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Review

Understanding the impacts of inorganic species in woody biomass for preprocessing and pyrolysis–A review

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ABSTRACT

Woody biomass represents an abundant resource for sustainable biofuels, biochemicals, and bioproducts. Technologies for converting woody biomass have been established for decades, and research consistently highlights the critical role of inorganic species and ash plays in feedstock handling and conversion processes, including equipment plugging, corrosion, and catalyst deactivation. A thorough understanding of the variability, transport behavior, and downstream impact of inorganic species in woody biomass is essential for defining feedstock quality specifications and developing effective management strategies for conversion processes. This review compiles critical information in five main sections: 1) inorganic species concentration in woody biomass, based on anatomical fractions and their sources of variability; 2) technique features for quantifying inorganic elemental chemical analysis; 3) impacts of inorganic species on biomass preprocessing; 4) impacts of inorganic species on pyrolysis, and 5) mitigation strategies. Additionally, this review explores future challenges and opportunities in addressing the impacts of inorganic species on biomass quality. These insights aim to support the sustainable development of the biomass-to-bioenergy pipeline and ensure high-quality lignocellulosic feedstocks for efficient downstream conversions. The findings offer valuable guidance to policy makers, industry stakeholders, and researchers in developing effective strategies for managing inorganic species in woody biomass and fostering the sustainable processes for lignocellulosic biorefineries.

1. Introduction

Production of biofuels and bioproducts from lignocellulosic feedstock has attracted considerable attention due to its abundance, carbon-neutral features, and potential to promote circular economy [1]. The 2023 Billion-Ton Report released by the U.S. Department of Energy (DOE) predicted the U.S. could produce 1 billion dry tons of biomass per year. This biomass can be converted to biofuel, biopower, and bioproducts to displace one-third of the nation's petroleum consumption [2]. By 2040, the total available biomass for bioenergy is about 826 million dry tons, with a roadside cost of \$60/dry ton. More than 15 % comes from woody energy crops. Compared to the northern and western regions of the U.S., the southern region provides more biomass, potentially more than 68 % (about 57–78 million dry short tons) [3].

Lignocellulosic biomass is composed of heterogeneous organic polymers, including carbohydrates and aromatic lignin along with smaller amounts of inorganic constituents with variable compositions. While the organic components, cellulose, hemicellulose, and lignin, in plant biomass are converted into fuels and chemicals, the inorganic components, such as ash, present challenges for preprocessing and conversion operations. The presence of inorganic components could lead to equipment wear and failure [4]. Beyond equipment wear and

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abrasion, ash and inorganic compounds could contribute to plugging, fouling, and slagging, which reduce product yields and increase conversion costs. Therefore, understanding the inorganic composition of lignocellulosic feedstocks and its impact on biomass processing is critical to biorefinery success [5–9].

Inorganic matters in biomass include 1) crystalline mineral species such as silicates, oxyhydroxides, sulfates, carbonates, chlorides, and nitrates, 2) poorly crystallized mineraloids, and semi-crystalline species; and 3) non-crystalline or amorphous species [10]. The woody biomass can contain over 20 % inorganic components, including heavy metals such as iron- and nickel-containing compounds [11]. The thermal transformation of lignocellulosic material may liberate alkaline metals, like silica or chlorine (Cl), resulting in reactor corrosion or toxic emissions into the atmosphere. Moreover, a high biomass ash content can result in equipment wear and unexpected downtime [12]. However, inorganic compounds also act as catalysts in pyrolysis and gasification.

Alkali metals and alkaline earth metals (AEMs) in biomass pose additional challenges for thermal conversion processes. The consequential alkali vapors generated from biomass thermal conversion are a critical problem, as they increase facility maintenance costs and reduce equipment lifespan. Potassium (K), one of the most common alkali metals in biomass, is present as oxides, chlorides, and sulfates, which can lead to slagging and fouling during thermal conversion [13]. Moreover, high alkali metal and ash contents significantly lower the efficiency of biomass pyrolysis and increase net power consumption [14]. Hence, understanding the inorganic elemental composition, chemical forms, and distribution within plant tissues is crucial for improving the efficiency of biomass thermal conversion and developing effective ash-mitigation strategies [15].

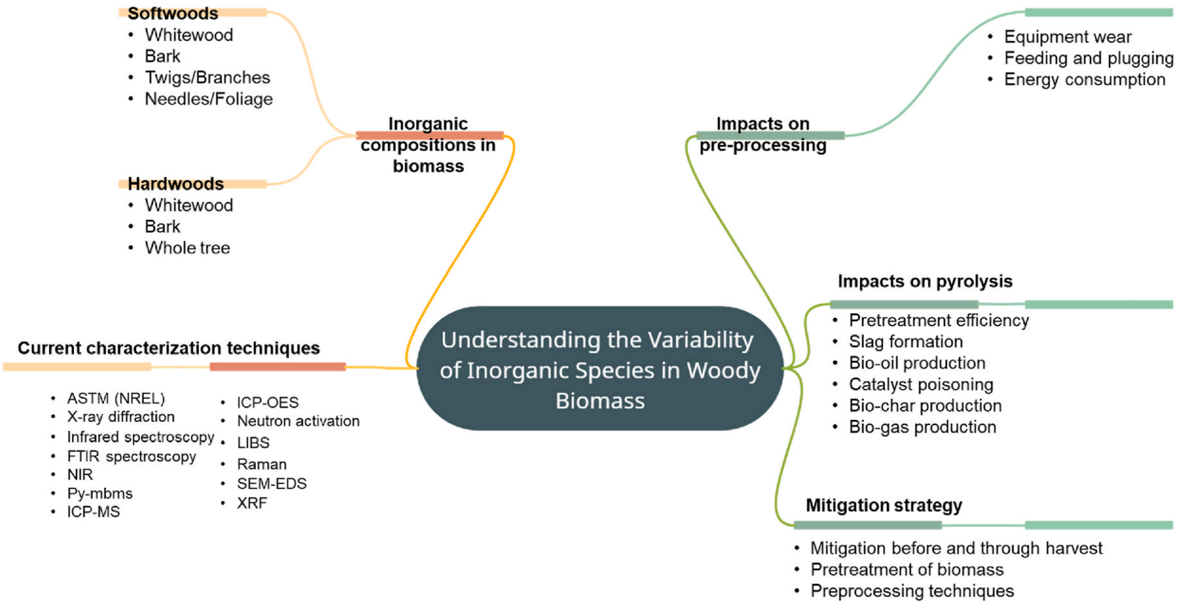
This review offers insight into the sources of lignocellulosic biomass variability related to biorefinery unit operations, the unique characteristics of inorganic species at anatomical and tissue scales, and their effects on biomass handling, feeding, process configuration, and conversion into fuels and chemicals. The focus is specifically on woody feedstock materials, including pine, willow, poplar, and mixed forest residues. This study highlights (1) the inherent variability of intrinsic and extrinsic inorganic species in biomass; (2) the features, advantages, and limitations of existing characterization techniques; (3) the impacts of inorganic species on preprocessing operations; (4) their effects on biomass pyrolysis; (5) potential strategies for mitigating inorganic components; and (6) future research needs and perspectives (see Scheme 1). The goal of this review is to identify knowledge gaps to optimize biomass utilization and mitigate negative effects on feedstock-to-bioenergy through thermochemical conversions.

2. Variability and characterization methods for inorganic species

Biomass contains significant ash constituents, including alkali and alkaline earth metals, such as silicon (Si), chlorine (Cl), and sulfur (S). Ash content can be divided into intrinsic and extrinsic ash. Intrinsic ash includes Si, micronutrients (e.g., Iron (Fe), Manganese (Mn), Zinc (Zn), Copper (Cu), Boron (B), Cl, and Molybdenum (Mo)), and macronutrients (e.g., Nitrogen (N), Phosphorus (P), K, Calcium (Ca), Magnesium (Mg), and S). These elements accumulate naturally in plants through uptake, transport, or assimilation depending on their availability in the soil [16]. In contrast, extrinsic ash is introduced from soil, dust, and other contaminations accumulated during harvest and collection [10]. Plants have intrinsic inorganic species that mediate biological mechanisms during growth and transport. Additionally, plants collect physiological inorganic components from the soil, and soil amendments, which consist of the essential macronutrients (Ca, K, S, Mg, N, P) [17], micronutrients (Zn, Fe, Mn, Cu, Cl, B, Mo, and Nickel (Ni)) [18,19], and other non-essential elements such as Sodium (Na) and Si [20–25].

2.1. Intrinsic inorganic species

Table S1 shows the total ash content in softwood and hardwood biomass categorized by anatomical fractions, including whitewood, branches, bark, needles, and leaves. Woody biomass contains relatively low ash content compared with herbaceous biomass. The average ash content in softwood biomass is 0.2–1.1 wt %, while in hardwood biomass, it is slightly higher, around 2 wt %. Differences in ash content are related to tree age and growing conditions, including the nutrient contents in the soil [17]. The composition of inorganics in different woody biomass is shown in Fig. 1. Woody fractions mainly contain Ca and significant quantities of K, both essential nutrients for tree metabolism [26–31]. Smaller amounts of Si, Aluminum (Al), Mg, P, and Mn are also present. Mineral nutrients are transported from the soil to tree tissues through xylem cells. The inorganic compounds migrate through the heartwood to sapwood and cambium into mature cell walls [26,32]. Therefore, the inorganics are unevenly distributed throughout the tree



Scheme 1. Schematic of connections among review topics and coverage in the literature.

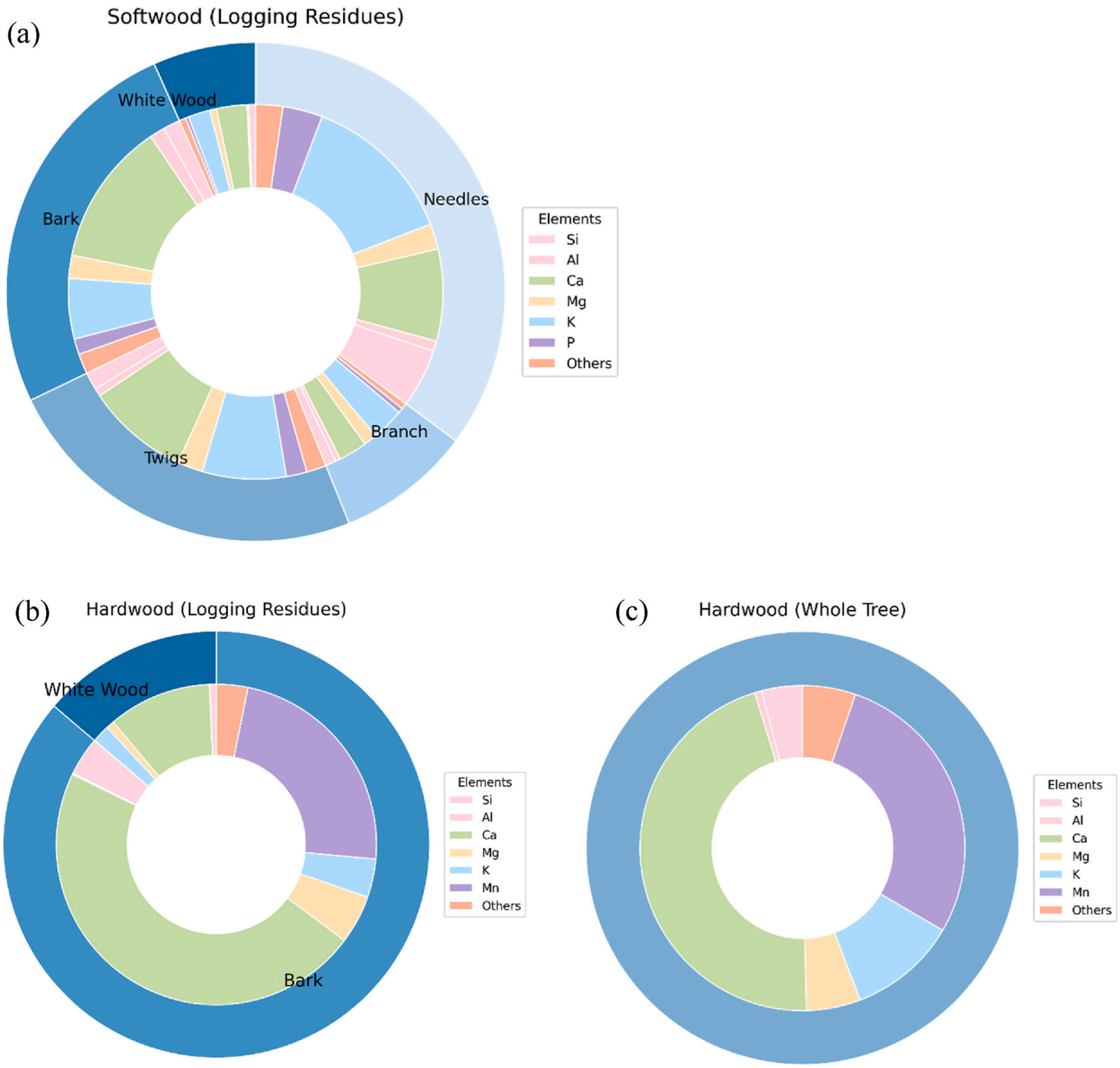


Fig. 1. Inorganic compositions in woody biomass anatomical fractions for (a) softwood logging residues, (b) hardwood logging residues, and (c) hardwood whole tree biomass (Detailed information in SI).

tissues, which necessitates the study of inorganic composition at the tissue or anatomical-fraction scale. Different plant parts—tissues and anatomical fractions, may be best suited to particular uses based on their chemical compositions and physical properties.

Fig. 1 and Table S1 show the ash content in different anatomical fractions of woody biomass. In softwood (Fig. 1a), inorganic compounds are more concentrated in bark (up to 7.1 %), foliage (~5.2 %), and needles (up to 2.2 %). The ash content in the whitewood of pine includes Ca (~600 ppm) and K (407–710 ppm). Bark fractions have over four times higher elemental contents with Ca (2390–6530 ppm) and K (1600–3180 ppm) as the dominating species, followed by P (1260 ppm), Mg (874 ppm), and Mn (343 ppm). However, their Cl contents (147 ppm) are only moderately higher when compared with the whitewood (85 ppm). The needles had the highest K (4770 ppm). The concentration of P, K, Cl, and Mg increases from whitewood to twigs, bark, and needles. However, the ash content is lower in Douglas fir, with Ca being the dominant component and abundant in the bark. For spruce, the inorganic contents are low across the whole tree, and the ash composition of different anatomical fractions is similar to that of pine.

The inorganic composition of hardwood is shown in Fig. 1b and c. The Ca is significantly higher in hardwoods than softwoods and is located mainly in the bark. The highest concentrations of inorganic elements were observed in different anatomic fractions, with Ca predominantly found in the bark, and K primarily located in the needles. In contrast, the elements found in the lowest concentration are Na and Fe [33]. The inorganic species K, P, and Mg, enriched in the woody biomass anatomical fractions, are mobile elements [34]. They are readily soluble in sap solutions and translocate between different parts of the trees [35,36]. The bark contains more inorganics than the whitewood because the bark collects inorganic impurities like sand and soil [37,38]. Therefore, it is suggested that the bark be treated as a distinct feedstock [39].

2.2. *Extrinsic inorganic species*

Extrinsic ash typically comes from rocks, soil, and other forms of contamination acquired during harvest, collection, transportation, and processing. Therefore, minimizing introduced contamination from soils during forestry harvest and materials handling is beneficial.

Fig. 2 illustrates the elemental analysis of anatomical fractions as received, using pine as a representative feedstock due to its prevalent use in industry. It also shows the chemical composition of the extrinsic, soil-derived inorganic particles adhered to the biomass through surface interactions while harvest and transportation. Extrinsic inorganic compounds were recovered from as-received dry biomass using a

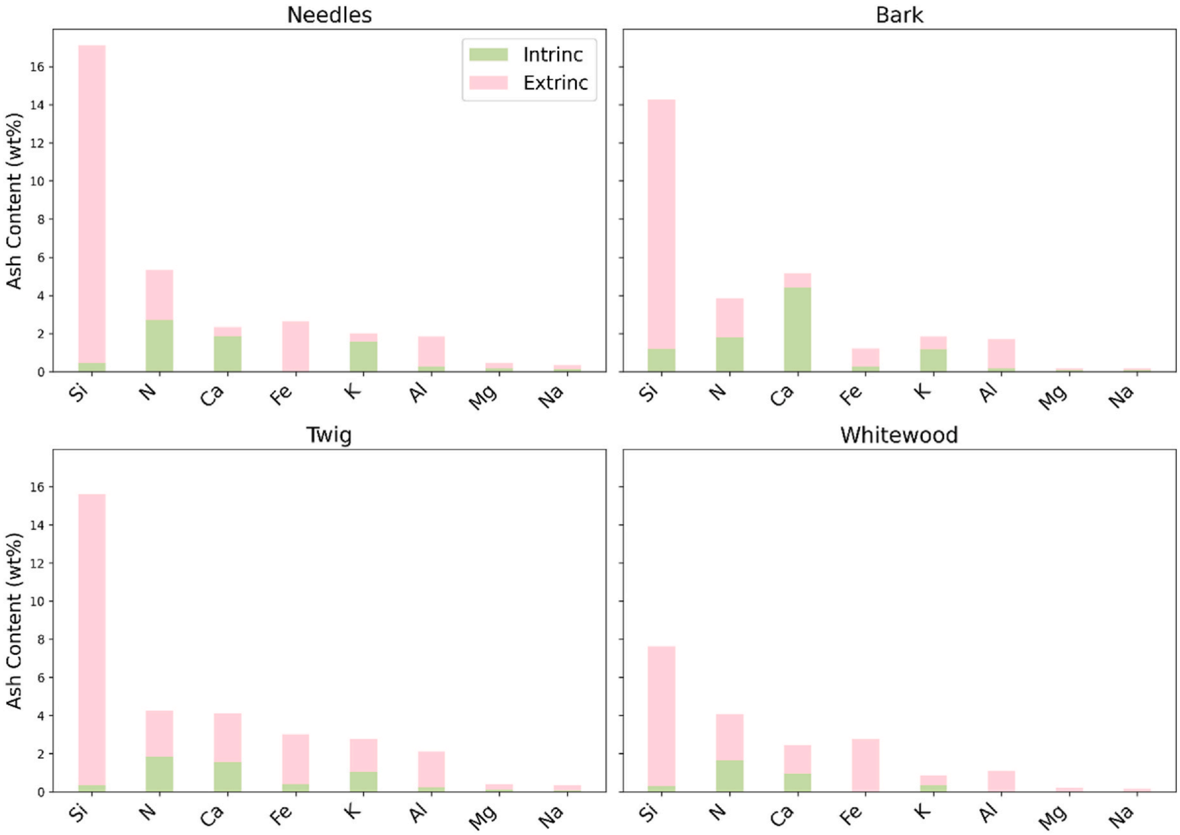


Fig. 2. Chemical compositions of pine anatomical fractions and their extrinsic inorganic compounds (modified from Ref. [4]); extrinsic inorganic compounds were separated from as-received dry biomass samples using a composition-preserving, sieving-sonication process. [Note: No standard deviation (% STDEV) was reported in this reference.].

composition-preserving method involving sieving, and sonication to separate a mixture of organic debris and inorganic dirt particles, as described by Lee et al. [4]. Extrinsic inorganic compounds were measured using newly developed sieving-sonication method to extract extrinsic inorganic compounds without altering their compositions. As shown in Fig. 2, the presence of extrinsic inorganic compounds at least doubles the total inorganic contents in biomass compared to the as-received (intrinsic inorganics) biomass samples. This pattern is particularly observed from Si; the extrinsic Si content is about 11 times higher than as-received content in bark and about 44 times higher in twigs. Similarly, the extrinsic Fe and Al contents are about 13 and 9 times higher, respectively.

The levels of extrinsic inorganic matter increased due to contamination during the processes of harvest, collection, and storage. For example, Si along with Fe, Al, Na, and Titanium (Ti) can be introduced during biomass harvest as clays, sand, and other soil contaminations [40–48]. Lee et al. [4] performed the characterization of inorganic compounds relative to the plant microstructure and found that Si exists in lower concentrations in the epidermis than in the inner tissues (0.14–0.18 wt% vs. 0.36–0.87 wt%). However, their scanning electron microscopy (SEM) energy dispersive x-ray spectroscopy (EDS) maps revealed higher content of Si in the needles (0.65 wt%) than the other anatomical fractions (0.11–0.24 wt%). Additionally, bark has the second-highest Si content (0.24 wt%), while twigs and whitewood fractions show similar levels of 0.11–0.15 wt%. These results highlight the significant presence of inorganics in biomass following processing.

High extrinsic inorganics can lead to operational difficulties [49]. Si can pose operational challenges due to its presence in quartz, which causes abrasion in handling and feeding equipment. Moreover, its presence as silicates with other inorganics can result in slagging during biomass combustion for bioenergy conversions [50,51].

2.3. Characterization techniques

Using a thermogravimetric analyzer to exam weight loss in biomass under 750 °C is the standard approach for determining ash content, as developed by National Renewable Energy Laboratory (NREL) and American Society for Testing and Materials (ASTM E1755-01) [52,53]. These conventional methods require labor-intensive sample preparation, high reagent costs, and time-consuming data generation. Additionally, they cannot differentiate between extrinsic and intrinsic ash [54]. However, a recent study reported non-destructive methods that distinguish between extrinsic and intrinsic ash in lignocellulosic by combining X-ray fluorescence (XRF) and near-infrared (NIR) spectroscopy [55]. Advanced characterization tools for inorganic species in woody biomass are essential for understanding the sources of variability and their distribution in plant tissues, as well as for quantifying their impact on lignocellulose preprocessing and conversion processes. Such knowledge can guide the design and development of mitigation strategies to manage the impacts of inorganics on biomass conversions [52]. Moreover, the concentration of the inorganic presents in biomass could be more accurately identified using non-destructive characterization techniques compared to conventional high-temperature ash characterization [56]. Thus, non-destructive tools that provide both quantitative and qualitative elemental analysis, along with rapid and reliable assessment of inorganic material attributes, can facilitate the identification of biomass type and origin while guiding quality management for subsequent conversions [50,57].

2.3.1. Conventional ash characterization

Table S2 summarizes techniques from the literature for characterizing total ash and inorganic composition characterization in biomass. Dry-ashing and wet-ashing are two major approaches used in conventional ash characterization. Dry-ashing directly heats biomass in a muffle furnace at the temperatures ranging from 500 °C to 600 °C. During heating, most minerals are converted to oxides, sulfates, phosphates, chlorides, and silicates. Some elements, including Fe, Selenium (Se), Lead (Pb), and Mercury (Hg), may partially volatilize during this process. Wet-ashing uses acids or oxidizing agents and is often preferred over dry-ashing because it solubilizes minerals without causing volatilization [58]. However, conventional methods that remove extractives require preconditioning, indicating that reliable results can only be obtained from samples within a specific range of particle sizes. Additionally, these methods require a relatively large amount of material (>300 mg) [59].

2.3.2. Non-destructive tools for ash determination

Infrared spectroscopy (IRS) is a valuable non-destructive analytical tool that detects the fundamental molecular vibrations excited by infrared light. Infrared spectroscopy yields qualitative and quantitative data with minimal or no sample preparation, at high throughput [60–62], and with multiple constituents analyzed simultaneously [63]. In addition to measuring inorganic composition, IRS also provides other structural information about the major polysaccharides in the biomass [63]. However, IRS cannot probe for trace elements, non-structural components, or compounds with concentrations below 1 g/L [64,65]. Fourier transform infrared spectroscopy (FTIR) is a powerful tool using infrared light. Sample preparation is crucial for FTIR, as it analyzes individual components extracted from the plant cell wall [66,67]. In addition to inorganics determination, Allison et al. demonstrated that FTIR can predict N content and alkali index in energy grasses converted from lignocellulosic feedstock [18]. The application of infrared light in NIR spectroscopy helps identify the biomass structure. NIR spectroscopy and pyrolysis-molecular beam mass spectrometer (py-MBMS), coupled with multivariate analytical techniques, can provide rapid and accurate predictive tools for biomass composition, including ash content [68–71]. However, these methods require sophisticated and costly equipment, as well as established analytical model using calibration samples. Additionally, NIR spectroscopy has limitations in predicting ash content because it only measures organically bound inorganics [64,72,73].

Powder X-ray diffraction (PXRD) has been frequently utilized to identify crystalline cellulose peaks and estimate an overall crystallinity index (CI). It also serves as a valuable tool to identify inorganic species such as quartz, carbonate, phosphate, sulfate, and metal oxides [56,74,75]. Quantification of minerals by PXRD is possible with careful experimental design, sample preparation (milling and mounting), data processing, and selection of standards. Moreover, additional techniques such as FTIR and elemental analysis are often required for mineral identification and selection of quantification standards [76].

Most analytical techniques used to quantify inorganic elements associated with biomass samples suffer low sensitivity and cannot be used for trace elements. Inductively coupled plasma mass spectrometry (ICP-MS) can detect down to the parts-per-trillion within liquid samples and can thus be used to measure trace elements. It also enables online determination of K and Na during single particle biomass combustion [77]. Furthermore, the accuracy and sensitivity of the measurements can be improved as the proper sample preparations and dilutions [78].

Neutron activation is another non-destructive technique that allows for the direct determination of trace element concentrations in biomass [79]. However, the major drawback is that the detection limits and analytical uncertainties are generally higher compared to standard ash characterization methods [16].

Laser-induced breakdown spectroscopy (LIBS) is an emerging non-destructive and rapid method that has shown comparable analytical results to established techniques for determining inorganic species across diverse lignocellulosic feedstocks [80]. Unlike wet-ashing techniques, LIBS requires little sample preparation and no reagent consumption, and it provides rapid data acquisition [73]. However, the major limitation of LIBS is the requirement of high inorganic compound concentration in biomass for detection [81].

Combining energy-dispersive X-ray fluorescence (ED-XRF) and NIR spectroscopy was introduced to enhance distinguishing between intrinsic ash and ash originating from contamination [55]. This technique creates new opportunities for developing instrumentation to monitor variable ash content in lignocellulosic feedstocks for biorefineries efficiently [55]. ED-XRF can also provide semiquantitative elemental analyses of individual biomass samples for their thermal conversion processes [8]. Additionally, scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM-EDS) can facilitate the analysis of the extrinsic inorganic particle composition. Coupling SEM-EDS with XRD can further identify specific mineral species present in the extrinsic inorganic particles [52,82].

Table 1 lists the suggested uses of advanced non-destructive technologies for characterizing inorganic species in lignocellulosic feedstocks. High-throughput analytical techniques, such as NIR coupled with Py-MBMS and wet chemistry, have been proven as effective characterization approaches to detailed chemical information of biomass samples with minimal sample preparation [73]. Wet analytical chemistry, IRS, ICP-OES, and LIBS, can provide rapid results with high chemical resolution. Unlike wet chemistry and XRF, LIBS and IRS require minimal or no sample preparations. Conclusively, most of these techniques can offer comprehensive features, enabling the best biomass characterization and screening based on the targeted biomass thermochemical conversion strategies.

3. Impacts of inorganic species on biomass preprocessing

Biomass preprocessing—including transportation, feeding, and particle size reduction—is essential before pyrolysis and accounts for approximately 4 % of total operational costs [83]. Inorganic species play a critical role in biomass preprocessing, posing challenges to process optimization. One major concern is equipment wear during biomass preprocessing, which directly impacts the feasibility of commercializing biorefineries and has drawn considerable attention [84]. In general, inorganic compounds in biomass feedstocks are the primary contributors to equipment wear, as they are harder and more abrasive than organic tissues [4]. Beyond equipment degradation, inorganic species affect multiple aspects of biomass processing. They complicate biomass transportation and feeding systems, leading to logistical challenges and reduced efficiency. Additionally, their presence increases energy consumption during preprocessing, ultimately hindering the overall efficiency of biomass pyrolysis and other conversion processes.

3.1. Equipment wear caused by inorganic species and ash

Machine wear is a critical issue in biomass preprocessing, driven by both extrinsic and intrinsic inorganic compounds. Extrinsic inorganic species, primarily from soil contamination, have the greatest impact due

Table 1
Suggested advanced biomass characterization techniques for elemental composition based on specific features of the technique.

Features	Characterization techniques
High throughput	NIR, py-MBMS, wet chemistry
High resolution	NIR, py-MBMS, LIBS
Low sample preparation	LIBS > wet chemistry; infrared spectroscopy > XRF
Non-destructive	Infrared spectroscopy, neutron activation, LIBS, Raman, FTIR, XRF
Broad application	LIBS > ICP-OES
Trace elements	ICP-MS, neutron activation, XRF
Low detection limit	ICP-MS, ICP-OES
Structural information	Infrared spectroscopy, Raman
Field portable	Raman, XRF, NIR, FTIR
Distinguish intrinsic/ extrinsic	ED-XRF coupled with NIR

to their abundance on biomass surfaces. Large particles (>200 μm) with sharp edges exert high contact forces, accelerating equipment wear and erosion. This abrasion substantially increases equipment maintenance costs and shortens machine lifespan, necessitating wear-resistant materials.

In woody biomass, bark typically contains higher extrinsic inorganic content than other fractions (Figs. 1 and 2). However, Lee et al. [4] reported that pine bark, despite having less ash than the needles, caused more wear on machines. Among inorganic species, Si and Al containing compounds can be harder than the steel used in comminution equipment, which causes excessive wear during biomass preprocessing [84, 85].

Intrinsic inorganic matter also contributes to abrasion and erosion of equipment. During particle size reduction, inorganic compounds redistribute according to particle size fractions. The 0.6–2 mm fraction has been reported to contain the highest inorganic and ash content, increasing the risk of machine wear without proper particle screening [86]. However, the intrinsic inorganic species vary across different feedstocks.

3.2. Impact on feeding, plugging, and energy consumption

The presence of inorganic species in biomass significantly affects various aspects of biomass processing, including feeding, plugging, and energy consumption. Soil contamination exacerbates plugging during conveyance, while high inorganic content increases the energy required for grinding and particle size reduction [87]. Variability in ash content further increases energy consumption, generates substantial fines, and reduces processing throughput, posing significant techno-economic challenges [15].

Fine extrinsic ash particle coat the biomass surface, significantly reducing biomass processing efficiency [88]. These fine particles also retain moisture, creating strong adhesive forces that act as binders, leading to plugging, arching, rat-holing, and discontinuous flow. Additionally, inorganics can absorb heat during preprocessing, further impacting energy efficiency [89]. Biomass with higher ash content requires approximately 30–42 % more energy for preprocessing [90,91].

Inorganic species include ash, Si, Al, Fe, and various minerals such as silicates, sulfates, and phosphates. These compounds contribute to various levels of impact on biomass preprocessing, including machine erosion, corrosion, and increased energy consumption (Table 2). Minerals like silicates, sulfates, and phosphates are particularly hard compounds that are most likely to cause damage to biomass preprocessing equipment. Corrosion-induced material loss during biomass processing can cause secondary erosion to machines, further reducing processing efficiency and machine lifespan [92]. The increased costs associated with the issue of inorganic species have become a critical challenge for lignocellulosic refineries. The mechanochemical process was considered as a cost-effective approach for removing inorganics from wood chips before biomass conversion [93]. Total process costs were projected to range from \$6 to \$9/dry ton of biomass for removing 62 %, 39 %, and 88 % of total ash, alkaline earth metals, phosphorus (AAEMS + P), and Si, respectively. These findings highlight the significance of minimizing dirt contamination, controlling inorganic content, and implementing effective preprocessing methods to reduce wear, energy consumption, and processing issues in biomass utilization.

Table 2
Impact of inorganic compounds on preprocessing.

Inorganic content	Formula, element, or note	Impact	References
Ash	SiO_2, Fe_2O_3, Al_2O_3, MgO, K_2O, CaO, etc.	• Higher power consumption during grinding, resulting in reductions of processing rate. • Higher mechanical wear rate, equipment erosion or corrosion.	[94,95]
Intrinsic and extrinsic inorganic compounds		• Combined contribution to machine wear. • Extrinsic minerals made a significant impact even though they only accounted for a small fraction of the total ash.	[4]
Silica and silicates	SiO_2, $NaAlSi_3O_8$ $CaAl_2Si_2O_8$ $CaMgSi_2O_6$ or $CaMg(SiO_3)_2$ $NaAlSi_3O_8$-$CaAl_2Si_2O_8$ $KAlSi_3O_8$ α-$CaSiO_3$ or α-$Ca_3Si_3O_9$	• Harder than the steel used in milling and can cause excessive wear. • Cause the most friction against the screw extruder and the briquette die.	[4,84]
Aluminum and alumina	Al, Al_2O_3	• Harder than the steel used in milling and can cause excessive wear.	[4,84,85]
Iron	FeO, Fe_2O_3, Fe_3O_4	• Preprocessing equipment wear.	[85]
Sulfates and phosphates	$Ca(PO_4)_3(Cl,F,OH,CO_3)$ or $Ca_5(PO_4)_3(Cl,F,OH,CO_3)$ $Ca(PO_4)_3(OH)$, $Ca_5(PO_4)_3(OH)$ or $Ca_{10}(PO_4)_6(OH)_2$	• Harder than mild steel and more likely to damage biomass preprocessing equipment.	[84]

4. Impacts of inorganic species on biomass pyrolysis

In addition to impacting biomass preprocessing, inorganic species present in biomass have become one of the decisive factors determining biomass behavior during thermal degradation, directly influencing conversion efficiency and quality in biomass pyrolysis [96–98]. Depending on their composition, these inorganics introduce new functionalities, exhibiting either catalytic or negative effects on biomass pyrolysis reactivity according to their specification. As a result, they can alter feedstock conversion and upgrading processes, ultimately modifying the chemical composition of bio-oils produced from biomass pyrolysis [99].

Unlike extrinsic inorganic species that mainly affect biomass preprocessing, intrinsic inorganics can facilitate the depolymerization of cellulosic polymers by promoting carbon-carbon bond cleavage and lowering reaction temperatures. In some cases, inorganics were found to act on primary pyrolysis reactions altering the distribution of organic compositions [100]. Sekiguchi and Shafizadeh reported that intrinsic inorganic compounds increase char and gas yields at the expense of liquid pyrolysis oil [101]. After the biomass thermochemical conversion, inorganic compounds are generally concentrated in the solid biochar residue [102]. Generally, lower ash content can increase oil yields by 1–5 % for 1 % of ash removed from native biomass [103].

Fast pyrolysis is widely used in biomass thermal conversion to produce various forms of bioenergy. Inorganic species, major components of ash present in lignocellulosic biomass, can significantly impact the biomass thermal conversion, influencing both biofuel quality and yields. Typically, woody biomass is preferred for fast pyrolysis due to its lower ash content compared to herbaceous feedstock (between 1.1 and 4.8 % wt.), leading to higher bio-oil yields and minimizing issues such as catalytic poisoning or equipment fouling during pyrolysis and gasification [103,104]. However, the presence of different inorganic species can significantly affect biomass thermochemical conversion, either inhibiting or catalyzing the process. Table 3 summarizes the specific impacts of the major inorganic species found in woody biomass.

Table 3
Impact of the inorganic composition of woody materials on pyrolysis.

	Impact on Pyrolysis	Reference
Alkali metals		
Potassium	•Strong catalytic effect on lignin thermal degradation. •Negatively affects catalytic fast pyrolysis. •Affects the biomass carbohydrate degradation pathway. •Increases biochar yield. •Lowers liquid fuel yield. •Causes fragmentation of the monomers that make up the polymer chains, rather than significant depolymerization in the absence of monomers.	[15,16,100,103,105–113]
Sodium	• Catalyzes lignin thermal degradation. •Trace amounts of sodium salts can significantly increase charcoal production. •Causes secondary cracking of vapors.	[15,16,100,104,106,107,109,110,112,115,116]
Alkaline earth metals		
Calcium	•Catalytic effect on lignin thermal degradation. •Lowers tar yield. •Increases biochar yield.	[16,100,104,110,115]
Magnesium	•Catalytic effect on lignin thermal degradation.	[16,100,104,110,115]
Others		
Iron	•Catalyzes cellulose degradation to both levoglucosan and levoglucosenone.	[16,104,110,115]
Sulfur	•Causes machine corrosion. • Sulfur tolerant catalysts or sulfur adsorbents (zinc oxide) are required. • Causes catalyst poisoning. • Formation of hydrogen sulfide in the pyrolysis vapor inhibits the deoxygenation reactions on the catalyst. • A scrubbing system is required to reduce harmful hydrogen Sulfide emissions.	[16,106,109,114–118]
Chlorine	• Causes machine corrosion. • Increases volatilization and formation of chlorine. • High concentration of chlorine increases fine particulate emissions. • Toxic to catalysts. • Iron or copper chlorides increase levoglucosan yields. • Likely bond with trace elements, such as Pb and Zn to form lead (II) chloride and zinc chloride in the volatile phase at high temperatures. • 20 ppm of hydrogen chloride cause degradation in solid-oxide fuel-cell performance. • Slag is formed after Cl reacts with alkali metals such as K and Na.	[16,106,109,114,119–121]
Bromine	• Hydrogen bromide formation increases volatility of biofuels.	[109,122]
Phosphorous	• Increases biochar yields. • Strong catalytic effect on lignin thermal degradation. • Phosphate deposition by catalysis from alkali metals causes catalyst poisoning. • Without alkali, formation of phosphoric acid accelerates the deposition of coke.	[110,117]
Nitrogen	• Nitrogen catalyst targets tar destruction and ammonia reduction. • Ammonia is the most-significant species from fuel nitrogen conversion (concentrations from 500 to 30000 ppm). • Scrubbing or catalytic elimination can remove the precursor to nitric oxide emission in downstream units. • Generates an unpleasant smell.	[109,114,116,117]
Silica	• Secondary catalyst for the cleavage of macromolecular polymers. • Facilitates char and biogas production. • Contribute to fouling on reactor surfaces. • Lowers overall thermal efficiency.	[104,105,110,125]
Heavy metals		
Cu, Zn, Cd, Pb, Hg, Mn, V, As	• Formation of fly ash and aerosols that lead to particulate emissions. • Affects the utilization and disposal of char/ash. • Cause machine abrasion, corrosion, and erosion. • Requires gas cleaning device. • An additional bio-oil cleaning step is needed. • Hinders energy and mass transport.	[16,104,110]

4.1. Catalytic effect on biomass pyrolysis

4.1.1. Alkaline metals (IA metals)

Ash formed from alkaline metals during biomass thermal conversion plays a critical role in biomass pyrolysis. Alkaline metals (primarly Na and K) catalyze biomass thermal conversion, exhibiting the greatest effect on improving biogas and biochar yields and quality from biomass pyrolysis by reducing oxygen content and modifying its chemical composition [123]. During the biomass pyrolysis, the presence of alkaline cations promotes the cleavage of bonds between monomers, leading to depolymerization, dehydration, decarboxylation, demethylation, decarbonylation, etc., of the polymer chains [106].

Among alkaline metals, Na and K also have strong catalytic effects on lignin thermal degradation by interacting with acidic functional groups and enhancing ether bond cleavage, forming simple phenolic compounds and decreasing the yield of bio-oil while increasing char and gas yield from 400 to 900°C (Fig. 3) [124,125]. Wang et al. reported the decrease in aromatics and olefin production with a higher concentration of alkali metals [126]. Even trace amounts of these metals influence pyrolysis rate, product yield, and degradation temperature and alter decomposition mechanisms [127]. Moreover, Li et al. reported that alkali metals effectively reduce the initial pyrolysis temperature by lowering activation energy for municipal waste and biomass in briquettes [127]. The similar results were also reported in the thermal decomposition kinetic investigation of Douglas fir, and the initial pyrolysis temperature was reduced to at around 350°C [128].

Alkaline metals further affect carbohydrate degradation pathway, illustrated in Fig. 4, and catalyze secondary fragmentation of volatiles, resulting in increased biogas and biochar production [100,103]. Consequently, the products of biomass pyrolysis can be tailored by augmenting the biomass with metal ions. Notably, potassium was reported stronger catalytic effects than sodium due to its larger metal strength [114,129]. Among potassium and sodium salts, carbonate salts demonstrate the highest catalytic efficiency, followed by sulfate and chloride salts [130]. Pushkaraj et al. demonstrated that small amounts of sodium chloride contribute to the formation of levoglucosan, the major pyrolysis product of cellulose for further product upgrading [100]. K-catalyzed pyrolysis increases biochar yield while reducing the average first-order activation energy required for the process [131]. Higher K concentrations in feedstocks also increase the production of acidic products from biomass thermal conversions, as well as lowering the bio-oil quality [131]. Conversely, reduced K can minimize further

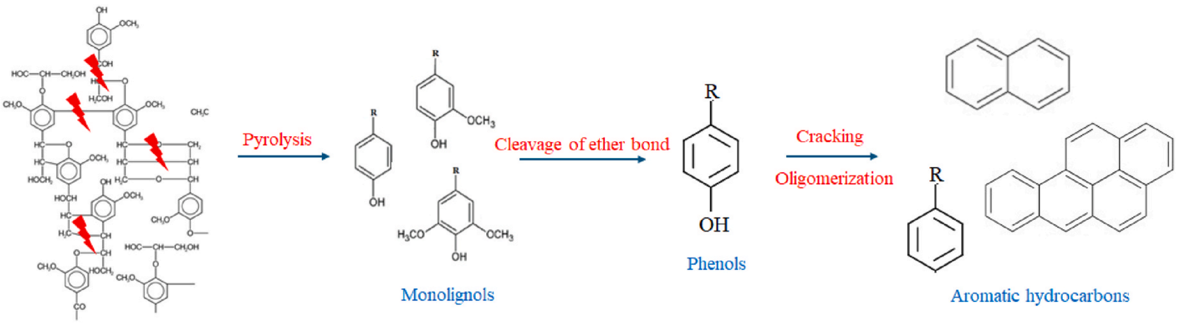


Fig. 3. Pyrolysis of lignin for bioenergy conversions.

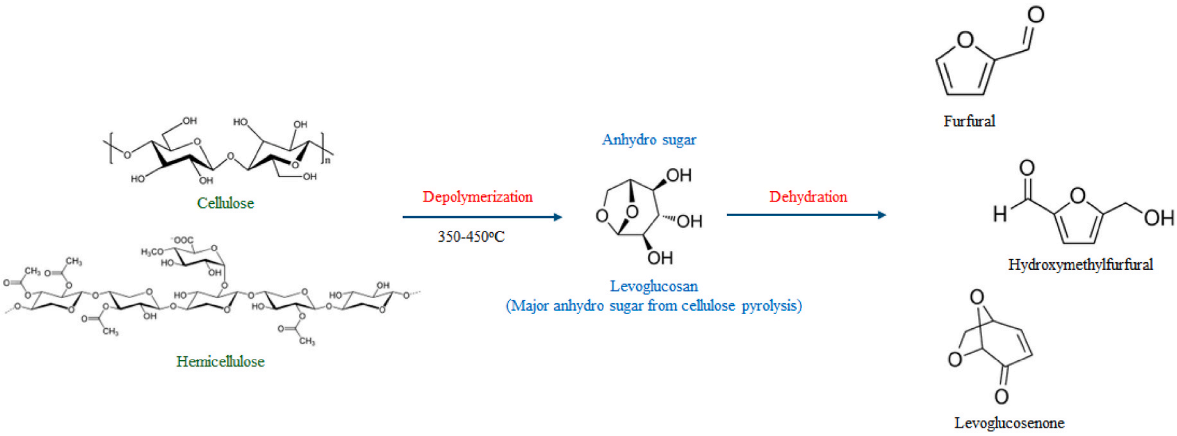


Fig. 4. Structural carbohydrate depolymerization during pyrolysis for biofuel conversions.

catalytic cracking or aging reactions [131].

4.1.2. *Alkaline earth metals (IIA metals)*

Alkaline earth metals (AEMs), including Ca and Mg, also exhibit catalytic effects on biomass pyrolysis. These metals have been reported to break weak bonds in favor of stronger ones, leading to an increase in condensed aromatic rings [132]. However, their impacts on lignin thermal degradation are less compared to alkaline metals such as K and Na [16,100,103,109,113].

In addition to lignin degradation, AEMs have a strong affinity for oxygenated groups, promoting deoxygenation via dehydration and depolymerization of cellulose and hemicellulose to form anhydro sugars. Wang et al. reported that AEMs greatly catalyzed cellulose and hemicellulose pyrolysis below 330 °C, reducing the initial and peak pyrolysis temperatures [133]. Kleen and Gellerstedt showed that Ca^+ enhances the yield of anhydrous sugars from pulp pyrolysis [120]. Shimada et al. observed that alkaline earth metals also promote the formation of low-molecular-weight products, such as levoglucosan, from cellulose depolymerization during pyrolysis. Notably, these also exhibit catalytic effects on reducing the bulk cellulose-decomposition temperature and activation energy by up to 20 % [119].

Furthermore, Ca and Mg also enhance the secondary dehydration of anhydro sugars, i.e. levoglucosan, anhydro glucofuranose, cellobiosan, etc. to form furans, light oxygenates, and char (Fig. 4) [134–137]. However, compared to alkaline metal ions (K^+, Na^+), Ca^{2+} and Mg^{2+} are a weaker cracking catalyst in biomass pyrolysis with lower levoglucosan yields [138].

4.1.3. *Other inorganics*

Other inorganic elements present in biomass can also catalyze biomass pyrolysis, particularly Fe, Cl, and P. Fe and Cl primarily facilitate cellulose degradation to increase levoglucosan yields [16,100,109,119]. Phosphorus salts were reported as a catalyst for lignin degradation, resulting in increased char yields [139]. While Si is generally considered inert in pyrolysis reactions. Bulushev and Ross found that silicon dioxide can act as a secondary catalyst, cracking macromolecular polymers and increasing char and gas formation [140].

Alkaline metals, alkaline earth metals (AEMs), and other inorganic species are primarily present in biomass ash. Ash content is the critical indicator for predicting the yield and quality of bioenergy from lignocellulosic biomass pyrolysis. During pyrolysis, even trace amounts of ash in biomass—as low as 0.1 wt%—can exert a catalytic effect, resulting in reduced bio-oil yield [103,130]. Moreover, biomass with high ash content significantly alters the product distribution, leading to increased yields of char and non-condensable gases [131]. This shift in the product profile can substantially impact overall process efficiency and the production of liquid and gaseous biofuels. In general, higher inorganic content results in greater bio-charge and bio-char yields, accompanied by a decline in liquid biofuel output.

Li et al. reported that a 7.4 % ash content increase in biomass would marginally increase biochar yield (by 0.03 kg per kg of biomass) but significantly reduce biofuel yields by 53.4 % (46.6 gallons per ton) [141]. This reduction might be mitigated by using biomass with a higher O/C ratio ranging from 0.88 to 1.12.

4.2. *Negative effects on biomass pyrolysis*

Although inorganic species can facilitate the degradation of structural carbohydrates and lignin during pyrolysis, their accumulation can also negatively influence biomass pyrolysis. Additionally, machine abrasion and further corrosion and erosion caused by these species pose major challenges for biomass pyrolysis.

4.2.1. Inhibitory effect on biomass pyrolysis

The inhibitory effect of inorganic species on biomass pyrolysis can be the catalyst poisoning derived from deposition of volatile compounds, particularly phosphate from reaction between alkaline metals (K, Na) and P [115]. Moreover, hydrogen sulfide present in pyrolysis vapor was reported to inhibit the deoxygenation reactions on the catalyst [142]. However, these effects have primarily been observed in pyrolysis processes with external catalyst additions. The inhibitory effects of the intrinsic inorganic species on biomass pyrolysis efficiency remain unexplored and require further investigations. A deeper understanding of these effects is essential for optimizing pyrolysis processes and developing strategies to mitigate negative impacts on product yields and quality.

4.2.2. Impacts on pyrolysis operation and its costs

However, alkaline metals and AEMs can lead to operational issues, such as reduced heat transfer, gas-flow disturbance, gasifier damage, corrosion, and erosion due to fouling, slagging, and agglomeration [143]. Another critical challenge posed by inorganic species is machine wear caused by heavy metals. Elements such as Cu, Zn, Cd, Pb, Hg, Mn, V, and As contribute to abrasion, corrosion, and erosion, further exacerbating equipment degradation. Additionally, these heavy metals promote biochar formation and intensify slagging, fouling, and agglomeration [103,109].

Beyond machine damage caused by inorganic species, they significantly impact heat transfer efficiency, gas flow stability, and overall gasifier performance [103,109]. These issues will substantially lower the economic feasibility of biomass pyrolysis for bioenergy production. Moreover, variations in ash content among different lignocellulosic feedstocks lead to differences in the energy content of bioenergy products after pyrolysis [144]. While the composition of pyrolysis bioenergy cannot be predicted from biomass ash content alone, limited data suggest that bioenergy yield from biomass pyrolysis is affected by both feedstock ash content and process conditions [145].

Blending different feedstocks is seen as promising strategy to mitigate ash content and secure economic sustainability for biomass pyrolysis. Lan et al. investigated the economic viability of fast pyrolysis biorefineries using blended feedstocks. Their analysis suggests that a more competitive minimum fuel selling price (MFSP) can be achieved using low-ash pellets mixed with pine residues. Notably, a 2000 dry metric tons per day biorefinery operating at 90 % capacity with blended pellets (75 % switchgrass, 25 % pine residues, 2 % ash) could produce fuel with an MFSP between $4.49 and $4.71 per gallon of gasoline equivalent. In contrast, a traditional centralized biorefinery operating at 80 % capacity achieves an MFSP between $4.72 and $5.28 [146]. However, several uncertainties still require further investigation. Future research is needed to understand how inorganic species impact the cost-effectiveness of bioenergy from biomass thermal conversions. This includes optimizing reaction design, mitigating inorganic elements, and improving related post-treatment and processing methods to address the economic and technical challenges posed by inorganic species in biomass conversion processes.

4.3. Impacts of waste and emissions on the environment

Additional gas and bio-oil cleaning steps are required for product recovery to ensure the better quality of bioenergy produced from biomass pyrolysis [147]. However, these cleaning processes not only increase operational costs but also introduce environmental concerns through waste streams. Emissions of sulfides and nitrogen oxides derived from S and N compounds are the major concerns, which contribute to air pollution and exacerbate impacts on ecosystem [109], such as acidification and eutrophication impacts. Additionally, the fine particulate emissions ($PM_{2.5}$) resulting from high Cl concentration present another are another critical air pollution concern [148].

Heavy metal content in biomass presents further environmental and ecological risks [16]. Emissions of heavy metal-based fly ash aerosols can lead to contamination of air, water, and soil, posing serious pollution hazards [149]. These challenges underscore the need for effective environmental management and safe handling practices for pyrolysis products and by-products [150].

5. Mitigation strategies

Implementing effective strategies to mitigate inorganic species in woody biomass is essential for optimizing its use as a renewable energy source. High inorganic content in biomass can lead to operational challenges in different conversion pathways, such as increased wear and tear on equipment, higher maintenance costs, and reduced efficiency due to slagging, fouling, and corrosion in boilers and gasifiers. Furthermore, ash disposal presents an environmental and economic challenge, as it requires safe handling and storage to prevent pollution. Mechanical, chemical, physiochemical, and advanced biomass pretreatments, summarized in Table 4, have been introduced to biomass processing prior to biomass pyrolysis to mitigate the inorganic content and make it a more efficient and cleaner fuel (Fig. 5). This not only enhances the overall performance and lifespan of bioenergy conversion systems but also contributes to the sustainability of biomass as a renewable energy source by reducing its environmental impact and lowering operational costs.

5.1. Mitigation before and through harvest

Mitigation strategies to reduce inorganic content in biomass are essential for minimizing nutrient loss from harvest sites, as well as reducing equipment maintenance and ash disposal costs in biorefineries. However, limited research has explored effective approaches to lower inorganic species content in woody biomass. One promising method is to optimize harvest window to reduce inorganic components that negatively affect fast-pyrolysis product distribution [131]. For example, analysis indicated that loblolly pine samples harvested in January (summer) had the highest ash content, demonstrating a statistically significant difference compared to samples collected in other seasons [152]. Similarly, white gum sampled in fall and late winter had the highest ash content [153].

Adjusting harvest practices is another approach to lower ash content, especially to reduce levels of water-soluble elements like Cl, S, K, Na, P, and N [150]. By incorporating a trommel screen in forest residue grinding process, the ash content can be notably decreased from 4.0 % to 1.4 % for roundwood residues, and from 11.9 % to 6 % for smaller chipping residues [154]. However, trommel screening could lead to approximate 50 % wt [156] biomass loss. Huber et al., reported that screens could decrease the ash content and improve biomass quality for pyrolysis, particularly in small wood fractions [154,155].

Moreover, the harvest practice can be adjusted particularly for reducing the extrinsic inorganic species present in biomass, including controlled felling to minimize soil contact, adopting clean collection practices to avoid ground debris, and debarking to remove mineral-rich bark. Additionally, utilizing machinery designed to limit soil and debris pickup [211]. Selecting low-risk harvest sites and performing post-harvest cleaning, such as air blowing or washing, can further help to lower ash content. By adopting these strategies, it becomes feasible to reduce biomass ash content, which in turn decreases equipment maintenance requirements and ash disposal costs. Moreover, these approaches enhance the quality and efficiency of biomass utilization processes, leading to more sustainable and cost-effective energy conversions.

5.2. Mechanical pretreatment

Particle size reduction and sieving are the principles used in biomass mechanical pretreatment. Particle size reduction not only increases

Table 4
Strategies for mitigating inorganic compounds prior to biomass pyrolysis.

	Pros	Cons	References
Biomass processing during harvest: Optimize harvest window	- Minimize nutrient loss from harvest site - Reduces equipment maintenance and ash disposal cost in biorefineries		[150, 151–157]
Adjust harvest practices	- Remove Cl, S, K, Na, P, N. - Decrease ash content from 4.0 % to 1.4 % for roundwood residues, and from 11.9 % to 6 % for smaller chipping residues by trommel screen. - Improve biomass quality for pyrolysis. - Decrease equipment maintenance requirements and ash disposal costs.	- Inconsistent ash reduction - Higher costs for Screened Round wood - Reduced Grinder Utilization. - ∼50 % mass loss	
Mechanical pretreatment: Air classification	- Effectively removes exogenous ash components like silica, alumina, and iron - Reduces propensity for slagging and fouling in biopower applications - Blending ash-reduced fractions can reduce overall feedstock costs compared to single feedstock - Removes over 40 % of ash content by concentrating it into <7 % of total biomass. - Reduces overall ash content from 1.69 % to 1.07 % for forest thinnings and 1.09 %–0.68 % for logging residues	- Removing physiological ash components within plant cells is more difficult - Significant biomass loss (up to 34 %) when removing high-ash light fractions. - Ineffective at removing biologically derived inorganics like calcium, potassium, magnesium, and phosphorus	[158–160]
Vibratory Sieving	- Reduces ash content by segregating minerals into different fractions - Simple and straightforward pretreatment process - Low-cost ash mitigation process	- May not completely remove all inorganic contaminants - Potential for feedstock losses during the sieving process	[161]
Biomass washing: Water washing and leaching	- Field rain washing is inexpensive and returns nutrients to the soil - Simple and straightforward treatment process - Removes extrinsic ash components - Decrease ash-related issues - Surfactant can be added to the water to increase the ash removal efficiency - Reduces alkaline metal and AMEs content in biomass - Decreases harmful emissions of pollutants like chlorine, sulfur, and nitrogen - Minimal impact on physicochemical properties of biomass	- Field rain washing is limited by weather unpredictability and can lead to organic degradation and material loss - May not be sufficient to remove intrinsic ash species - Using surfactant-mediated inorganics removal approaches is non-selective, potentially extracting lipids and proteins during the process - Can disrupt the chemical structure and morphology of biomass - High water consumption required for the washing process - Wastewater generated needs to be treated, adding operational costs - Potential loss of water-soluble nutrients in the biomass - Consumes energy for heating, pumping, and drying the treated biomass	[93,109, 158, 162–165]
Hot-water extraction	- Reduces Na, K, and Ca concentrations, increasing the higher heating value (HHV) of the bio-oil - Improved bio-oil composition - Effectively remove a large fraction (50–100 %) of the alkaline metals and AEMs, especially the monovalent K. - Limited increases in the yields of sugars in the bio-oil.	- Cannot completely remove the divalent AAEM species like Mg and Ca from the biomass. - Downstream drying of treated biomass requires high energy input, increasing costs in the bio-oil. - Disposal of leachate containing removed inorganics can be challenging	[108,123, 166–168]
Chemical pretreatment: Acid Pretreatment	- Effectively removes inorganic minerals, alkali and alkaline earth metals from biomass. - Causes structural changes, increasing biomass energy density and accessibility - Increased aromatic hydrocarbon yield by removing alkali and alkaline earth metals. - Prevents catalytic effects of inherent or added alkaline metals and AEMs - Improves bio-oil yield and quality - Improves pyrolysis selectivity towards bio-oil, reduces char and gas yields.	- Acidic leachate requires proper disposal, increasing operational costs - Additional pretreatment increases complexity and cost of the overall process. - Corrosive nature requires expensive non-corrosive reactor materials. - Limited understanding of the removal mechanism of inorganic species - Can lead to 4–15 % biomass loss - Difficulty removes calcium in some feedstocks	[123, 169–183]

(continued on next page)

Table 4 (*continued*)

	• Alters bio-oil composition, increasing oxygen content and anhydro sugars. • Reduces ash-related issues like slagging, fouling, and corrosion • Concentrates inorganics in the solid char for separate utilization. • Passivates AAEMs by converting them into normal and acid salts. • Increases sugar yields, especially levoglucosan (up to 55 % of cellulose)	• Can cause agglomeration and clogging issues during continuous pyrolysis • Decreases H_2 production at both low and high temperatures	
Alkaline pretreatment	• Effective at removing inorganic contaminants from biomass • Solubilizes lignin and enhances cellulose digestibility • Enhances depolymerization of hemicellulose and cellulose, benefiting thermochemical conversion • Increase alkali and alkaline earth metal (AAEM) content in biomass • Enhances gas and char yields at low pyrolysis temperature (823 K) • Promotes H_2 production at both low and high temperatures	• Forms salts to inhibit the bio-oil production • Generates significant amounts of wastewater requiring further treatment. • Reduces bio-oil yield • Inhibits syngas energy output at high pyrolysis temperature • Removes lignin resulting in reduction in energy density	[172,181,184–189]
Thermal pretreatment: Hydrothermal pretreatment	• Retain structural carbohydrates and lignin • Majorly disrupt biomass structure and retain structural carbohydrates and lignin • Effectively removes heavy metals like Ni, Ag, Pb, Zn, Cu, As, Cd, Cr • Effectively reduces ash content • Reduces slagging and fouling tendencies of biomass ash • Decreases alkali index and chlorine content to avoid corrosion • Wide adaptability to different feedstocks	• Silica content in ash is relatively stable and not significantly reduced • Some inorganics like Pb and As are not effectively removed • Potential loss in overall yield of useable pyrolysis oil products • Can lead to catalyst deactivation	[190–198]
Torrefaction	• Removes ash with depolymerization of structural carbohydrates • Significantly reduces ash-forming elements like Na, Mg, K, and Ca. • Improves fuel quality by reducing oxygen content and increasing energy density • Improves bio-oil quality • Increases phenol content • Increases anhydro sugar yields • Increases syngas yields • Mitigates slagging, corrosion, and agglomeration during biomass thermal conversion	• Lowers bio-oil yields • Complex and energy-intensive pre-treatment process • Potential loss of desirable organic components • Challenges in scaling up the technology	[99,165,199–204]
Advanced process: Sequential pretreatment processing	• Combining different pretreatment technologies • Optimizes inorganic removal • Provides flexibility to process various resources of feedstocks	• Additional preprocessing steps incur capital and operating costs • Additional optimization is needed	[205–207]
Microwave-assisted pretreatment	• Applied to acid, alkaline, and hydrothermal pretreatment • Increases energy efficiency • Increases organic accessibility	• Causes microexplosions and uneven treatment of biomass • Potential incomplete transformation of pyrolysis intermediates, leading to lower organic product yields	[178,184,208–210]

biomass surface to increase pyrolysis efficiency but also mitigates ash and inorganic content through subsequential screening [83]. Additionally, extrinsic inorganic species can be removed from the screening process.

Air classification and mechanical sieving are widely used in mechanical pretreatment. Air classification, known as air aspiration, separates biomass into different fractions based on density profiles [160]. Thompson et al. reported that the lighter fraction contains higher ash forming elements, such as Si and alkaline metals [159]. Air aspiration has demonstrated strong performance in removing approximately 40 % of extrinsic inorganic compounds and ash content from biomass before pyrolysis. Additionally, it offers significant flexibility in adapting to various feedstocks while maintaining economic viability, with an estimated cost of $2.23 per ton of biomass [158,159]. However, air classification is limited in removing intrinsic inorganic species and results in approximately 34 % biomass loss [159].

Similar to air aspiration, vibratory screening separate biomass into different fractions. This process utilizes a vibrating screen or series of screens to classify particles, with smaller particles passing through the screen openings while larger particles are retained [161]. Pradhan examined the ash removal efficiency of pinewood and sweet gum and reported that vibratory screening can remove 17–27 % ash content

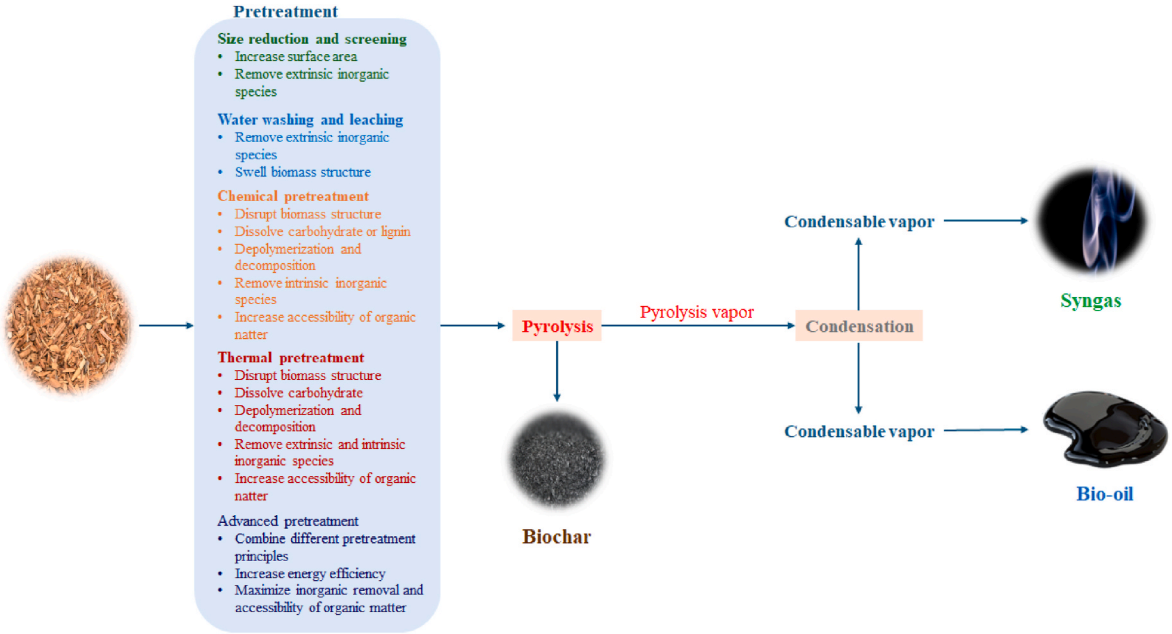


Fig. 5. Biomass-to-bioenergy pipeline via pretreatment and pyrolysis.

[212]. However, this process primarily removes extrinsic inorganic species and also contribute to mass loss.

5.3. *Biomass washing*

Biomass washing is an economical approach to remove inorganic species from biomass and decrease ash-related issues, such as slagging, fouling, and corrosion [158,164]. Additionally, water washing and leaching also mitigate the harmful emissions from Cl, S, and N [164].

Both rainfall in the field and laboratory-scale soaking, leaching, and dewatering have demonstrated efficacy in reducing inorganic content [93]. Meester et al. reported that about 80–90 % of K and Cl can be removed by multiple extractions with fresh water [213]. Compared to water washing in the lab, field rain washing is inexpensive and returns nutrients to the soil but is limited by weather unpredictability and can lead to organic degradation and material loss by 15 % [93]. Water washing can be performed using spraying, flushing, or soaking with cold or hot water in a temperature-controlled environment. Among these methods, soaking proves more effective than spraying or flushing, with hot water achieving better results than cold [162]. Water washing at near boiling temperatures can remove ash species introduced in soil contamination [109]. Surfactants can be added to the water to increase the ash removal efficiency. However, using surfactant-mediated inorganics removal approaches is non-selective, potentially extracting lipids and proteins during the process [163].

While water washing primarily removes extrinsic inorganic matter, it can also cause minor disruptions to biomass morphology and, resulting in loss of water-soluble nutrients, such as carbohydrates and proteins [213]. Additionally, the process incurs increased operational costs due to high water consumption, wastewater treatment requirements, and the need for additional energy to dry the biomass after washing [164]. The cost of water leaching to remove alkaline metals and AEMs was estimated at $8 per ton of dry biomass [213].

Hot-water extraction (HWE) has demonstrated success in removing inorganic metal species from the biomass that would increase bio-oil yields [123]. Mohammed et al. reported that hot-water treatment significantly reduced Na, K, and Ca concentrations, increasing the higher heating value (HHV) of the bio-oil [166]. Tarves et al. demonstrated that treating shrub willow by HWE reduced the concentration of inorganic minerals, such as Ca, K, Mg, P, and S, improving bio-oil's heating value by 5 % with reducing hemicellulose content [167]. Similar results were reported by Mourant et al. indicating that Na, K, and Mg were significantly removed using HWE at near to boiling temperature [108]. Overall, HWE is an efficient and cost-effective technique, removing inorganic minerals or alkali metals and enhancing bio-oil quality [123]. However, hot water extraction has lower removal rate of AEMs due to its lower mobility and weaker divalent binding strength with water compared to alkaline metals [214]. Similar to water washing and leaching, the resulting waste stream can also pose potential environmental burdens.

5.4. *Chemical pretreatment*

Acid and alkaline pretreatments have merged as effective methods for modifying the composition of lignocellulosic feedstocks, particularly in removing inorganic species and altering the structure of biomass components [215]. Various acids and bases have been used to mitigate ash-forming elements before pyrolysis. These pretreatments play a crucial role in enhancing pyrolysis efficiency and improving bio-oil quality.

5.4.1. *Acid pretreatment*

Acid pretreatment primarily solubilizes hemicellulose and alters biomass structure, facilitating removal of soluble metals and insoluble minerals not physiologically bound to the plant tissue. Sulfuric acid, hydrochloric acid, phosphoric acid, acetic acids, nitric acid, and formic acids have been commonly used in biomass pretreatment for bioenergy conversions [169,170,172,174,175]. The cleavage of C-O bond among structural carbohydrates and lignin as well as the breaking of ether bond within lignin by acids facilitates hemicellulose solubilization and inorganics extraction. It results in increasing heating value of biomass with condensation of aromatic compounds in biomass [123,179]. Reducing alkaline inorganic species mitigates their catalytic effect on thermal decomposition of biomass, resulting in higher yields and better quality of bio-oil [103]. The efficiency of inorganic leaching increases with higher concentration of the acidic solvent [123].

Meesters et al. demonstrated that acid pretreatment can be designed to extract targeted AEMs with over 80 % efficiency [213]. Acid leaching with acetic acid removed approximately 60–86 % of alkaline metals and AEMs [177]. Hong et al. investigated the efficiency of inorganic removal from white pine and rice husk using 7 % HCl, H_3PO_4, and H_2SO_4, reporting 55–88 % removal rates for alkaline species. Among these acids, HCl and H_3PO_4 exhibited the highest efficiency for AEMs removal, achieving rates of 78–88 % [170]. Stefanidis et al. found that nitric acid treatment at room temperature for 5 h removed over 90 % of inorganics [169]. Pascoli dilute H_2SO_4 treatment achieved up to 81 % ash removal from pine chips [176]. Liu et al. examined the effect of HCl concentration on microalgae pretreatment, finding that increasing HCl concentration to 4M removed approximately 79 % of Al. However, the overall ash removal rate remained relatively low at 38 % [151].

In addition to inorganic removal, acid pretreatment passivates alkaline metals and AEMs, enhancing biomass susceptibility to thermal decomposition and significantly increasing bio-oil yields while improving quality by reducing acidity and viscosity [178]. Usino et al. found that mahogany bio-oil yields increased by over 70 % after acid pretreatment with 1 % H_2SO_4. Results of 46 % bio-oil yield increase were reported by Pittman et al. from corn stalks acid pretreatment using 2 % H_2SO_4 [183]. Pienihäkkinen et al. reported that nitric acid pretreatment increased bio-oil yields by over 40 % from the pyrolysis of eucalyptus residues and wheat straw [168]. Similar results were observed in the pyrolysis of pretreated agricultural residues. Wang et al. reported that 10–46 % increase in bio-oil yield from sweetgum, switchgrass, and corn stover after 1 % H_2SO_4 pretreatment [173]. Zhou et al. summarized that about 20 % reduction in bio-oil acidity was achieved from HCl pretreatment [173]. Tan et al. concluded that acid pretreatment not only enhances bio-oil yields but also increases sugar yields for bioenergy conversion [216].

Although acid pretreatment is seen as an efficient approach for inorganic mitigation, the post-treatment of acid leachate presents challenge due to its potential content of hazardous metals. Additionally, the corrosive nature of acid increases operational and maintenance costs, necessitating expensive non-corrosive construction materials for reactors [131]. However, its high sugar content resulting from cellulose and hemicellulose degradations offers opportunities for producing value-added coproducts [123]. Notably, Horhammer et al. demonstrated that a subsequent neutral wash following dilute acid pretreatment further enhanced mineral removal from biomass, eliminating all K and Mn, and 94 % of Mg [95]. To date, limited studies have examined the economic and environmental costs and benefits of acid pretreatment and its overall impact on biomass pyrolysis. This lack of research leaves gaps in our understanding of how to design a sustainable inorganic mitigation for bioenergy production from biomass pyrolysis.

5.4.2. *Alkaline pretreatment*

Alkaline pretreatment is another process to enhance biomass depolymerization by disrupting glycosidic and ester bonds between lignin and hemicellulose, leading to primarily solubilizing lignin and keeping most parts of cellulose intact [166,181]. NaOH, KOH, $Ca(OH)_2$, and NH_4OH have been commonly used in alkaline pretreatments [123,184,185]. With the solubilization of lignin, biomass structure is disrupted, resulting in cellulose swelling, increasing accessibility to biomass decomposing, and facilitating ash removal [181,217].

Unlike acid pretreatment, alkaline pretreatment could decrease bio-oil yield. Wang et al. found about 10 % reduction in bio-oil yield from loblolly pine pyrolysis after pretreatment with 0.5 % NaOH [172]. Similar results were reported by Mohammed et al. indicating 9 % decrease in bio-oil yield from the pyrolysis of 5 % NaOH pretreated Napier grass [166]. However, Ellison et al. observed a marginal increase in bio-oil from the pyrolysis of NaOH pretreated eucalyptus residue [186]. Hernández-Beltrán et al. concluded that the effects of alkaline pretreatment on the composition of bio-oil, biochar, and biogas vary depending on the biomass feedstock [188]. To optimize bio-oil yield while preserving cellulose, Raveendran et al. investigated the combination of acid and alkaline pretreatments. The results demonstrated increase in bio-oil yields of 16–23 %, 39 % and over 75 % were explored from the pyrolysis of 10 % HCl-5 %NaOH pretreated corn residues, rice husk, and subabul wood, respectively [218].

In addition to its impact on bio-oil yield, alkaline pretreatment has been shown to reduce biochar yield. Palniandy et al. reported a 10 % decrease in biochar yield from the pyrolysis of rice husk. Notably, alkaline pretreatment increases the carbon content of biomass and alters the functional groups of biochar, enhancing its catalytic activity and potential applications [187].

Though alkaline effectively solubilizes lignin, alter biomass structure, remain cellulose, and enhance inorganics mitigation, the potential formation of salts, such as NaCl, Na_2SO_4, K_2SO_4, and K_2CO_3, from alkali agents could reduce the bio-oil yields [130]. Similar to acid pretreatment, leachate from the alkaline poses required waste treatment to mitigate pollution risks, leading to an increase in operational and capital costs [189].

5.5. *Thermal pretreatment*

Thermal pretreatment is seen as a physiochemical process to deconstruct biomass to enhance its accessibility for further conversions. Hydrothermal and torrefactions have been widely used in biomass pretreatment for bioenergy production through different conversion routes. For its applications in inorganic mitigation, the hydrothermal process utilizes pressure and high temperature to solubilize inorganic species; torrefaction directly uses heat to break down the biomass structure, removing the intrinsic inorganic matter, which then improves pyrolysis efficiency and bioenergy quality [193].

5.5.1. *Hydrothermal pretreatment*

Hydrothermal pretreatment effectively removes loose dirt and structural ash, eliminating up to 90 % of Ca, S, P, Mg, and K, and less than 50 % of Fe and Mn from biomass, resulting in a homogenized, carbon-rich, energy-dense solid fuel, biochar, through pyrolysis [190]. Processing temperature and duration are critical factors influencing biomass deconstruction and ash mitigation. A wide range of temperatures (120–230 °C) and processing times (30 min to 2 h) have been explored. Reza et al. reported that hydrothermal carbonization at 130 °C for 2 h, assisted by citric acid, effectively reduced heavy metals, lowering structural ash content by 77–93 % without adversely affecting the cellulose, hemicellulose, or lignin composition [191]. Lu et al. observed an increase in ash content reduction from 13 % to 21 % as the processing temperature rose from 120 °C to 180 °C. Vallejo et al. highlighted that intrinsic inorganic species migrated from biomass to the liquid phase during hydrothermal processing, reducing the impact of minerals on biomass pyrolysis and product properties. Additionally, they demonstrated the broad adaptability of hydrothermal pretreatment to various feedstock blends, achieving up to a 70 % increase in energy yield and a 46 % improvement in higher heating value (HHV) from different biomass sources [195].

However, solely hydrothermal process primarily reduces biomass crystallinity and partially solubilizes hemicellulose [219]. The effectiveness of hydrothermal treatment in enhancing ash reduction, improving product HHV, and increasing pyrolysis efficiency is significantly higher when assisted by additional chemicals [191,195]. The use of chemical additives introduces additional leachate handling requirements, similar to acid and alkaline treatments, and may lead to catalyst deactivation during pyrolysis [197,198]. Other challenges associated with hydrothermal pretreatment include biomass loss and the limited reduction of Si, Pb, and As [195–197]. Furthermore, limited studies have investigated the effects of hydrothermal processing on bioenergy product yield and distribution, resulting in knowledge gaps regarding the optimization of hydrothermal pretreatment for regulating bioenergy production from pyrolysis.

5.5.2. *Torrefaction*

Unlike hydrothermal process, torrefaction is performed at 200–300 ^{0}C with low heating rate under inert atmospheric conditions to deconstruct biomass [123]. The extent of biomass deconstruction is primarily determined by torrefaction temperature. In light torrefaction (200–235 °C), hemicellulose degradation occurs through the breaking of glycosidic bonds and the dehydration of hydroxyl groups. Mild torrefaction (235–275 °C) primarily breaks glycosidic and hydrogen bonds, leading to the depolymerization of free hydroxyl groups in hemicellulose and cellulose, resulting in further structural breakdown. Severe torrefaction (275–300 °C) causes near-complete degradation of hemicellulose and significant depolymerization of cellulose [123,200]. With depolymerization and degradation of cellulose and hemicellulose, the intrinsic ash-forming species can be removed. Sutapa and Hidyatullah reported that about 50 % total ash were removed from Calliandra wood by torrefaction [220].

In addition to ash reduction, torrefaction also increases energy density and improves fuel quality by reducing oxygen content [199,201,202]. Tumuluru et al. reported that the heating value of pine chips doubled after torrefaction at 270 °C, while the total lignin content increased to 68 % after the torrefaction at 300 °C [204]. Ren et al. found that torrefaction increased syngas quality, resulting in a 32 % increase in H_2 content and a 12 % increase in CO content [99]. Moreover, torrefaction increased carbon conversion to 93 % to syngas production [204]. For bio-oil production, torrefaction increases anhydro sugar production due to its primary effects on depolymerization of structural carbohydrates [204]. However, it leads to lower bio-oil yields. Tran et al. observed about 4.5 % reduction in bio-oil yield from pyrolysis of torrefied spruce wood [203]. Moreover, torrefaction effectively alleviates the slagging, corrosion, agglomeration issues by reducing ash-forming inorganic species from biomass [199].

However, torrefaction pretreatment can result in up to 30 % biomass loss, leading to a significant reduction in organic compounds available for biofuel conversion [204]. Additionally, high energy requirements and fine particulate emissions are major concerns associated with torrefaction [165]. Its complex and energy-intensive nature poses significant challenges to large-scale commercialization.

5.6. Advanced preprocessing

5.6.1. Sequential pretreatments

The sequential pretreatments, integrating different biomass deconstruction processes to optimize ash mitigation and increase bioenergy yield and quality, have been introduced. Wigley et al. combined dilute acid pretreatment with torrefaction in series for pinus radiate wood, resulting 25 % increase in bio-oil yield [205]. Chen et al. applied sequential wash and torrefaction pretreatment to cotton stalk, which increased HHV of biofuel by 37 % but its bio-oil yield decreased from 42 % to 24 % [206]. Zhang et al. reported marginal increase in bio-oil yield and its HHV by about 2 % from the sequential light torrefaction and acid leaching for rice husk [207].

While sequential pretreatment offers flexibility in processing diverse feedstocks, it also presents challenges. Additional processing steps increase operational costs, and further optimization is required to integrate different processing technologies into a single pipeline. Future research is necessary to inform the development of more efficient sequential pretreatment systems for bioenergy conversion. This includes optimizing process parameters, exploring cost-effective integration strategies, and assessing the overall economic and environmental impacts of these integrated systems.

5.6.2. Microwave-assisted pretreatment

Microwave technology has been applied to facilitate acid, alkaline, and hydrothermal pretreatment, effectively reducing biomass crystallinity, improving inorganic species mitigation, increasing organic matter accessibility, and enhancing energy efficiency [178,208,209]. Álvarez-Chávez et al. found that microwave-assisted hydrothermal pretreatment achieved a 58 % ash reduction and increased organic species accessibility by 18.5 % for spruce wood [208]. Chen et al. reported that microwave-assisted formic acid pretreatment increased aromatic product yields from lignin decomposition to 30 % [178]. Moreover, microwave-assisted alkaline pretreatment has been shown to enhance biomass porosity and surface area while selectively solubilizing lignin and hemicellulose, improving the removal of intrinsic inorganic species [184].

However, excessive microwave exposure can cause microexplosions within the biomass structure, leading to uneven heating, microstructural collapse, and increased recalcitrance [210]. The rapid heating associated with microwave-assisted pretreatment may also result in incomplete transformation of depolymerized intermediates, reducing organic product yields and biofuel production [178]. Therefore, further optimization of microwave assisted pretreatment is necessary to maximize the efficiency of biomass pyrolysis.

6. Current status and future perspectives

Inorganic species, particularly alkaline metals and alkaline earth metals, impact biomass pyrolysis efficiency and bioenergy quality (Table 3). Mechanical pretreatments have been employed to remove extrinsic inorganic matter, mitigating operational challenges and facilitating subsequent processing [158,159]. Various chemical, physicochemical, and thermal pretreatments have also been introduced to address inorganic-related issues, particularly by removing intrinsic inorganic species. However, these approaches alter biomass decomposition pathways, affecting syngas, bio-oil, and biochar yields, as well as their physicochemical properties (Table 4). Compared to conventional biomass pretreatment using single processing technology, hybrid mechanical-physicochemical methods have been explored to enhance biomass deconstruction while preserving organic matter [178,205–207]. Among these, advanced hydrothermal pretreatment applications have shown great potential for integration with mechanical and chemical processes, improving ash mitigation through biomass deconstruction [193,195,196]. Additionally, these methods offer flexibility for processing different biomass types based on their chemical composition, providing a promising solution to challenges related to feedstock variability, uncertainty, and supply [221]. Further research is needed to optimize consolidated biomass pretreatment strategies for accommodating biomass variability and feedstock blends.

Gaining a deeper understanding of inorganic migration mechanisms is a crucial area for future research, particularly in the development of consolidated pretreatments that integrate multiple technologies within the biomass-to-bioenergy pipeline. Leveraging the principles of mechanical, chemical, and hydrothermal pretreatments to explore their synergistic effects could mark a significant milestone in bioenergy conversion. Additionally, strategically balancing gas, liquid, and solid bioenergy yields through intelligent pretreatment design presents a critical opportunity to enhance the efficiency and viability of biomass pyrolysis. Furthermore, integrating advanced modeling techniques, such as computational fluid dynamics and machine learning algorithms, could greatly improve the ability to predict and control inorganic migration during pyrolysis, leading to more precise process optimization and improved bioenergy yields [222].

Economic and environmental viability is another major concern for consolidated pretreatments. Multiple operations increase operational costs and capital investments. Several studies have conducted the techno-economic analysis of biomass pyrolysis for bioenergy production from wood using different pretreatments prior to pyrolysis, and the cost biofuel ranges from $2.58-$4.13/gal [112,223,224]. However, limited studies have performed the cost analysis and prediction focusing on biomass pretreatment for further pyrolysis. Though higher operational costs are expected for the consolidated pretreatments due to additional processing units, the higher quality of biofuel and coproduct recycling can increase the total revenues and improve overall economic feasibility.

For carbon footprint of biofuel production, Lan et al. reported that a range from 40.8 to 41.2 g CO_2e/MJ biofuel from pine residues via fast pyrolysis was observed [225]. Notably, the carbon footprint can be reduced by over 50 % when biochar is reused as a soil amendment, resulting in 19.0–19.7 g CO2e/MJ. This study has provided great insight into the carbon dynamics of biofuel production from woody biomass. However, limited studies have highlighted the environmental impacts derived from the leachate from biomass pretreatment. It will help the development of consolidated biomass pretreatment and pyrolysis to optimize bioenergy production and sustainably manage its coproducts and waste streams.

Future research should also focus on reducing the high feedstock costs, which are a major economic driver, and exploring innovative pretreatment methods to enhance the overall efficiency and environmental performance of biofuel production systems.

7. Conclusion

This review synthesizes insights from peer-reviewed publications to examine the quantity, distribution, and roles of inorganic species within anatomical fractions of woody biomass. It addresses three key research questions: (1) the total ash content and specific inorganic species introduced at various stages of biomass supply logistics, (2) the specific impacts of extrinsic and intrinsic inorganic species on biomass pyrolysis, and (3) the critical unit operations and most efficient methods for managing ash, specifically whether these opportunities are best implemented early in the supply chain or by integrating preprocessing with biorefinery operations. However, few studies have focused on these areas, and significant data gaps hinder our ability to assess the economic and environmental trade-offs between increased processing costs to achieve lower ash levels and the higher biofuel production costs associated with elevated ash content.

A comprehensive understanding of inorganic profiles in biomass is fundamental to effective ash mitigation and process optimization in thermochemical conversions. Biomass pretreatment strategies present opportunities for advanced process design, improving bioenergy conversion efficiency while enhancing co-product utilization and waste management. Optimizing these factors is essential for increasing the economic and environmental viability of biomass-to-bioenergy pathways, particularly through pyrolysis. Consolidated biomass pretreatment offers a promising approach, not only for effectively mitigating inorganic species and enhancing product yields and quality but also for facilitating coproduct recovery and waste minimization.

Future studies should focus on leveraging the synergistic effect integrating different pretreatments to improve the overall efficiency of biomass pyrolysis. Additionally, advanced modeling techniques, such as dynamics simulation and machine learning, should be employed to better predict and control inorganic migration during thermal conversion. Further research should also explore the economic and environmental trade-offs associated with various ash mitigation strategies, assessing their feasibility across different biomass supply chains. By addressing these gaps, future studies can contribute to the development of more efficient, cost-effective, and sustainable biomass-to-bioenergy processes, ultimately enhancing the viability of biofuels as a renewable energy source.

CRediT authorship contribution statement

Ling Ding: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Ming-Hsun Cheng:** Writing – review & editing, Conceptualization. **Yingqian Lin:** Writing – review & editing, Writing – original draft, Visualization, Conceptualization. **Kuan-Ting Lin:** Writing – original draft, Data curation, Conceptualization. **Kenneth L. Sale:** Writing – review & editing. **Ning Sun:** Writing – review & editing. **Bryon S. Donohoe:** Writing – review & editing, Writing – original draft. **Allison E. Ray:** Writing – review & editing, Funding acquisition. **Chenlin Li:** Writing – review & editing, Supervision, Project administration, Investigation, Conceptualization.

Declaration of competing interest

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.energy.2025.135697.

Data availability

Data will be made available on request.

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