that wastepaper briquettes burned faster than wood and coal briquettes and burned in the same burn time as kindling firewood. If the goal is to produce temperature with this pellet, and the demand is 400ºC, the paper pellets can meet this requirement, and the need to refuel would be around 40 minutes after the start of combustion.

The phenolic paper pellets presented a ΔT similar to kraft paper pellets, suggesting that phenol, acting as a flame retardant, requires higher temperatures for thermal degradation. Therefore, it can be inferred that phenolic papers alone need temperatures exceeding 300°C for effective combustion, as demonstrated by the TGA. This insight is significant because the proportion of phenolic paper in the pellet composition can influence the required initial combustion temperature. Consequently, this factor should be considered in waste management strategies aimed at energy recovery. The quantity of each residue type, whether phenolic or non-phenolic, may vary depending on industrial production requirements. Thus, a higher initial combustion temperature may be necessary if a larger proportion of phenolic materials is present.

Upon analyzing the mixture pellets' result containing phenolic and non-phenolic materials, we observed a higher ΔT of 231°C. Since the thermal degradation of non-phenolic samples begins at lower temperatures than that of phenolic ones, the energy required for the combustion of the mixture pellets is lower than that of the phenolic paper pellets. This highlights the significant advantage of blending phenolic and non-phenolic papers for combustion.

Galiwango et A.Gabbar (2022) investigated the activation energy required to ignite municipal paper and polypropylene residues during co-pyrolysis. Their modelling study revealed that polypropylene residues required higher activation energy compared to paper. Specifically, the activation energy for polypropylene was 187.25 kJ/mol according to the Coats-Redfern model, while paper required 159.25 kJ/mol. This difference was attributed to the presence of aliphatic compounds and aromatic rings in polypropylene, which increase the energy needed to break the inherent chemical bonds. Additionally, the study found that when paper and polypropylene residues were mixed, the activation energy required decreased to 143.74 kJ/mol compared to the activation energy of homogeneous residues. This suggests a beneficial energy profile for mixed polymer-lignocellulosic residues, which is common in municipal solid waste.

The presence of phenol can be beneficial as it enhances the calorific value of the samples. Phenol contributes to a higher energy content, which contrasts with paper, which typically has a lower calorific value. The strategy of mixing the residues to produce the pellets for combustion may reduce the ignition temperature of the pellets, reducing this retarding effect that phenol attributes to its samples. Moreover, the mixed pellets would be the only ones capable of meeting a temperature production demand higher than 400ºC, reaching temperatures over 500ºC.

The emission of carbon monoxide (CO) and carbon dioxide (CO_2), along with the consumption of oxygen (O_2), were measured during combustion. Figure 3.7 presents the evaluation of the formation of these gases over time for the pellet's mixture at a temperature of 300°C and $\alpha = 1$.

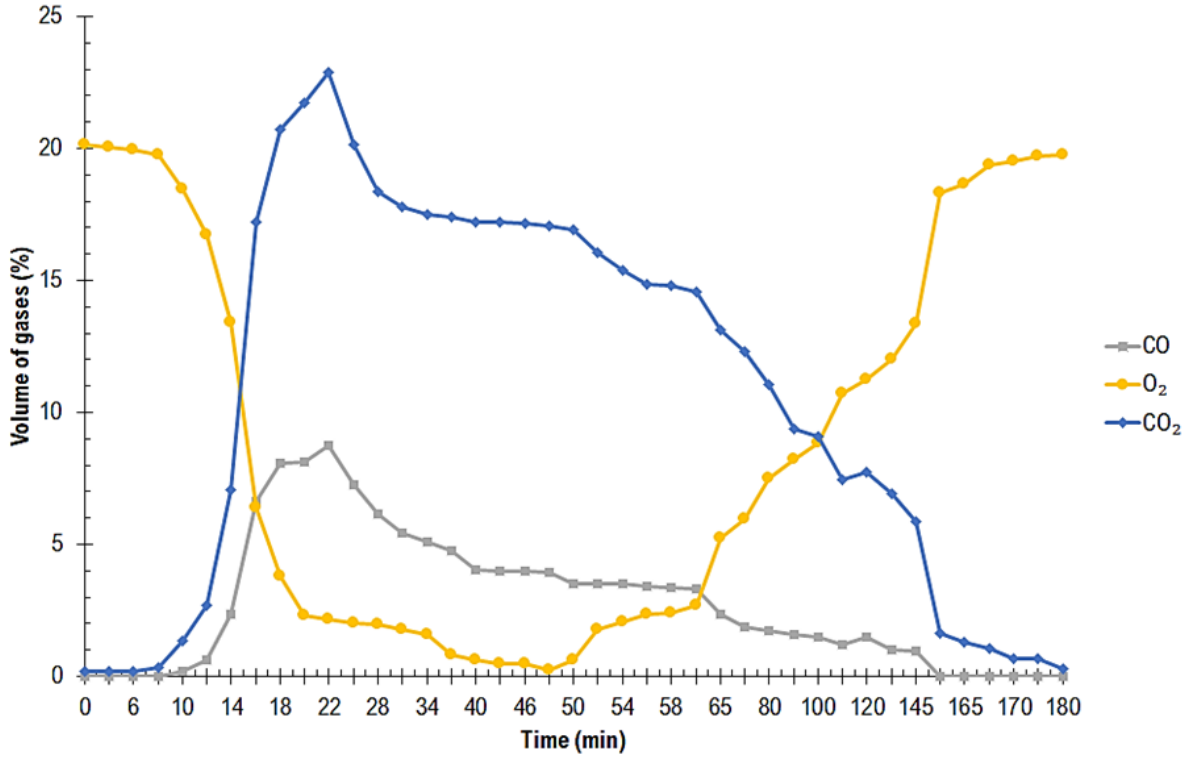

Figure 3.7 - Evaluation of CO and CO_2 formation and oxygen consumption as a function of time for mixture paper pellets at T=300°C, α=1.

At the beginning of the combustion, the oxygen concentration was around 20%. Between 10 and 20 minutes, the O_2 concentration declines, dropping to near zero as combustion intensifies and remains in a low concentration when it reaches a low point of 0.24% in 48 minutes. After that point, it starts slightly to rise again as the pellets finish burning. The carbon dioxide (CO_2) starts in a low concentration, and between 10 and 20 minutes, there is a sharp increase in the concentration, peaking at 23% at 22 minutes as the combustion products are released. Then, it gradually decreases over time, declining to around 0.3% at the end of the combustion. Carbon monoxide (CO) starts very low, and then the concentration starts to rise, peaking around 18 and 22 minutes with a concentration of 8.7%, and then, gradually decreasing until the end of combustion.

The emission peak of CO_2 and CO occurred between 20 and 30 minutes. The CO_2 indicates the maximum rate of fuel oxidation. The CO indicates incomplete combustion processes alongside

complete combustion. The formation of CO may have occurred due to low oxygen mixing with the fuel leading to a partial combustion of the carbon.

$$2C + O_2 \rightarrow 2CO$$

It could also have happened due to parallel reactions of competition that can occur during the combustion of solid fuels, involving carbon dioxide, as described in the Boudouard reaction.

$$C + CO_2 \rightleftharpoons 2CO$$

The presence of CO in the combustion of papers has been reported in the literature by Xiu et al. (2018) and Yaqub et al. (2021). According to the authors, briquettes maintained their shape until the end of combustion, which could restrict air penetration and thus result in more significant CO emissions.

Also, the phenolic resin's thermal decomposition can lead to CO formation. One of the main reactions is the oxidation of the phenol present in the resin structure. As the temperature increases, the chemical bonds in the resin begin to break down, producing volatile gases, including CO. Same results were found by Bouajila et al. (2003) and Bobrowski et al. (2018). They found that carbon monoxide emissions occurred between 200 and 600°C, and the higher the temperature, the higher the CO formation. In high temperatures, the formation of CO could be even higher than that of CO_2.

Figure 3.8 evaluates the formation of these gases over time for the non-phenolic paper pellets at 300°C and $\alpha = 1$.

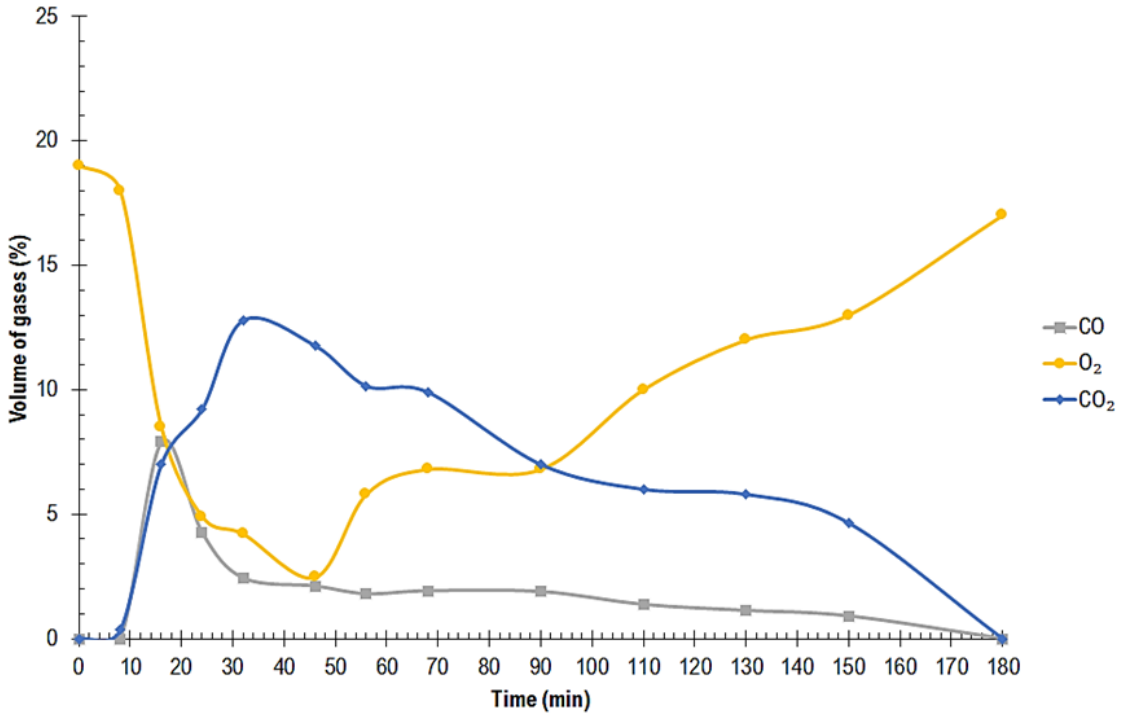

Figure 3.8 Evaluation of CO and CO_2 formation and oxygen consumption as a function of time for non-phenolic paper pellets at T=300ºC, α=1.

During the combustion of non-phenolic paper pellets, the oxygen (O_2) consumption in the gas samples collected was significantly reduced to a volume of 2.4%. This indicates that the pellets reacted with oxygen in the combustion reaction, leading to the oxidation of the material and the generation of heat. After 8 minutes of reaction, the oxygen level dropped drastically, as shown in Figure 3.9. Concurrently, carbon dioxide (CO_2) and carbon monoxide (CO) gases started to be produced in more significant volumes. The peak production of CO_2 was 12.8% in 32 minutes, while CO reached a peak of 7.9% around 16 minutes. Agarwal et al. (2014b) mention that the gases involved in the decomposition of paper consist of H_2O, CO_2 and CO.

Similar results were observed by Yaqub et al. (2021) in the chemical looping combustion of paper waste at 800ºC. The paper waste reacted immediately, producing high peaks of CO and CO_2. The authors attributed the high concentration of CO to the insufficient contact between the gasification products and the oxide carrier.

Xiu et al. (2018) reported similar findings with high CO emissions for paper briquettes. They attributed this result to the mechanical properties of the briquettes preventing the optimal mixing of oxygen with the fuel, thereby increasing the formation of CO, as discussed before.

The emission of CO during biomass combustion has also been reported. Ozgen et al. (2014) registered that firewood oak emitted 21,720 mg of CO per MJ in a traditional combustion stove. It is a value significantly higher than the 1.7 mg/MJ reported by Wiinikka & Gebart (2004) for wood pellets in a laboratory pellet burner. Additionally, emission of 9.6 mg/MJ was measured from a mixture of wood and reed canary grass in a 40-kW boiler by (Kortelainen et al., 2015).

The gas emission of the combustion phenolic paper pellets is shown in Figure 10. As shown in the Figure 3.5 (b), the thermal degradation of phenolic paper pellet presented two peaks of degradation, 388°C and 409 °C. We considered this peak of temperature close to each other. Hence, the temperature of 390°C was chosen as the initial combustion temperature.

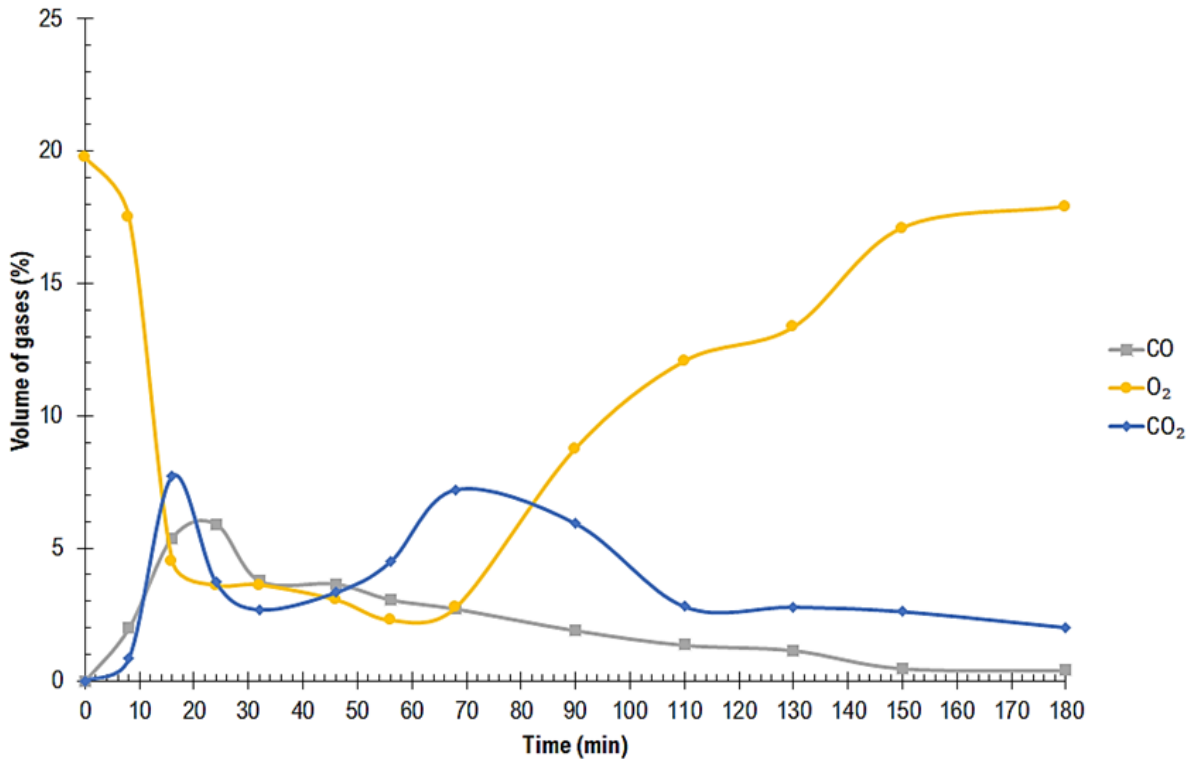

Figure 3.9 Evaluation of CO and CO_2 formation and oxygen consumption as a function of time for phenolic paper pellets at T=390°C, α=1.

Figure 3.10 shows the oxygen (O_2) consumption and production of CO_2 and CO during the combustion of phenolic paper pellets. The oxygen level reduced to 2% around 55 minutes after the start of the reaction. For CO_2, it was observed two peaks of production. Its behaviour was similar to the TGA, which also presented 2 peaks of significant mass loss. Initially, CO_2 production increased to 8%, then, between 20 and 60 minutes, it dropped to 3% before rising back to 8% again. During this range, CO production was slightly higher than CO_2. This behavior can be explained by the thermal decomposition of the phenolic resin. Several reactions can lead to CO formation, such as the oxidation of phenol present in the resin structure and the oxidation of formaldehyde. Additionally, the thermal decomposition of phenolic resin at high temperatures can release CO, as mentioned. Similar findings were reported by (Bouajila et al., 2003) and (Bobrowski et al., 2018) aligning with the findings in this research.

Influence of the air coefficient

Any conventional fuel requires a stoichiometric amount of oxygen according to its chemical composition to theoretically achieve complete combustion (Marangwanda et al., 2020). When this air coefficient is equal to one (α =1), it means that the exact stoichiometric air supply for the complete combustion is used (Abdoli et al., 2018). In this work, the air coefficient was varied between 1.0 and 3.0 to study the influence of the air coefficient on the combustion and heat production of the pellets. The mixture pellets were chosen for this test.

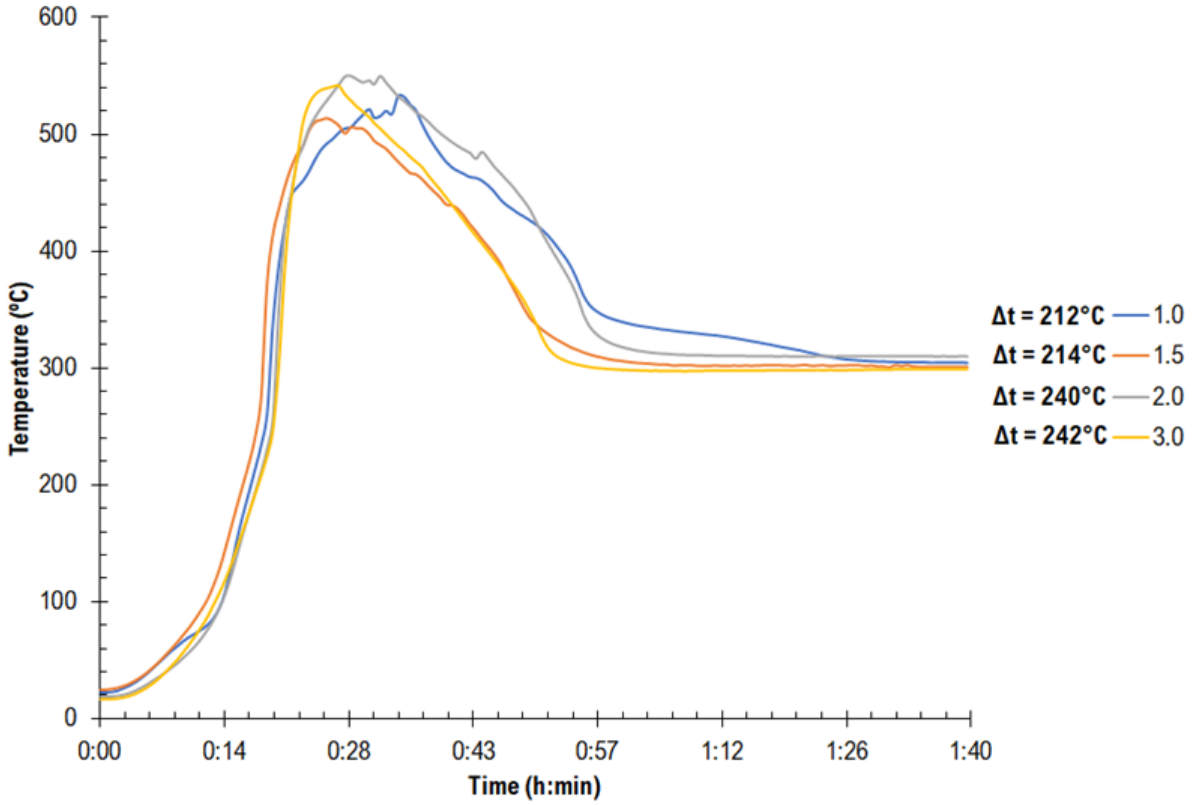

Figure 3.10 - Influence of the air on the combustion of mixture pellets at α = 1.0, α = 1.5, α = 2.0, α = 3.0 at 300°C.

Figure 3.11 shows the air coefficient's influence on the pellet mixture's combustion at 300ºC. The delta temperature (ΔT) for each air coefficient (α) were 212ºC, 214ºC, 240ºC and 242ºC, respectively.

The volume of air supplied affected the heating rate and thermal behavior during combustion. The curves for α=1.0 and α=1.5 showed a more gradual combustion and reached a slightly lower peak temperature compared to α=2.0 and α=3.0. In contrast, the curves for α=2.0 and α=3.0 reached an earlier, higher, and more pronounced temperature peak. It suggests that a greater air coefficient accelerated combustion, resulting in higher temperature peaks and, consequently, a higher ΔT.

Figure 3.12 shows the graphs with the volume percentages of CO, CO_2 and O_2 during the combustion versus time for the α 1.0, 1.5, 2.0 and 3.0. Gas sample was made every 20 minutes of reaction.

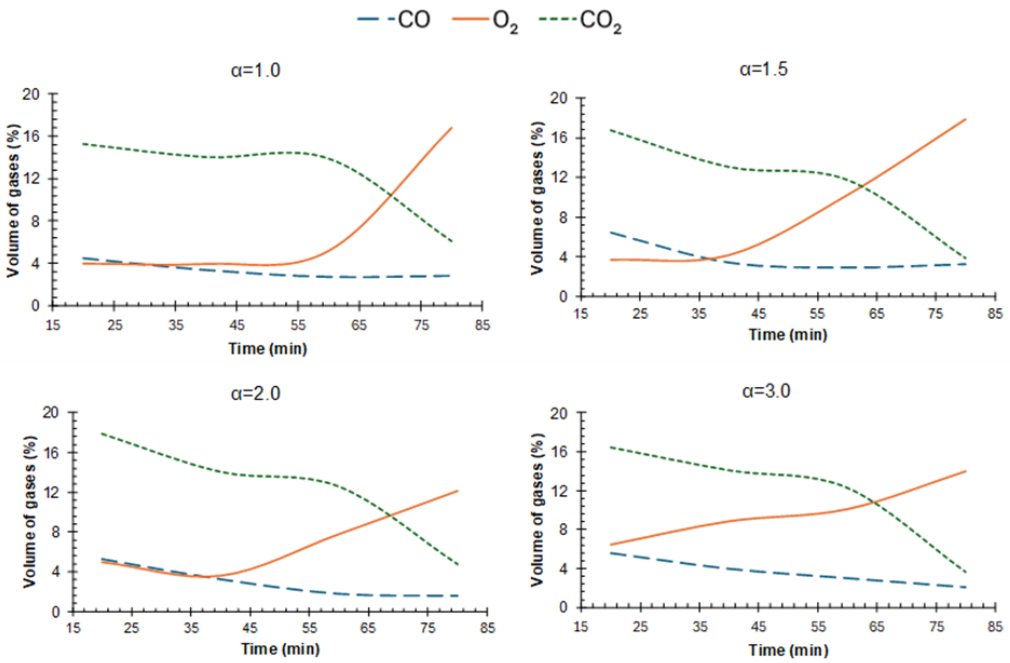

Figure 3.11 - Influence of the air on gas emission on the combustion of mixture pellets at α = 1.0, α = 1.5, α = 2.0, α = 3.0 at 300°C.

There was some similar behaviour in the graphics. At α = 1.0, CO after 20 minutes of reaction was around 5% and continued a bit flattened until the reaction's end. CO_2 starts high and gradually decreases, showing that the carbon material was consumed, and that combustion was going on. O_2 levels in the first measurement were 4% and continued flat till 55 minutes of reaction, then returned to a higher level. Inversely, during this time, CO_2 was high and started dropping when oxygen started to increase, reflecting the consumption of oxygen due to combustion.

At α = 1.5, a similar trend in CO was observed. The concentration was higher than that in α = 1.0 at the beginning and then decreased along the reaction until the end. The CO_2 level peaks at

the beginning and decreases faster than in α = 1.0. Oxygen concentration: the more CO2 was produced, the more O2 was consumed.

At α = 2.0, the levels of CO were a bit higher at the beginning and fell with time; this decline becomes very pronounced towards the end of the combustion period, indicating a more complete combustion process. CO_2 levels went up high and then fell with time, while the O_2 raised oxygen consumption and more complete combustion compared with α=1.5.

At α = 3.0, the levels of CO were at their highest amongst the samples at the beginning and decreased with time. CO_2 level follows a similar trend but started high and then decreased along the reaction. The O_2 levels were in higher concentration, probably due to the excess of oxygen present in the air flux. In general, with increasing α values, more efficient burning happened. In the beginning, CO and CO_2 levels were high, with more oxygen consumption at the same time.

According to Caposciutti et al. (2018) and Huang et al. (2021) it is necessary to administer a volume of air higher than the stoichiometric value for the complete combustion of the fuel due to the difficulty of guaranteeing a perfect mixture of the air with the fuel and the limited time of permanence of this in the combustion chamber.

Similar results were found by Meng et al. (2019) in the combustion of blended corn straw and pinewood wastes, increasing air coefficient increased the average peak temperature. Porteiro et al. (2010) burning wood biomass, olive residue, and almond shells reported that for some biomasses, increasing air in the combustion induced a maximum burning rate and maximum temperature. However, for some other biomass, the author observed the excess air induced maximum temperature, but increasing beyond that air coefficient, the burning rate decreased. These results are in accordance with Elorf et al. (2022) and Yang et al. (2004).

Xiu et al. (2018) reported that paper briquettes generated higher temperatures under natural airflow. Their study indicated an average burning temperature of 286°C for waste paper briquettes, which was higher than the temperature of wood briquettes (227°C) but comparable to that of kindling firewood.

Malaťák et al. (2017) found in their studies that combustion of Timothy grass in briquette form at an air coefficient of $\alpha<2$ generated incomplete combustion of biomass with high concentrations of carbon monoxide. In $2<\alpha<3$ was the moment where the CO presented the lowest level; then, excess air coefficient ($\alpha>3$) concentration of carbon monoxide emissions increased again, and the authors attributed this result mainly to low temperature and premature cooling of the flue gas. A similar trend was also found by Eskilsson et al. (2004) and Malaták & Bradna (2017) in burner furnace. Choiński et al. (2023) reported that increasing air coefficient increases CO_2 emissions of combustion of wood pellets in a 25-kW fixed-bed boiler.

3.6. Conclusion

The experimental work conducted in this study showed that:

Pelletizing

1. It is feasible to make pellets from industrial paper residues in this study.
2. The presence of phenolic resin increases the pellets' energy content, making them competitive against other sources like wood biomass.
3. Pellets manufactured from a blend of the remaining papers met the properties requirements for pellets by CANplus certification except for ash content.
4. This pellet mixture had high ash (6.38%) and Ca content (14671 ppm), which would most likely create problems with ash in the combustion system. Hence, pretreatment of the pellets or an appropriate ash handling system must be considered if this pellet is used.

Combustion

The pellets' combustion performance differed between the pellet groups in this work.

1. The combustion temperature of the mixture pellets was determined to be 300ºC and provided ΔT maximum of 232°C, indicating the highest peak of temperature.

2. Their combustion performance was dependent on the pellet composition. Non-phenolic paper pellets burn relatively fast, which indicates the need for a more frequent feed of the combustion system. For phenolic paper pellets, higher temperatures had to be reached for proper combustion. Blending these two groups of paper residues might be a good strategy to improve the energy recovery from these residues through combustion.

3. Higher CO_2 content in the flue gas from the combustion of non-phenolic paper pellets and of mixture pellets.

4. The combustion was influenced by the air coefficient: an increase in this coefficient from 1.0 to 3.0 showed that the pellets oxidate faster and earlier, producing higher temperatures.

This work demonstrated for the first time the technical feasibility of energy recovery by combustion of paper residues from the HPL industry. The results showed the great potential of these residues to displace the use of fossil fuels in the industry.

Funding

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4. ENERGY RECOVERY FROM CANNABIS RESIDUES BY COMBUSTION WITH AND WITHOUT STEAM EXPLOSION PRETREATMENT IN DIFFERENT AIR COEFFICIENTS

4.1. Avant-propos

Auteurs et affiliations:

Rafael Eloy de Souza: PhD Candidate, Université de Sherbrooke, Faculté de génie, Département de génie chimique et génie biotechnologique.

Eduardo Lins de Barros Neto: Professor titular, Universidade Federal do Rio Grande do Norte (Brésil), Département de génie chimique.

Jean-Michel Lavoie: Professor titular, Université de Sherbrooke, Faculté de génie, Département de génie chimique et génie biotechnologique.

Bruna Rego de Vasconcelos: Professor agrégrée, Université de Sherbrooke, Faculté de génie, Département de génie chimique et génie biotechnologique.

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Titre en français: Récupération d'énergie à partir des résidus de cannabis par combustion avec et sans prétraitement par explosion de vapeur à différents coefficients d'air.

Contribution au document: This study explores the energy recovery of green cannabis residues through combustion. It investigates the pelletization process of this biomass and the impact of steam explosion pretreatment on pellet properties. The study also evaluates the effect of the air coefficient on gas emissions during pellet combustion and how the pretreatment influenced the combustion process.

Résumé français: Les résidus de cannabis sont considérés comme une biomasse résiduelle potentielle pour la valorisation énergétique, et l'expansion du marché du cannabis au Canada a créé une niche d'opportunité pour la valorisation de ces résidus. Cette étude vise à évaluer le potentiel de récupération d'énergie à partir des résidus de cannabis par combustion. Une caractérisation chimique de la biomasse a été réalisée. Les résidus de cannabis ont également été prétraités par explosion de vapeur pour observer l'influence du prétraitement sur les propriétés des pellets et sur la combustion. Les résidus de cannabis ont été granulés et ont été combustés à 290°C dans un réacteur à lit fixe. Trois coefficients d'air différents (α=1,0, α=1,15, α=1,3) ont été étudiés. La quantification des gaz de combustion a été effectuée par chromatographie en phase gazeuse et avec un détecteur de NOx. Le prétraitement par explosion de vapeur a amélioré la valeur calorifique et la durabilité des pellets tout en réduisant la teneur en cendres. Cependant, le prétraitement a eu un impact négatif sur la combustion, entraînant une combustion incomplète. Pour la combustion des pellets de cannabis sans prétraitement, le meilleur coefficient d'air était α=1,15, ce qui a conduit à une meilleure conversion de l'oxygène et à une émission plus élevée de CO_2, indiquant une meilleure qualité de combustion. Pour les émissions de NOx, dans les deux traitements, plus le coefficient d'air était élevé, plus les émissions étaient importantes. Ces résultats indiquent le grand potentiel de récupération d'énergie à partir des résidus de cannabis par combustion. Le contrôle des paramètres de combustion s'est révélé essentiel pour obtenir une meilleure qualité de combustion.

Mots-clés : Résidus de cannabis, bioénergie, combustion, explosion de vapeur, granulés.

4.2. Abstract

Cannabis residues have been considered a potential biomass residue for energy recovery, and the increase in the cannabis market in Canada has created an opportunity niche for valorizing these residues. The study aims to assess the potential of energy recovery from cannabis residues via combustion. Chemical characterization of the biomass was carried out. The cannabis residue was also pretreated by the steam explosion to observe the influence of pretreatment on pellets properties and combustion. Cannabis residues were pelletized and combusted at 290°C in a fixed-bed reactor. Three different air coefficients (α) were investigated (α=1.0, α=1.15, α=1.3). Flue gas quantification was performed using gas chromatography and a NO_x detector. Steam explosion pre-treatment improved the calorific value and the pellet's durability and decreased ash content. However, the pretreatment negatively impacted the combustion, leading to an incomplete combustion. For the combustion of the cannabis pellets without pretreatment, the best air coefficient was at α=1.15, leading to higher oxygen conversion and CO_2 emission, indicating better combustion quality. For NO_x emission, in both treatments, the higher the air coefficient, the higher the emission. These results indicate the great potential of the energy recovery from cannabis residues via combustion. The control of the combustion parameters was essential to achieve better combustion quality.

4.3. Introduction

The planet's sustainability is seriously threatened by the increasing demand for energy from burning fossil fuels, which is a concern in many countries worldwide (Gurney et al., 2022). Burning fossil fuels significantly contributes to greenhouse gas emissions (GHG), leading to climate change (Gurney et al., 2022). Alternative methods for energy production that are environmentally friendly and economically viable are being studied to reduce GHG emissions significantly and meet the requirements of the Paris Agreement (Brar et al., 2024).

Bioenergy has emerged as a sustainable alternative to fossil fuel sources, with the potential to meet energy demand while reducing greenhouse gas emissions (Brar et al., 2024). Renewable sources such as forests, agriculture residues and invasive plants have gained attention due to

their potential as feedstock for bioenergy (Ginni et al., 2021). These sources are produced in large volumes, often as residues of a product, making them a cost-effective and carbon-neutral resource for bioenergy production (Ginni et al., 2021).

Cannabis residues hold a particular potential for bioenergy due to the growth of the cannabis industry. The cannabis market reached US$41.6 billion worldwide in 2022 and is expected to have a Compound Annual Growth Rate (CAGR) growth of 15% between 2023 and 2028 (Statista, 2023). Canada was the second country to legalize the production and sale of cannabis, being one of the biggest sellers along with the United States. Since the legalization in 2018, Canada's sales increased from CA$ 151.5 million in the first three months to exceeding CA$ 5 billion in 2023 (Statistics Canada, 2024). A growth of CA$ 6.5 billion is expected by 2025 and CA$ 10.5 billion by 2030, growing at a CAGR of 16% between 2020 and 2030 (Cannabis Business Plan, 2024).

As the demand and annual production of cannabis increase, it is estimated that 70-75% of the output is process residue, amounting to approximately 10.5 t/ha of green waste (Kraszkiewicz et al., 2019). Solid cannabis plant residues such as branches, leaves, and bushes are destroyed using incineration onsite and offsite disposal (Brar et al., 2024). However, this residue can be repurposed as bioenergy, thereby reducing GHG emissions (Parvez et al., 2021).

Cannabis residues stand out as a promising bioenergy resource**,** with a high energy yield of approximately 100 GJ/ha/y, making them potentially competitive with other biofuels (Brar et al., 2024; de Medeiros Dantas et al., 2023). Hemp has an energy yield 120% higher than wheat straw (Prade et al., 2011). When compared to other hemp biofuels like biogas and liquid fuels, they might have the highest yield of energy content, as they are not separated in any way (Parvez et al., 2021; Prade et al., 2011). Solid biofuels could come in densified forms, briquettes, or pellets (Prade et al., 2011). Pellet forms can help with more efficient handling, transportation and combustion. They require low processing for manufacturing and can be sold locally for heating purposes (Prade et al., 2011).

Technologies most applicable for the thermochemical conversion of pellets are combustion, gasification, and pyrolysis (Kpalo et al., 2020). Combustion is the technology most used for

producing energy and heat from biomass, and it is a technique that allows easy adaptation of the already established system of energy conversion technologies (Briones-Hidrovo et al., 2021; REN21, 2020). Moreover, it is a low-cost, highly reliable, and short-term option for biomass utilization (Kpalo et al., 2020). Cambero et al. (2015) showed that small-scale (0.5 MW and 2MW) cogeneration plants using forestry biomass residues from two remote communities in Canada generated the cheapest electricity when compared to a large-scale (5MW) plant. Briones-Hidrovo et al. (2021) evaluated the energy recovery of residual forest biomass by combustion and gasification, and the combustion process returned 17% more energy. According to Carotenuto et al. (2022), this advantage could result in an energy return of 40% higher.

The steam explosion has been suggested in the literature as a pre-treatment for biomass before pelletization to improve bonding characteristics, hydrophobicity, dimensional stability, durability, and higher heating value (HHV) of wood pellets (Alizadeh et al., 2022). Onyenwoke et al. (2022) improved the HHV and tensile strength of steam-pretreated sawdust and oat straw pellets. Yu et al. (2022) mentioned that the equilibrium moisture content of pellets of wood biomass pretreated with steam explosion decreased from 10% to up to 5.9% compared to the non-treated biomass. Reducing the hygroscopicity of pellets prolongs their shelf life and decreases handling and transportation costs (Yu et al., 2022).

Parvez et al. (2021) have highlighted the lack of research on cannabis residues, as the waste is not well known. The performance of this material in thermochemical conversion could be studied, offering more accurate information on its viability as feedstock and expanding its market (Parvez et al., 2021).

The objective of this study was to evaluate the energy recovery of cannabis waste via combustion. The influence of the air coefficient on gas emission and the impact of steam explosion pretreatment on the production of pellets and combustion were also investigated.

4.4. Material and methods

Feedstock

The variety used in this study was the Powdered Donut, with a production cycle of 85 days, consisting of 28 days for the vegetative stage and 57 days for the flowering stage. The production yield was 125g of flower per plant. Cannabis was cultivated in Varennes, Quebec, Canada, and harvested in March, April, and September. The part of the plant used for this study was the plant's stems. The feedstock was characterized as shown in Table 4.1.

Table 4.1 - Physical-chemical characterization of cannabis. Adapted from (de Medeiros Dantas et al., 2023).

Parameter	Value
Moisture Content (w/w)	6.12 ± 0.11
Ash Content (w/w)	3.97 ± 0.28
Extractive Content (w/w)	15.69 ± 0.06
a-Cellulose Content (w/w)	39.55 ± 1.20
Hemicellulose Content (w/w)	22.64 ± 0.60
Lignin Content (w/w)	18.38 ± 0.12
Nitrogen (w/w)	0.77 ± 0.04
Carbon (w/w)	46.91 ± 1.08
Hydrogen (w/w)	6.14 ± 0.12
Sulfur (w/w)	< 0.2
Oxygen (w/w)	43.68 ± 0.94
High Heating Value	17.3 MJ/kg

The cannabis stems were milled in a Ritsch Mill (ZM200) at 6000 rpm, equipped with a 500 μm sieve. Then, a part of the cannabis was pelletized, and the other part was pretreated by the steam explosion and subsequently pelletized. So, two types of cannabis pellets were produced: pellets without steam explosion pretreatment and pellets with steam explosion pretreatment.

Steam explosion pretreatment

A batch pilot unit was used for the steam explosion treatment. The pressure and temperature were 200 psi and 200°C, respectively. First, 200 g of biomass was introduced into the reactor and exposed to the saturated steam at 200°C and 200 psi for a steaming time of 2 min. Then, a

release valve in the reactor was opened to bring the reactor rapidly to atmospheric pressure. The steam-exploded sample was recovered in the bottom vessel and it was dewatered in a laboratory filter press. A controlled pressure of 100 psi was applied for 10 minutes until the water content was reduced to the minimum, and then the samples were frozen to preserve the material. Before pelletizing, the sample was dried in an oven at 105°C.

Pellets production

For this experiment, the CANplus certification (Biomass, 2013) was used as a direction for pellet production, with the goal of obtaining property values as close as possible to what the certification requires to ensure better-quality pellets.

The cannabis residue pelletization was carried out in a pellet mill, a Buskirk Engineering laboratory-scale unit, with a production capacity of 68 kg/hour, which was equipped with a variable-speed drive. The die used for production had a hole diameter of 6.35 mm and a length of 25.4 mm. The die was heated by a silicone rubber heating tape with a time percentage dial control (BSAT) to control the temperature during pellet production. The temperature was set at 70ºC, and the operating temperature ranged between 70ºC and 100ºC. Temperature control during production was measured by a thermal camera Model PTi120, Fluke. After the production, the pellets were cooled down at room temperature and then sieved in a sieve with holes of 3.15 mm in diameter.

Pellets characterization

The Cannabis Pellets were Characterized at the Biomass Technology Laboratory (BTL) to Evaluate their Physical, Thermal, and Chemical Properties.

Dimension

The dimensions of cannabis pellets were assessed using a digital micrometer with 0.001 mm of precision. The pellet diameter and length measurements were taken according to the ISO 17829:2015 standard (International Organization for Standardization, 2015b).

Moisture and ash content

The sample was dried in an oven at 105°C ± 3ºC until it had a constant weight to measure its moisture, as per the standard ISO 18134-1:2015 (International Organization for Standardization, 2015d). The ash content was then determined in the dried sample after complete carbonization through incineration in a muffle furnace at 575°C ± 25ºC, following the standard ISO 18122:2015 (International Organization for Standardization, 2015a).

Mechanical durability

The durability of the pellets is essential since pellets with low durability are sensitive to mechanical stress during transport and handling, which results in the formation of fines. The durability was determined by weighing 100 g of each pellet batch. The samples were set to tumble for 2 min at 50 rpm and then sieved on a 3.15 mm diameter sieve as described in the ISO 17831-1:2015 (International Organization for Standardization, 2015c). The durability was determined according to Equation 4.1.

$$Durability\ (\%) = \frac{m_2}{m_1}\ x\ 100 \qquad \text{(Eq. 4.1)}$$

Where m_1 represents the weight of the sieved pellet before the thumbling procedure, in grams, and m_2 is the mass of the sieved pellet after the completion of the thumbling process, also expressed in grams.

Fines content

The fine content of the cannabis pellets was determined by weighing 0.5 kg of pellets, which were then placed on a 3.15 mm diameter sieve and manually sieved to separate fines from the pellet. Then, fine content and pellet mass were weighted, and fine content was determined according to ISO 18846:2016 (International Organization for Standardization, 2016).

Thermogravimetric analysis

The cannabis pellets underwent thermogravimetric analysis (TGA) using a TA Instruments Q 500 instrument in New Castle, DE, USA. Each treatment was represented by a 50 mg sample. The analysis involved raising the temperature from room temperature to 1000ºC at a rate of 20ºC/min with an air atmosphere with a 90 mL/min flow. The TGA aimed to determine the combustion temperature, which was identified as the temperature corresponding to the peak mass loss in the TGA.

High heating value (HHV)

The high heating value is essential to assess the fuel's energy content. It was measured according to ISO 18125:2017 (International Organization for Standardization, 2017). In this experiment, one grama of sample was positioned in the calorimeter bomb model 6400 from Parr Instruments Moline, IL, USA. The temperature increase and the net calorific value were recorded in megajoules per kg (MJ/kg).

Elemental Analysis

The chemical composition of the pellets, including hydrogen (H), carbon (C), oxygen (O), nitrogen (N), and sulphur (S), was analyzed using a Flash 2000 Organic Elemental Analyzer from Thermo Fisher Scientific. This analysis was essential for determining the process's equivalent air ratio.

Combustion tests

The combustion experiments were performed in a semi-pilot fixed-bed reactor, as shown schematically in Figure 4.1. The combustion system comprises an air supply system by compressed air, a combustion reactor made of stainless steel with a 2-inch and 48 inches long diameter, and a gas cooling system connected to an exhaust and gas sampling system. Manual rotameters regulated the airflow at the entrance, and a digital flow meter was used at the combustion system's end to ensure the airflow mass in the reactions. The air and pellets were injected from the top of the reactor. Thermocouples controlled the temperature in the reactor in three different zones. It introduced 30g pellets into the reactor when the combustion system reached the combustion temperature, and the reaction time was set to 120 minutes.

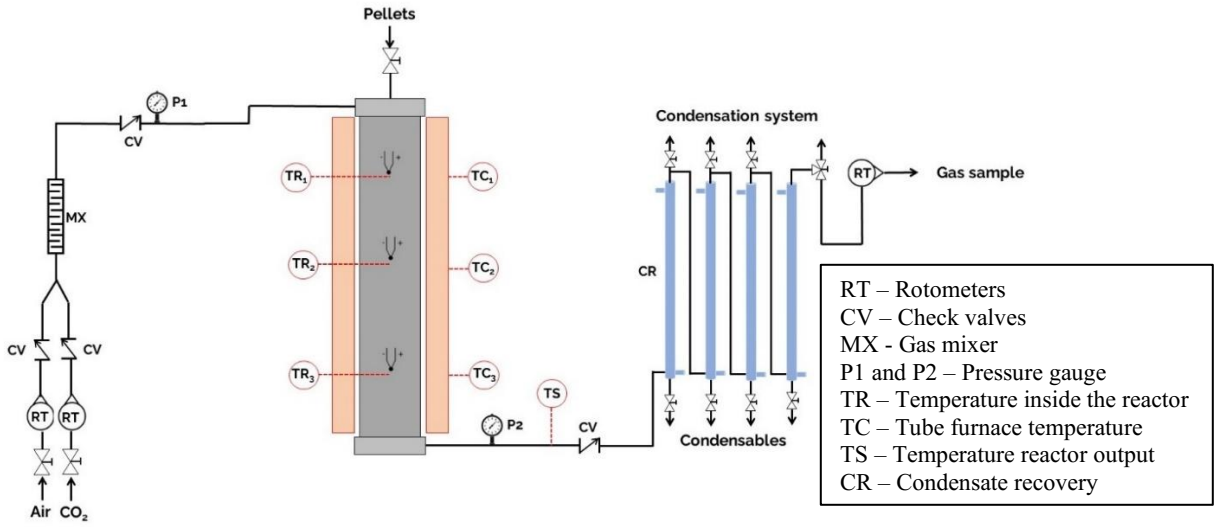

Figure 4.1 - P&ID of the semi-pilot fixed-bed set up for biomass combustion.

The gases generated during combustion were directed into a refrigeration system, with a glycerol solution at -10°C recirculating to cool the gases and separate potential condensable gases. Reactor pressure was monitored using a manometer. Gas chromatography (model Scion 456 GC, Bruker) quantified gases at the reactor's exit. The gas chromatograph (GC) was equipped with two thermal conductivity detectors (TCD) and two columns for gas separation: Molsieve 13X (80/100 mesh, 1.5 m x 1/8" IS) and Hayesep N (80/100 mesh, 0.5 m x 1/8" IS). It had a flame ionization detector (FID) for detecting and separating long-chain hydrocarbons,

equipped with a BR-1 column (10 m x 0.15mm, 2 μm). The measured gases included O_2, CO_2, CO, and CH_4. The NO_x was measured using a NO/NO_x detector (RASI 300C, Eurotron; range 0-5000 ppm).

Equation 4.2 was used to determine the air supply for the combustion of the cannabis pellets, based on the elemental analysis, where "x" and "y" are the coefficients necessary to obtain the stoichiometric air supply according to (Silva et al., 2018).

$$C_{x_1}H_{x_2}O_{x_3}N_{x_4}S_{x_5} + \eta.(3.76N_2 + O_2) \rightarrow aCO_2 + bH_2O + cO_2 + dN_2 + +eCO + fCH_4 + fNO_x + gSO_x \quad \text{(Eq. 4.2)}$$

The excess air coefficient (α) for combustion studied was α=1, α=1.15, and α=1.3, and the volume of air was calculated according to Equation 4.3.

$$\alpha = \frac{V_a}{V_a^o} \quad \text{(Eq. 4.3)}$$

Where V_a is the real volume of air supply for the combustion; V_a^o is the stochiometric air supply volume, and α is the air coefficient.

For the complete combustion, the flue gas is composed by CO_2 and water. However, other chemical reaction involving carbon dioxide and carbon may happen and contribute to the formation of CO, especially in systems where solid carbon is burned, as demonstrated in the Boudouard reaction (Eq. 4.4) (Chien & Chuang, 2011).

$C + CO_2 \rightleftharpoons 2CO \qquad \Delta H_{298}^O - 172.5\ kJ/mol$ (Eq. 4.4)

Dorokhov et al. (2021) have mentioned other parallel reaction that can occur during the combustion of solid fuel that can contribute to the formation of CO, H_2 and CH_4.

$2C + O_2 \rightarrow 2CO \qquad \Delta H_{298}^O - 221.0\ kJ/mol$ (Eq. 4.5)

$C + H_2O \rightarrow CO + H_2 \quad \Delta H_{298}^O\ 131.3\ kJ/mol$ (Eq. 4.6)

$C + 2H_2 \rightarrow CH_4 \qquad \Delta H_{298}^O - 74.8\ kJ/mol$ (Eq. 4.7)

4.5. Results and discussion

Pellets characterization

The properties of the cannabis pellets were carried out after the pelletization process regarding their chemical and physical properties. Figure 4.2 shows the pellets with and without steam explosion pretreatment.

Figure 4.2 - Cannabis pellets. (a) without steam explosion pretreatment, (b) with steam explosion pretreatment.

The cannabis pellets that were not pretreated by steam explosion had a slightly less dark colour than the pretreated pellets. Similar results were found by Lam et al. (2015) in producing pellets made from empty fruit bunch and palm kernel shells and Lam et al. (2012) in the steam-treated biomass of Western Douglas Fir. In both types of biomasses, the pretreatment changed the color of the biomass, and the authors attributed the darker color to a reaction of hemicelluloses and the degradation of extractives in the presence of heat during the steam explosion process. Similarly, Yu et al. (2022) showed that the greater the severity of the treatment, the darker the colour of the pellet.

The pellets' properties were made and compared with the specifications recommended by CANplus certification. Table 4.2 shows the specifications of graded wood pellets based on CANplus requirements (Biomass, 2013). Grade B pellets are used in the industry.

Table 4.2 - Key specifications of graded wood pellets based on the CAN/CSA-ISO 17225 Part 2 Standard (Biomass, 2013).

Property	Unit	Grade B*	Pellets without pretreatment	Pellets with pretreatment
Diameter	mm	6 ± 1 or 8 ± 1	6.13	6.14
Length	mm	$3.15 \leq L \leq 40$	24.5	23.8
Moisture	% mass	≤ 10	6.1	5.2
Ash	% mass	≤ 2.0	4.0	0.9
Durability	% mass	≥ 96.5	95.1	96.7
Fines	% mass	≤ 1.0	1.4	1.0
HHV	MJ/kg	≥ 18.6	17.5	18.6

*CAN*plus* (B): is the grade used for industrial pellets.

The dimensions of the pellets produced, in both cases, are in accordance with CANplus specifications. The values for moisture content are also in accordance with what is required by the certification for Grade B. For the durability test, the pellets without steam explosion pretreatment presented durability values of 95.1%, which is a value close to that required by the certification. For the pellets pretreated, it was found that pretreatment had an impact on durability. The pellets presented durability of 96.7%, fitting CANplus specifications. According to the results of Takada & Saddler (2021), steam explosion increased the durability of poplar pellets by more than 13%, demonstrating that the steam explosion technique has a positive effect on improving this property of the pellets. According to the author, lignin might become partly fluidized and relocated from the cell wall to the fiber surface. Similar results were also reported by Tang et al. (2018), wood pellets of ground poplar increased durability from 77.6% to 97.9% with the steam explosion pretreatment.

The durability of the pellets is essential since pellets with low durability are sensitive to mechanical stress during transport and handling, which results in the formation of fines and makes their use difficult. Pellets with more excellent durability mean denser pellets, with higher energy content, which positively influences the combustion of this material (Machinery, 2014).

Ash content

The ash content, which was 4.0% for cannabis pellets, was reduced to 0.9% after pretreatment with a steam explosion, making the pellets reach the CANplus requirement concerning ash content. This may have occurred due to the expansion of the cellulose fibers during the steam

explosion treatment, which allowed the inorganic part of the material to be leached from the biomass. These results follow what was reviewed by Sarker et al. (2021). Tanase-Opedal et al. (2024) found similar results in their studies with straw of barley grain, a biomass with high ash content. According to these authors, straw with impregnation of water and steam explosed at 200°C for 8 minutes, and 1.6MPa were able to reduce ash content from 5.7% to 2.8% and even more for the straw impregnated with acetic acid (1 wt. %), decreasing from 5.7% to 1.8%. Furthermore, after washing and leaching the straw impregnated with water, ash was reduced from 5.7% to 0.98%. If acid acetic acid was used for presoak, washing, and leaching with water, the ash content was reduced from 5.7% to 0.73%.

High heating value (HHV)

The high heating value of the pellets increased after the steam explosion, going from 17.5 MJ/kg to 18.6 MJ/kg. This value corresponds to an increase of 6.29%. Tang et al. (2018) and Lam et al. (2012) attribute the increase in HHV after steam explosion pretreatment to the chemical modifications that occur in the structure of the biomass and the removal of low-volatility extractives. Different authors have found increased HHV when steam explosion is applied as a pretreatment for biomass. However, the results varied according to the biomass studied. Lam et al. (2015) found a 21% increase in calorific value in pellets made from empty fruit bunch. However, under the same conditions, they obtained an increase of just 2.6% for Douglas Fir pellets.

Onyenwoke et al. (2022) pretreated sawdust and oat straw with a steam explosion for pellet production. The HHV of both biomasses pretreated had an increment of energy content of about 9.5% and 7%, respectively, and it increased according to the severity of the treatment. As described by the same author, any chemical, physical or structural changes in biomass can potentially cause an increase in HHV due to changes in small carbon molecules. During the steam explosion, molecules with low volatile energy density can be removed by autohydrolysis, and the carbonization of biomass during the steam explosion can increase the concentration of this energy.

Thermogravimetric Analysis (TGA)

The TGA is a well-known technique for studying combustion behavior (Liu et al., 2021). Figure 4.3 (a) shows the TG and derivative thermogravimetric (DTG) curves of cannabis pellets without pretreatment. The TG curve presents the mass loss with the temperature increase, and the DTG is a derivative weight loss of the TG. The combustion of the cannabis pellets happened in 3 stages of thermal degradation. The first one consists of a weak peak at the beginning of the DTG that corresponds to removing the moisture content. It started around 25°C and ended at 52.2°C, corresponding to a mass loss of 0.83%. The second stage was the most significant mass loss of 76.46% at 289.5°C. That stage corresponds to the decomposition of the cellulose and hemicellulose. The decomposition ranged from 178°C to 300°C. The third stage, a short peak, corresponds to the combustion of the remaining char. The mass loss peak happened at 312.5°C with a mass loss of 11.7%.

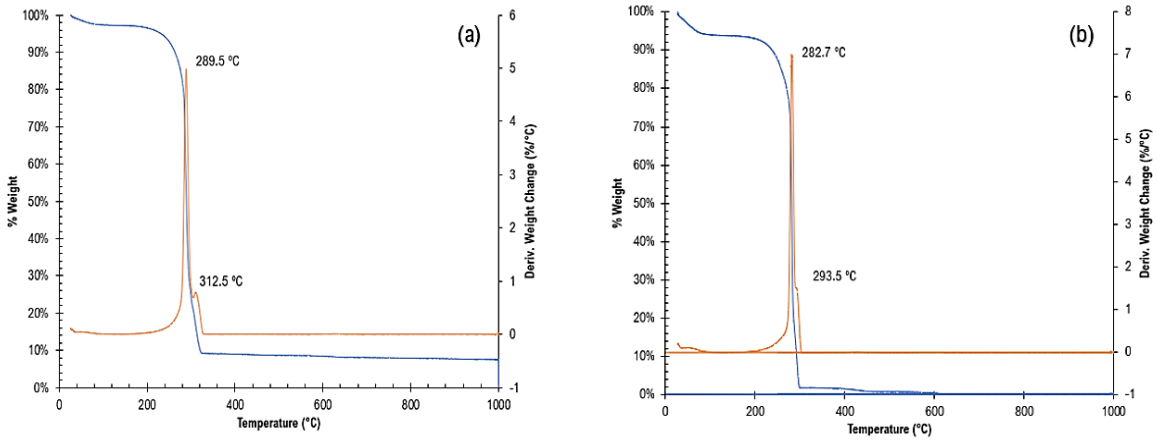

Figure 4.3 - TGA of cannabis pellets under an air atmosphere at 20°C/min. (a) without pretreatment and (b) pretreated by steam explosion.

Figure 4.3 (b) shows the TG and derivative thermogravimetric (DTG) curves of cannabis pellets pretreated with the steam explosion. The first stage corresponds to removing the moisture content, starting around 28.7°C and ending at 100°C, corresponding to a mass loss of 1.84%. The second stage, corresponding to cellulose and hemicellulose degradation, presented a mass loss of 80% and occurred at 282.7°C. This stage ranged from 180°C to 290°C. The third stage,

the combustion of the char remaining, had a higher mass loss at 293.5°C and ranged from 291°C to 300°C with a mass loss of 11.9%. These results showed that the steam explosion pretreatment shifted the curve of the second stage of the TG to the left, decreasing the decomposition temperature of the second stage when compared to the pellet without pretreatment. The temperature difference was 7°C, which might be considered insignificant between the treatments.

Elemental analysis

The steam explosion treatment had an impact on the elemental composition of cannabis. The carbon, hydrogen, oxygen, and nitrogen percentages in untreated cannabis biomass were 46.91%, 6.14%, 43.68% and 0.77%, respectively (Table 4.3). After pretreatment with the steam explosion, there was an increase in the carbon and nitrogen content to 47.5% and 0.85%, respectively. The concentrations of hydrogen and oxygen decreased to 5.55% and 42.97% respectively.

Table 4.3 - Elemental composition of cannabis pellets without pretreatment and with pretreatment.

Elemental Analysis (%)	Pellets without pretreatment	Pellets with pretreatment
C	46.91	47.50
H	6.14	5.55
O	43.68	42.97
N	0.77	0.85
S	< 0.2	< 0.2

Onyenwoke et al. (2022) also reported a change in the elemental composition of biomass treated with the steam explosion. They registered an increase in carbon content from 47.47% to 48.53% for sawdust and 44.24% to 44.89% for Oat straw (in conditions similar to this study). There was a reduction in hydrogen content from 6.66% to 6.43% (sawdust) and 6.05% to 5.92% (Oat straw). Oxygen went from 45.41% to 44.11% and 43.87% to 42.65% for sawdust and oat straw, respectively. The same tendency was found by He et al. (2020) for steam-treated rice straw at 200 °C for 10 min; according to their results, the carbon content increased from 42.14% to

46.47% while hydrogen decreased from 5.60% to 5.35% and oxygen decreased from 51.45% to 47.40%.

Anglès et al. (2003) reported that steam explosion treatment may cause lignin carbonization and even condense, contributing to increased carbon content. The reduction of oxygen and hydrogen Onyenwoke et al. (2022) and He et al. (2020) explain that loss could be attributed to carbon dioxide and water formation.

Emissions

Figures 4.4 and 4.5 present the outlet concentrations of measured gases as a function of time in the three different air coefficients: $\alpha = 1.0$, $\alpha = 1.15$, $\alpha = 1.3$.

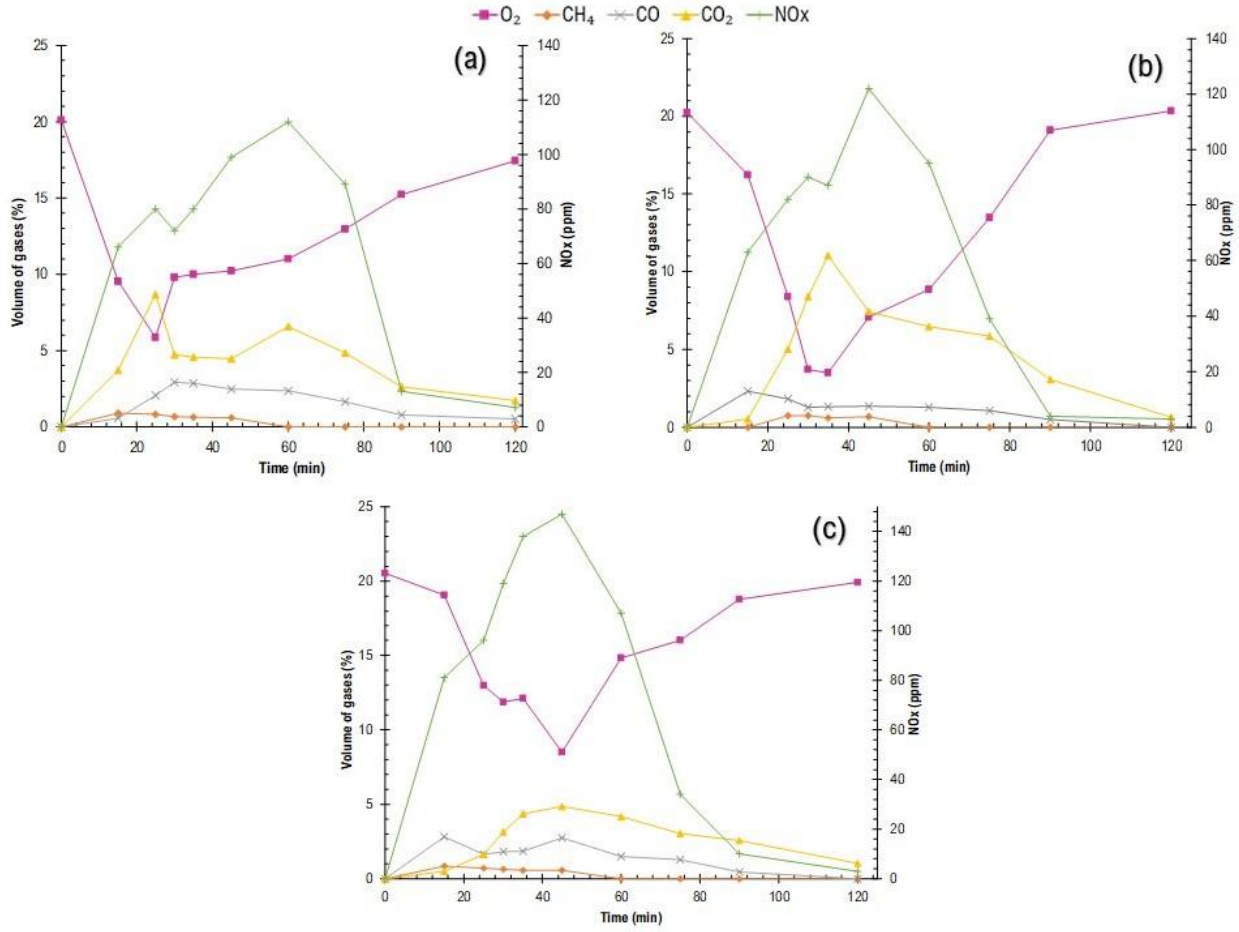

Figure 4.4 Influence of the air on gas emission on the combustion of cannabis pellets without pretreatment at (a) α = 1.0, (b) α = 1.15, (c) α = 1.3 at 290°C.

The experiment evaluated the impact of air coefficient values (α=1, α=1.15, and α=1.3) on the combustion and gas concentration of the two types of pellets. The results showed that the air supply impacted gas production during the combustion of cannabis pellets without steam explosion pretreatment, as shown in Figure 4.4. The flue gas composition indicated incomplete combustion when CO is formed, while the presence of CO_2 and low concentrations of CO and CH_4 indicated better combustion quality. The air coefficient of α=1.15 showed the best results for the combustion of the pellets with a low level of oxygen (3.5%) and a high concentration of CO_2 (11%) in the flue gas. The air coefficient of α=1.0 did not provide enough air for combustion, resulting in a minimum concentration of oxygen (5.8%) and a maximum production of CO_2 (9%). This result was expected since the literature shows that an airflow

higher than stoichiometric is necessary for solid fuels to ensure a better mixture of air and fuel (Caposciutti et al., 2018; Huang et al., 2021). The air coefficient of α=1.3 had an excess of air, leading to the production of CO and CH_4, probably CO was due to temperature loss by convection, which characterizes incomplete combustion, and methane by equilibrium reaction.

The higher the air coefficient, the lower was the CO content in the flue gas. The CO concentration in the flue gas was 2.85%, 1.83%, and 1.74% for α=1, α=1.15, and α=1.3, respectively. However, the lowest concentration of CO in the α = 1.3 air coefficient may be due to incomplete combustion of the pellets, as 8.5% of oxygen was still present in the flue gas.

Regarding the methane concentration (CH_4), all reactions ceased at 60 minutes, and the average concentration values were 0.6%, 0.49%, and 0.55% for α=1, α=1.15, and α=1.3, respectively. The NO_x production increased with the increase of air coefficient. The maximum peak values were 112 ppm for α=1.0, 122 ppm for α=1.15, and 147 ppm for α=1.3. It was expected that the NO_x emission as these gases is formed during the combustion of fuel that contains nitrogen in the composition because it promotes the oxidation of the nitrogen in the fuel (Xiu et al., 2018). Díaz-Ramírez, Sebastián, Royo et Rezeau (2014) studied the influence of air supply in the NO_x emission of 3 pelletized energy crops, such as *Brassica carinata*, *Populus* sp., and a blend of them, and found that higher air supply and temperature increases the specific NO_x emissions. The same tendency was found by Malaťák et al. (2017) for emissions of meadow hay and timothy grass.

For the combustion of pellets with steam explosion pretreatment (Figure 4.5), similar behavior was observed regarding the influence of the air coefficient on combustion. However, the combustion quality was affected compared to the combustion of pellets without steam explosion pretreatment.

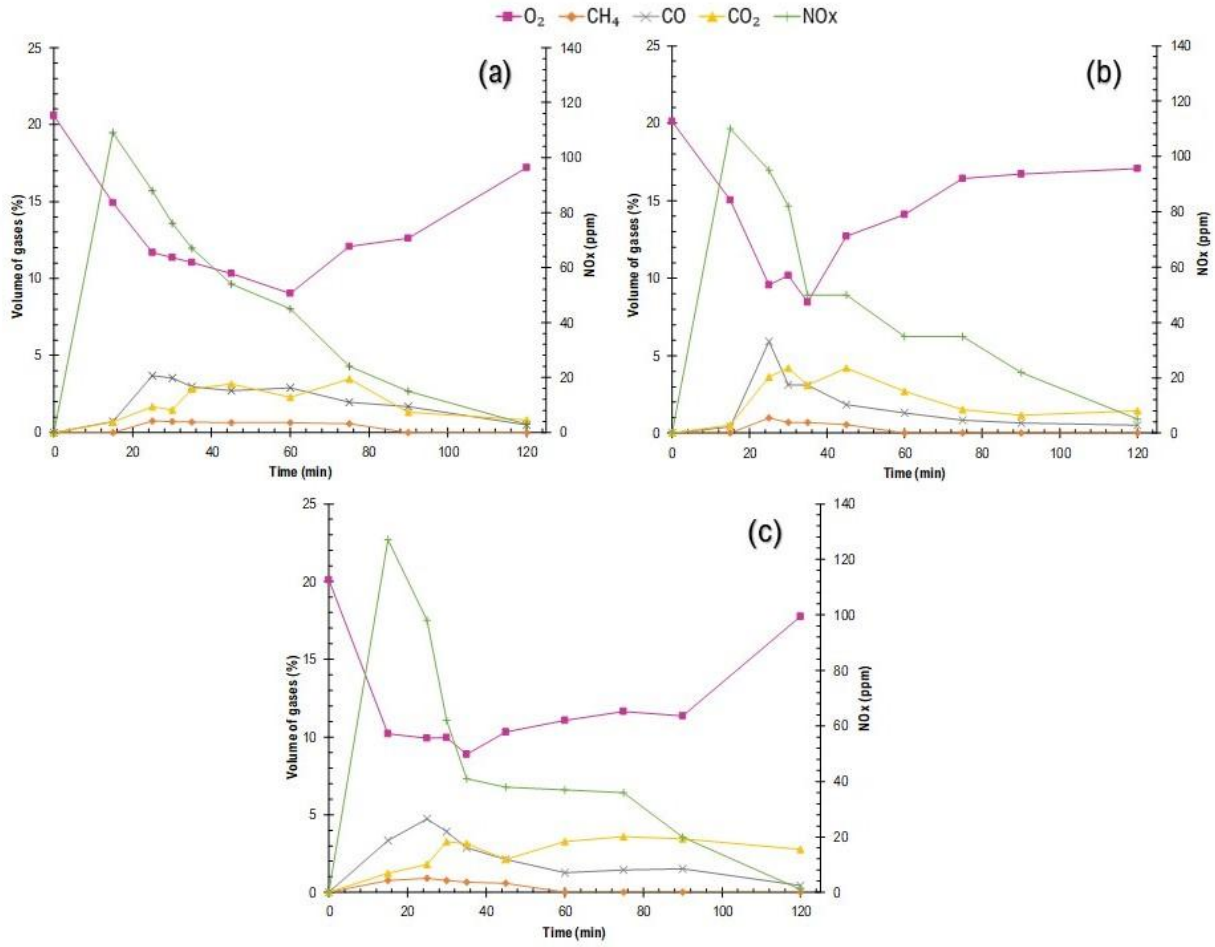

Figure 4.5 Influence of the air on gas emission on the combustion of cannabis pellets with pretreatment at (a) α = 1.0, (b) α = 1.15, (c) α = 1.3 at 290°C.

All air coefficients presented similar oxygen values in the flue gas with values of 8.4%, 9%, and 8.9% for α=1, α=1.15, and α=1.3, respectively. The three air coefficients showed higher CO production than CO_2. The peak CO production was 3.69%, 5.92%, and 4.74%, respectively, for α=1, α=1.15, and α=1.3. CH_4 was present in the composition of the gases with average concentration values of 0.5%, 0.49% and 0.62% for α=1, α=1.15, and α=1.3, respectively. The NO_x emission had maximum peak values of 109 ppm for α=1.0, 110 ppm for α=1.15, and 127 ppm for α=1.3. These results indicate the need to improve the combustion of these pellets for better combustion quality.

Biswas et al. (2011) reported that the fuel and combustion properties are affected by steam explosion treatment because it changes the chemical composition of the raw material. As evidenced in this work, although steam explosion pretreatment improved the properties of the pellets in terms of their calorific value and ash content, combustion was affected by the pretreatment.

Biswas et al. (2011) found significant differences in the pellets' char combustion. The time of char combustion, produced from several batches of pellets after pyrolysis, showed a constant increase with the severity of the steam explosion process. The greater the severity of the treatment, the longer the combustion time of the char under combustion conditions at 900°C and 21% O_2.

Several factors can support or explain this behavior. Sharma et al. (2004) reports that the steam explosion treatment alters the properties of the wood, where there is a rupture of cellulose and hemicellulose and the percentage of lignin is considered to increase, causing higher lignin content in the char. This lignin may cause a partial blocking of the pores of the char or decrease the reactivity of the char due to softening, melting and carbonization of lignin. Biswas et al., (2011) also reports that steam explosion can reduce the volatile materials of the raw material, which contributes to the reduction of the reactivity of the char due to the reduction of pores caused in the previous pyrolysis process. Matsumoto et al. (2009) have reported that the reduction in char reactivity may be related to the alkali metal content in the biomass.

Even though steam explosion improved the calorific value, durability, and ash content in the pellets, it also induced some disadvantages in the combustion process. Steam explosion is discussed in the literature in the impact of the treatment on pellet properties; however, according to Yu et al. (2022) and Zhang et al. (2022) steam explosion still has much space for improvement and further research is required to justify the dominant reason for the reduced char reactivity.

4.6. Conclusion

Cannabis residues could be a potential source of solid biofuel. The steam explosion pretreatment had an impact on the characteristics of the pellets produced by cannabis residues and on the colour of the cannabis residues. The biomass presented a darker colour than the non-treated biomass. The steam explosion improved pellet durability, increasing it from 95.1% to 96.7%. Ash content decreased from 4% to 0.9%. The high heating value of the pellets increased from 17.5 MJ/kg to 18.6 MJ/kg. After this pretreatment, the pellet's properties were in accordance with the CANplus certification.

Regarding the combustion behaviour of pellets non-treated by steam explosion, it was observed that the air coefficient influenced the emission levels, with the optimum at $\alpha = 1.15$. At this air coefficient, higher oxygen conversion and CO_2 production were observed, which indicates better combustion quality. NO_x emissions increased with the increase of the air coefficient. However, the combustion behaviour of pellets pretreated by steam explosion showed higher CO production and lower emission of CO_2, which indicated incomplete combustion at all air coefficients. That shows the need for optimized conditions for burning pellets treated by steam explosion.

These proved for the first time the potential for recovering energy from cannabis residues through combustion. Future research must focus on optimizing combustion conditions, understanding pellet reactivity modified by the steam explosion pre-treatment and understanding the combustion behaviour.

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5. CONCLUSION

5.1. Conclusion (en français)

Dans le contexte actuel du changement climatique, la transition énergétique visant à réduire les émissions de gaz à effet de serre et la dépendance aux ressources fossiles a été au centre de plusieurs études au sein de la communauté académique. L'objectif est de trouver des initiatives capables de décarboner les secteurs du transport, de l'industrie et de l'agriculture, pour n'en citer que quelques-uns. De plus, la réduction de la génération de déchets par ces secteurs a également été un sujet d'étude. Le concept d'économie circulaire a gagné en importance, car il encourage le recyclage, la réutilisation et la réduction des déchets générés, couramment observés dans les secteurs mentionnés. En tant que stratégie technologique et d'innovation pour le traitement et le recyclage des déchets, la conversion thermique est en cours de développement et d'application.

Ce travail a évalué la faisabilité technique de la valorisation de deux types de résidus solides industriels pour la récupération d'énergie par combustion. Le premier résidu provient du processus de production de stratifiés haute pression, actuellement disponibles sur le marché. Le deuxième résidu est un résidu solide vert provenant de la production de cannabis.

La première partie de ce travail visait à étudier l'efficacité de la valorisation énergétique des déchets de papier par combustion contrôlée à différentes températures et avec différents apports d'air pour identifier les conditions optimales permettant de maximiser la récupération d'énergie. Pour cette étude, des pellets ont été produits à partir d'un mélange de résidus fournis par l'industrie, et leurs propriétés ont été comparées aux exigences du standard CANplus. L'étude a montré que des pellets à haute valeur calorifique pouvaient être produits à partir des résidus de l'entreprise. Leurs propriétés respectent les spécifications CANplus, à l'exception de la teneur en cendres. La température de combustion étudiée avec le TGA a efficacement brûlé les pellets produits dans un réacteur à l'échelle de laboratoire. De plus, la quantité d'air fournie a significativement influencé la combustion. Un apport d'air plus élevé a entraîné une augmentation du delta de température obtenu à partir des pellets pendant la combustion. Cela signifie qu'ils pourraient fournir des températures plus élevées avec des coefficients d'air plus

élevés. Les résultats ont montré que le pellet mélangé offre une efficacité de combustion significative ; par exemple, dans les mêmes conditions expérimentales, les pellets composés de résidus mixtes ont produit le delta de température le plus élevé (ΔT 232°C) par rapport au papier phénolique ou au papier kraft lorsqu'ils étaient utilisés seuls et dans les mêmes conditions. Cela suggère que la combinaison des différents résidus peut améliorer la performance de combustion des pellets. De plus, il a été démontré que plus la teneur en phénol des résidus est élevée, plus la température de combustion doit être élevée, ce qui indique l'importance de connaître la teneur en résine dans le mélange.

La deuxième partie de ce travail a évalué la récupération d'énergie des résidus de cannabis par combustion. Elle a également étudié l'influence du coefficient d'air sur les émissions de gaz et l'impact du prétraitement par explosion de vapeur sur la production des pellets et sur la combustion. La partie de la plante de cannabis utilisée pour fabriquer les pellets était la tige. Un prétraitement par explosion de vapeur a été réalisé pour évaluer son effet sur les pellets et son influence sur la combustion. Le profil des émissions de gaz à différents coefficients d'air a également été évalué. La caractérisation et la combustion des pellets de résidus de cannabis indiquent un grand potentiel de récupération d'énergie. Le prétraitement par explosion de vapeur a amélioré la valeur calorifique et réduit la teneur en cendres des pellets. Cependant, le traitement par explosion de vapeur a induit des effets sur le processus de combustion des pellets, entraînant une combustion incomplète. Le coefficient d'air étudié a montré que le coefficient α=1,15 a donné les meilleurs résultats de combustion, avec une consommation d'oxygène plus élevée et un pic d'émission de CO_2 plus élevé, indiquant une meilleure qualité de combustion. Le coefficient d'air a eu un impact sur la production de NO_x, avec des coefficients d'air plus élevés entraînant des émissions de NO_x plus élevées. La combustion des pellets avec explosion de vapeur n'a pas montré une bonne conversion de l'oxygène, avec des niveaux d'émissions de CO_2 plus faibles et des concentrations plus élevées de CO, considéré comme un gaz de combustion incomplète. Ces résultats indiquent que le prétraitement par explosion de vapeur a amélioré la qualité des pellets. Cependant, il a eu un impact négatif sur la combustion des pellets prétraités.

Cette recherche montre le potentiel de valorisation des résidus solides industriels par combustion. Une combustion efficace de ces résidus pourrait apporter des avantages doubles

pour atteindre les objectifs de gestion durable des déchets et de récupération d'énergie. De plus, la réduction de la dépendance aux combustibles fossiles pour l'approvisionnement en énergie contribue à la réduction des émissions de GES provenant de sources non renouvelables, ce qui peut réduire les coûts liés aux crédits carbones, bénéficiant ainsi à la fois à l'économie et à l'environnement.

Bien que des résultats prometteurs aient été obtenus dans cette étude, la teneur en cendres des pellets doit être prise en compte dans le processus. La teneur en cendres entraînera une plus grande fréquence de maintenance des équipements de combustion en raison de la formation de scories dans le système, ce qui peut réduire l'efficacité de la combustion. Les systèmes de post-traitement des gaz, tels que la réduction des NO_x et des SO_x, doivent être envisagés pour la valorisation des résidus par combustion.

En conclusion, les résidus de papier et de cannabis ont un potentiel significatif pour la valorisation par combustion. Aborder les défis liés à la teneur en cendres et mettre en place des systèmes de contrôle des émissions efficaces peuvent offrir une solution viable pour la gestion des déchets et la production d'énergie, contribuant ainsi à un avenir plus durable et plus efficace en termes de ressources.

5.2. Conclusion (in English)

In the current context of climate change, energy transition with the aim of reducing greenhouse gas emissions and reducing dependence on fossil resources has been the focus of several studies in the academic community. The objective is to find initiatives that can decarbonize sectors of transportation, industry, and agriculture, to mention some. Moreover, the reduction of waste generation by these sectors has also been the focus of studies. The concept of circular economy has gained importance, as it encourages recycling, reuse, and reduction of waste generated, commonly found in the sectors mentioned. As a strategy of technology and innovation strategy for waste treatment and recycling, thermal conversion is being developed and applied.

This work assessed the technical feasibility of valorizing two types of solid residues from industry for energy recovery by combustion. One residue comes from the production process of high-pressure laminates, which are currently on the market. The second residue is a green solid residue from cannabis production.

The first part of this work aimed to investigate the efficiency of energy recovery from paper waste through controlled combustion at different temperatures and air supply to identify optimal conditions for maximizing energy recovery. For this study, pellets have been produced from a mixture of residues provided by the industry and their properties were compared to CANplus requirements. The study has shown that high calorific pellets could be produced with the residues from the company. Their properties fit CANplus specifications, except for ash content. The combustion temperature studied with TGA efficiently burned the produced pellets in a lab bench-scale reactor. Moreover, the amount of air supply significantly affected the combustion. A higher air supply resulted in a higher delta temperature obtained from the pellets during the combustion. It means that they could provide higher temperatures with higher air coefficients. Results have shown that the mixture pellet offers significant combustion efficiency; for example, under the same experiment conditions, the pellets composed of mixture residues produced the highest delta temperature (ΔT 232°C) compared to phenolic paper or kraft paper when they were used alone and in the same conditions. This suggests that combining the different residues can enhance the combustion performance of the pellets. Moreover, it was evidenced that the higher the amount of phenolic content in the residues, the higher the

combustion temperature needs to be, indicating the importance of knowing the resin content in the mixture.

The second part of this work evaluated the energy recovery of cannabis residues via combustion. It also investigated the influence of the air coefficient on gas emission and the impact of steam explosion pretreatment on the production of pellets and combustion. The part of the cannabis plant used to make the pellets was the stem. A steam explosion pretreatment was conducted to assess its effect on the pellet and its influence on combustion. The gas emission profile at different air coefficients was also evaluated. The characterization and combustion of the cannabis residue pellets indicate great potential for energy recovery. The steam explosion pretreatment improved the calorific value and decreased the ash content of the pellets. However, the steam explosion treatment induced effects on the combustion process of the pellets, leading to incomplete combustion. The studied air coefficient demonstrated that the coefficient of α=1.15 yielded the best combustion results, with higher oxygen consumption and a higher peak emission of CO_2, indicating better quality combustion. The air coefficient impacted NO_x production, with higher air coefficients resulting in higher NO_x emissions. The combustion of pellets with steam explosion did not exhibit good oxygen conversion, with lower levels of CO_2 emissions and higher concentrations of CO, which is considered a gas of incomplete combustion. These results indicate that steam explosion pretreatment improved pellet quality. However, it negatively impacted the combustion of the pretreated pellets .

This research shows the valorization potential of industry solid residues through combustion. Effective combustion of these residues could provide dual benefits toward achieving goals for sustainable waste management and energy recovery. In addition, reducing dependence on fossil fuels for energy supply contributes to reducing GHG emissions from non-renewable sources, which may reduce costs with carbon credits, benefiting both the economy and the environment.

Although promising results have been obtained in the study, the pellets' ash content must be considered in the process. The ash content will lead to a greater frequency of maintenance of the combustion equipment due to the formation of clinkers in the system, which can reduce combustion efficiency. Post-treatment gas systems, such as NO_x and SO_x reduction, shall be considered for the residue valorization through combustion.

In conclusion, the paper and the cannabis residues have significant potential for recovery through combustion. Addressing the ash content challenges and implementing effective emission control systems can offer a viable solution for waste management and energy production, contributing to a more sustainable and resource-efficient future.

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