

1 Introduction

Bioenergy is converted from available organic waste into useful heat, power, and fuels, thus closing material loops in a circular economy. Recent studies show how thermochemical and biochemical processes (combustion, gasification, pyrolysis, fermentation) valorize biogenic streams to reduce waste and increase resource efficiency [1–3]. Within solid bioenergy, densified fuels such as wood pellets and briquettes have become the main form due to their uniform geometry and high bulk density, which simplify handling, storage, and long-distance transportation. Further quality-related properties, such as durability, ash content, energy density, and heating value, determine end-use performance [4]. Wood pellets typically achieve $>16 \text{ MJ kg}^{-1}$ with low ash content, whereas agricultural residues help broaden feedstock supply but often require tighter control of ash-forming elements and binders [5,6].

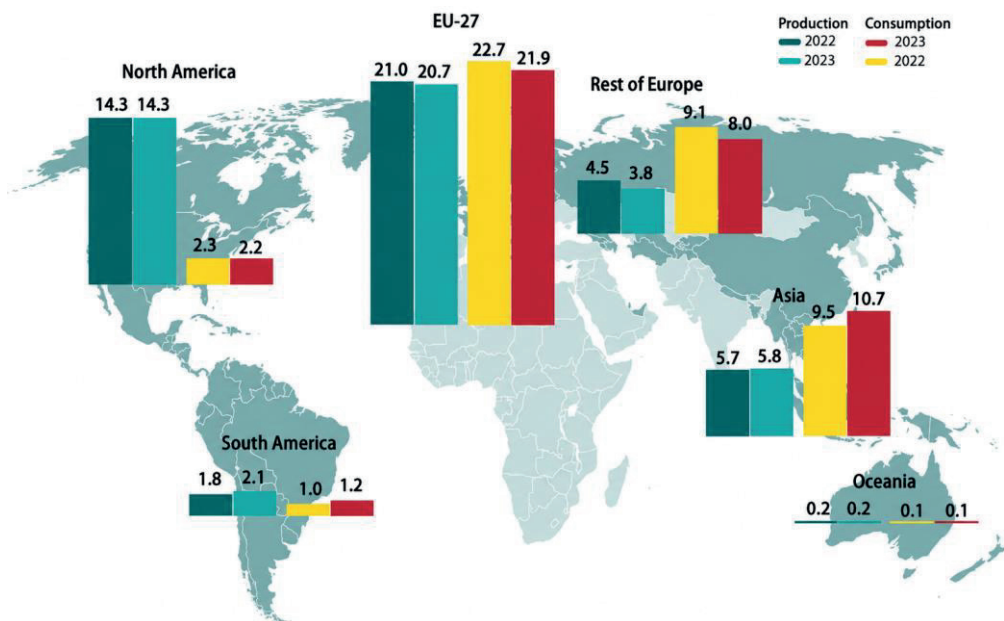


Figure 1-1: Global wood pellet production and consumption (in million tons) in 2022 and 2023 [7].

Global pellet production reached about 43 million tons (Mt) in 2022, with regional imbalances in supply and demand (Figure 1-1). The European Union represents the global leader by producing 20.6 Mt in 2022 and consuming the largest share at about 22 Mt, particularly for residential heating, and co-firing in power plants [7]. North America (the USA and Canada) accounts for 14.3 Mt, most of which



is exported to Europe and Asia. In Asia, pellet use has expanded rapidly, with consumption rising from 9.5 Mt in 2022 to 10.7 Mt in 2023, driven primarily by co-firing in coal-fired power plants in Japan and South Korea and by household heating programs in Northern China. In South America, production remains modest at 1.8-2.1 Mt, but Brazil, Chile, and Colombia are emerging exporters. In contrast, Africa and Oceania still play only marginal roles, with production <0.3 Mt. Pellets are therefore used across scales: in automatically fed domestic stoves and boilers, in commercial and industrial heat processes, and for utility applications including co-firing, thus leveraging standardized fuels to reach high appliance efficiencies and reduce lifecycle greenhouse emissions. As pellet technologies advance and supply chains professionalize, pellets are framed as a flexible option to enhance energy security and rural economies while reducing dependence on imported fossil fuels [5,8].

Densifying forestry biomass and agro-industrial by-products into higher-value energy carriers mitigates waste, enables consistent fuel quality for modern combustion systems, and supports the goal of a circular bioeconomy. Life-cycle assessments in Latin America indicate that well-configured pellet and briquette supply chains can deliver low energy and emissions intensities per unit of delivered energy. However, results depend on biomass sourcing, plant configuration, and modeling choices (e.g. land-use and soil carbon accounting) [9,10]. At the same time, real-world deployment underscores the importance of policy design, fuel standards, and robust logistics. In 2022, volatile energy markets shifted pellet use towards residential and commercial heating, which accounted for 56 % of EU consumption, which was the highest share since 2014 [7]. Experience from rural Northern China shows that household pellet heating programs depend on reliable feedstock collection, fuel-quality certification, and cost-reflective support. Without these, field emissions can diverge from lab performance and adoption slows. These insights show that pellet market growth is not just a technological question but also a coordination challenge across supply, appliances, and governance [9,10].

Taken together, the literature suggests biomass pellets as a practical, scalable solution in the energy transition. A standardized solid fuel that integrates residue streams into clean heat and power applications, aligns with circular-economy principles, and, when paired with fit-for-purpose standards and policies, can deliver tangible environmental and socioeconomic benefits.



1.1 Motivation

In contrast to loose biomass particles, pellets are favored due to their high bulk density, easier handling, and predictable behavior during thermochemical conversion. Their uniform shape and mechanical integrity facilitate efficient transport, long-term storage, and performance during combustion, pyrolysis, and gasification processes. Although strict requirements (uniform geometry, low water content and high durability) ensure a minimum pellet quality after production, subsequent humidity fluctuations and mechanical stresses throughout their logistics chain can substantially alter pellet structure. Nevertheless, the extent to which structural changes occur under different stress conditions is strongly influenced by the processing parameters applied during pelletization. Numerous studies have examined the effects of parameters such as particle size distribution and water content of wood shavings, die temperature and compaction pressure on pellet density and mechanical stability. However, detailed insight into the internal microstructural heterogeneity of pellets, which is decisive for their failure mechanisms, remains limited. Compaction in open press channels is a complex process that can induce pronounced radial and axial variations in porosity within individual pellets. This heterogeneous porosity distribution determines both the severity and the mode of pellet failure under the stress conditions encountered during handling, storage, and further processing. Moisture exposure is a key driver of structural evolution during pellet storage, as even moderate variations in water content affect the polymeric matrix and inter-particle bonding, leading to swelling, internal cracking, and irreversible changes in density and porosity. Experimental investigations, including studies conducted within the scope of this thesis, demonstrate that pellets subjected to cyclic humidity conditions do not fully recover their initial structural and mechanical properties upon re-drying [11,12], with direct consequences for fines generation, flowability, and energy density. These parameters influence the dynamics of subsequent processes, such as pyrolysis, a thermochemical process that yields volatiles and pelletized chars, which are subsequently gasified to produce synthesis gas. Furthermore, pellet chars must resist mechanical loads during transportation from the pyrolysis reactor to the gasification reactor. During pyrolysis, heat and mass transfer within the compact pellet geometry govern devolatilization behavior and, consequently, the resulting pellet char morphology. While loose biomass particles can be elutriated unreacted, pellets incur lower losses and often develop characteristic structural features, such as central cavities, anisotropic porosity gradients, and layered morphologies [13], which may be closely linked to the densification characteristics, internal density gradients, and orientation of wood shavings determined during pelletization.

Although the pelletization process, storage effects, and pyrolysis have each been investigated extensively as individual processes, they need to be examined within a unified methodological framework. As a result, a comprehensive understanding of how pelletization conditions alter the initial microstructure that governs both moisture-induced behavior during storage and devolatilization during pyrolysis lacks. Therefore, a systematic characterization of pellets from production through humidity exposure to thermochemical conversion, while clearly distinguishing the processes and conditions governing each stage, is necessary.

1.2 Objective and outline

To achieve this, model systems were established that enable both fundamental investigations under well-controlled conditions and transfer to industrially relevant setups. Because closed-die wood tablets are frequently investigated in the literature and their mechanical properties are often considered representative of industrial wood pellets, they were also analyzed in this study. Precise control of densification parameters during tableting enabled the study of the viscoelastic behavior of wood components, such as lignin, spring-back, and fiber orientation. To complement these idealized systems, single open-die pellets were produced to replicate the heterogeneous structure and the U-shaped layered orientation of the fibers in technical pelletization. By systematically varying press channel length (D/L ratios of 1:3, 1:4, and 1:5) and die temperature, the influence of densification pathways on relaxed density, porosity distribution, and mechanical strength was elucidated. Direct comparisons between tablets and pellets further enable assessment of the suitability of each model system for industrial wood pellets, thereby linking fundamental insights to practical relevance. Furthermore, industrial wood pellets were produced using a flat-die press. Dies of different press channel geometry (same D/L ratio as in single open-die experiments) were used, and process parameters such as die temperature, throughput, and specific energy consumption were recorded. The impact of the press channel diameter-to-length (D/L) ratio on the density distribution, layer-shape orientation of wood shavings, and the mechanical strength of wood pellets was analyzed in detail. The flat-die press also enabled large-scale production, ensuring sufficient material availability for subsequent storage and pyrolysis studies. Beyond production, the thesis addresses structural changes induced during storage under fluctuating humidity, emphasizing moisture sorption, swelling, crack propagation, and the partial recovery of strength after subsequent re-drying. Sorption behavior is assessed using equilibrium isotherms fitted with the Guggenheim-Anderson-de Boer (GAB) model, complemented by μ CT-based porosity evolution and mechanical stability tests at different water contents. The findings reveal whether



- and to what degree - pellets still meet the relevant standard requirements after being exposed to such storage conditions. The impact of thermal stress on pellet structure is investigated through comparative pyrolysis experiments conducted in a fluidized-bed reactor (FLB) and a control setup (CS). Here, structural and mechanical degradation of pellet chars are quantified, highlighting the formation of central cavities, pine-cone-like morphologies, and the role of fines enrichment in the pellet core. Special attention is given to methodological challenges in porosity analysis, such as closed pores, large cracks, and entrained sand particles. By combining μ CT with complementary density-based bookkeeping, consistent porosity estimates and sand-corrected char properties are obtained, enabling a robust interpretation of the effects of densification and pyrolysis on pellet-char structure.

The thesis is structured into six chapters. Chapter 1 introduces the motivation, objectives, and overall scope of the work. Chapter 2 presents the basic principles and current state of the art regarding wood pellets as a biomass fuel. It covers the relevant wood components governing pellet properties, feedstock preparation, pelletization processes, and operating parameters, including the roles of press-channel geometry, water content, particle-size distribution, and pressure and temperature. Furthermore, handling-related aspects, such as sorption characteristics and mechanical properties during storage, are discussed, followed by an overview of the fundamentals of thermal conversion, with a focus on biomass pyrolysis. Chapter 3 presents the fundamentals of X-ray micro-computed tomography (μ CT) and image processing, which serve as the central non-destructive characterization techniques throughout this thesis. Chapter 4 describes the materials and experimental methods, including wood-shaving preparation; compaction processes using closed-die, single-open-die, and flat-die presses; pellet conditioning; fast pyrolysis experiments; and the characterization methods applied. These methods include moisture determination; dynamic vapor sorption particle size distribution analysis; density and porosity measurements; swelling behavior during storage; laser scanning microscopy; mercury porosimetry; μ CT analysis; and mechanical stability testing, including compression, three-point bending, durability, and impact tests. Chapter 5 presents the results and discussion. It first introduces the model systems under investigation, beginning with tablets produced in closed dies, then pellets from single open-die and flat-die pelletization, and summarizes their key characteristics. The influence of die geometry on pellet properties in flat-die pelletization is then analyzed. Subsequently, the effects of storage on wood pellet properties are examined, including sorption behavior, mechanical properties at varying water contents, re-drying and recovery behavior, and their implications for pellet stability. Finally, the chapter addresses structural integrity during pyrolysis,



detailing the transformation from wood pellets to pellet chars, challenges in pellet-char characterization, and the mechanical properties of pellet chars. Chapter 6 summarizes the main findings of the thesis and provides concluding remarks. It highlights methodological advances in pellet and pellet-char characterization and discusses implications for pellet production, storage stability, and thermochemical conversion.

Within the framework of this thesis, several peer-reviewed publications [11,12,14,15] were produced. Their results are integrated into the corresponding chapters and complemented by additional, previously unpublished data.



2 Basic principles and state of the art

2.1 Wood pellets as biomass fuel

Wood pellets represent a climate-friendly alternative to fossil fuels, offering high energy density and significantly lower carbon dioxide emissions [6]. Compared with loose wood shavings, they offer advantages in handling and pose a lower risk of dust explosions [16–19]. According to ISO 16559, pellets are defined as “densified biofuels made from fragmented biomass with or without additives, usually cylindrical with broken ends and a length between 3.15 and 40 mm” [20]. For small-scale combustion systems, ISO 17225-2 specifies that only pellets with a diameter of 6–8 mm are suitable [4].

To withstand the mechanical loads encountered during transportation and storage, pellets must exhibit high mechanical durability. Pellets with insufficient durability are prone to breakage, thereby increasing the fines fraction in the bulk material. This in turn reduces energy density, impairs flowability, and lowers storage capacity. Durability is commonly assessed using a pellet tumbler test, which simulates mechanical stresses during conveying [21–23]. According to the DIN EN ISO 17225-2 standard [4], pellets must reach a durability of at least 97.5 % to be certified as ENplus A1, representing the highest quality grade. Nevertheless, even pellets that meet this standard may undergo structural and stability changes during prolonged storage under fluctuating humidity.

To ensure consistent quality across the supply chain, the ENplus certification scheme was introduced in Europe in 2010 and has since become the most widely implemented system, covering production, logistics, storage, and delivery. ENplus distinguishes three quality classes:

- ENplus A1, requiring raw materials from debarked roundwood or untreated sawmill by-products, with ash ≤ 0.7 wt.-%, nitrogen ≤ 0.3 wt.-%, and durability ≥ 98.0 %.
- ENplus A2, allowing forest residues and bark, with ash ≤ 1.2 wt.-%, nitrogen ≤ 0.5 wt.-%, and durability ≥ 97.5 %.
- ENplus B, designed for industrial pellets from untreated recovered wood, with ash ≤ 2.0 wt.-%, nitrogen ≤ 1.0 wt.-%, and durability ≥ 97.5 %.

For all quality classes, the maximum water content is limited to ≤ 10 wt.-%, and the fines fraction (<3.15 mm) must not exceed 1.0 wt.-% at the factory gate. In



addition, minimum ash softening temperatures are prescribed to prevent slagging and corrosion in combustion appliances [4,24].

Taken together, these standards and certification schemes highlight the importance of correlating pelleting conditions, mechanical pellet properties, and storage stability to guarantee consistent performance across different stages of the biofuel supply chain.

2.2 Components of wood relevant for pellet properties

In practice, wood pellets are produced from a variety of raw materials, including softwoods, hardwoods, and in some cases herbaceous biomass, and the intrinsic composition of these feedstocks strongly influences pellet quality. Wood is a hierarchical biopolymer composite composed of an interconnected network of cellulose, hemicelluloses, and lignin. On a dry weight basis, wood cell walls are dominated by carbohydrates (65-75 %), primarily cellulose and hemicellulose, combined with lignin (18-35 %). The elemental composition of dry wood averages ~50 % carbon, ~6 % hydrogen, ~44 % oxygen [25,26], and inorganics [27], while freshly harvested biomass can contain up to 45-50 % water. Microscopically, wood tissue is arranged in a highly ordered and interconnected network of cells, consisting mainly of axial tracheids and vessel elements for fluid transport, and ray cells for radial transport and storage. Cell walls are divided into the middle lamella, primary wall, and a three-layered secondary wall [28,29]. The middle lamella is highly lignified and binds adjacent cells, while the central layer of the secondary wall contributes most to mechanical strength. The density of the wood cell wall remains relatively uniform at about 1.46-1.56 g cm⁻³ [30,31]. The main structural polymers also exhibit distinct intrinsic densities: cellulose ~1.50-1.58 g cm⁻³, hemicellulose ~1.45-1.55 g cm⁻³, and lignin ~1.25-1.45 g cm⁻³ [32]. Softwoods generally contain 33-42% cellulose, 22-40 % hemicellulose, 27-32 % lignin, and 2-3.5 % extractives [33,34], whereas hardwoods contain 38-51 % cellulose, 17-38 % hemicellulose, 21-31 % lignin, and about 3 % extractives [34,35]. In comparison, herbaceous plants exhibit greater compositional variation, with 0-40 % lignin, 20-50% hemicelluloses, 25-95% cellulose, and 4-9% extractives [36,37]. Cellulose provides tensile strength through its crystalline fibrils; hemicelluloses act as a flexible, amorphous matrix that promotes fiber bonding; and lignin functions as a hydrophobic binder that strengthens and protects the cell wall. While cellulose, hemicelluloses, and lignin dominate the binding behavior of woody biomass, comparisons with feed pelleting research provide useful insights for understanding densification mechanisms. In animal feed, starch and protein soften under thermal and



moisture treatments, facilitating compaction and improving pellet hardness [38,39]. Fibers contribute in different ways depending on their properties: soluble fractions enhance cohesiveness by increasing viscosity, whereas coarse insoluble fibers interrupt the bonding network and can act as fracture initiation sites [40]. Lipids generally counteract binding by inhibiting hydrogen bonding between adjacent polymers [39]. In wood pellets, elevated extractive levels can lower die friction during pelletization but at the cost of creating weak interfacial layers that reduce durability [41]. However, studies have shown that isolated or chemically modified lignin significantly enhances both the durability and energy value of biomass pellets [42,43]. A key explanation for these improvements is the glass transition of lignin. When heated above its glass transition temperature (T_g), lignin changes from a brittle, glassy material to a rubber-like state with increased chain mobility, thereby lowering viscosity and enhancing interdiffusion under compression [44,45]. Upon cooling below T_g , secondary interactions are re-established, locking particles into a consolidated structure [46].

Beyond these general polymeric effects, different feedstocks show distinct pelletization behaviors. For example, softwoods and hardwoods generally yield dense, durable pellets with low ash contents. However, softwoods require greater milling energy due to their density, while hardwoods with higher acetylated hemicelluloses can release acetic acid during processing, increasing die wear [38]. Herbaceous feedstocks such as wheat straw, switchgrass, and corn stover contain higher levels of silica and alkali compounds, reducing durability and increasing slagging and fouling tendencies during combustion [47]. Agricultural residues such as olive tree pruning can produce pellets of good density and strength when water content is kept around 9-12 wt.-%, but stability declines at higher values, as hydrogen bonds between polymers are replaced by water bonds [22]. Similarly, investigations of barley and wheat straw, corn stover, and switchgrass confirmed that a water content of ~12 wt.-% and fine comminution (<1 mm) increased density and mechanical strength [48]. Agro-industrial by-products, such as rapeseed or soybean cakes, contain intrinsic proteins and fats that act as lubricants and binders during pelletization but increase water affinity and reduce storage stability. In contrast, nutshells or husks often form brittle pellets due to poor moisture retention, while citrus residues, rich in pectin, enhance cohesiveness and hydrophobicity [49].

Taken together, these examples demonstrate that pellet quality cannot be explained by chemical composition alone, but results from a close interplay between feedstock type and processing history. Thus, even when biomass polymers have the potential to bind, the effectiveness of densification depends

critically on how the raw materials are prepared before entering the pellet press. This makes feedstock preparation a decisive stage in the production chain.

2.3 Feedstock preparation

In practice, feedstocks enter the pelletization chain with widely varying initial conditions ranging from freshly harvested roundwood with high water content to sawdust, agricultural residues, or agro-industrial by-products with distinct particle sizes, morphologies and chemical additives. These differences necessitate systematic preparation steps (Figure 2-1) such as **drying**, **milling**, and **conditioning**, each of which directly influences the activation of natural binders, the homogeneity of the feedstock, and the efficiency of densification. Accordingly, the following section addresses feedstock preparation in detail, with emphasis on the key physical parameters water content (wc) and particle size distribution (PSD).

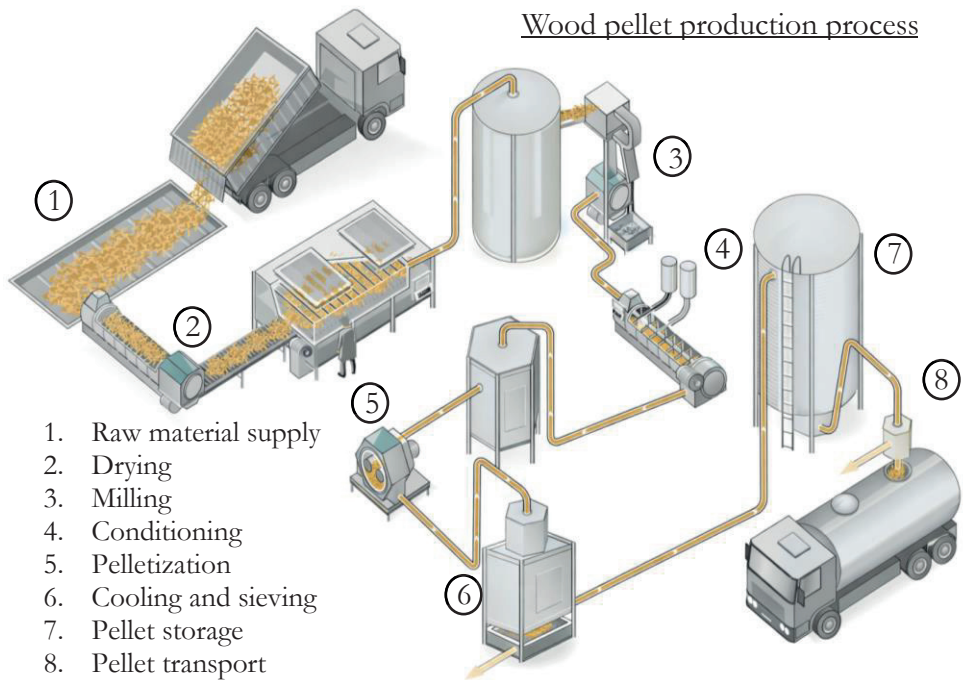


Figure 2-1: Schematic representation of the industrial wood pellet production process including raw material supply (1), drying (2), milling (3), conditioning (4), pelletization (5), cooling and sieving (6) with subsequent storage (7) and transportation (8) (modified after [52]).