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ALKYL-AROMATIC-MODIFIED PHENOLIC BINDERS FOR HIGH-PERFORMANCE WOOD WASTE BIOFUEL BRIQUETTES

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ABSTRACT

The utilization of wood waste for the production of solid biofuels is an important route toward sustainable energy and waste valorization. In this work, a modified phenol–formaldehyde resin (MPFR), synthesized by polycondensation of phenol and formaldehyde with an alkyl-aromatic fraction of catalytic cracking gas oil in the presence of an ionic liquid catalyst, was investigated as an efficient binder for biomass-based fuel briquettes. Beech and pine sawdust were used as fillers, and briquettes were produced by hot pressing at pressures of 50–150 MPa and temperatures of 50–100 °C with binder contents of 0.5–1.0 wt.%. The influence of binder content and biomass particle size on density, compressive strength, ignition behavior, combustion duration, calorific value, and ash content was systematically studied. The obtained briquettes exhibited densities of 1.4–2.0 g/cm^3, compressive strength of 2.2–3.5 MPa, calorific values of 9300–11000 kcal/kg, and combustion times of 16.3–26 min. Increasing the MPFR content and optimizing sawdust particle size significantly improved mechanical integrity and combustion performance compared with binder-free briquettes. Moisture uptake tests over three months showed less than 1% mass increase, indicating good storage stability. The results demonstrate that modified phenolic resin is an effective binder for producing high-performance biomass fuel briquettes from wood waste and offers a promising route for waste-to-energy applications.

Keywords: fuel briquettes; wood residues; modified phenol-formaldehyde resin; aromatic hydrocarbons; binder properties; combustion performance.

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1. Introduction

The growing demand for energy in all sectors of human activity has traditionally been met by petroleum and petroleum-derived fuels. However, the continuous exploitation of these non-renewable resources has led to their gradual depletion and increased environmental pressure, stimulating strong interest in renewable and sustainable energy alternatives. In this context, biomass derived from wood waste, agricultural residues, forestry by-products, and organic fractions of industrial and household waste has attracted considerable attention as a promising renewable energy source [1].

Among biomass-based fuels, fuel briquettes represent a practical and economically attractive option owing to their controlled shape and size, adjustable density, ease of transportation, and high energy efficiency [2]. Briquettes are typically produced by compressing dried biomass under high pressure, using materials such as sawdust, wood shavings, straw, sunflower husks, rice straw, buckwheat shells, peat, and coal fines. In principle, almost any plant-derived material can serve as feedstock; nevertheless, wood and wood-processing wastes remain particularly valuable because of their renewability, low ash content, and relatively high calorific value.

Fuel briquettes also offer important operational advantages, including compactness, convenient storage, and stable combustion behavior. They are widely applied in domestic stoves, fireplaces, boilers, and industrial heating systems. Owing to their favorable environmental and energy characteristics, wood-based briquettes are often described as a fuel that is “hotter than firewood and cleaner than coal”. Compared with conventional solid fuels, briquettes emit lower levels of carbon oxides, exhibit safer handling properties, reduce heat exchanger and chimney corrosion, and minimize soot and ash formation. In addition, their production enables the valorization of waste materials, providing both economic and environmental benefits [3, 4].

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Conventionally, wood briquettes can be produced without external binders, where softened lignin acts as a natural adhesive during pressing. Such briquettes typically contain 8–10% moisture and less than 1.5% residual ash, making them an environmentally friendly fuel for residential, municipal, and industrial heating.

The depletion of traditional energy resources, their increasing cost, and the need to mitigate environmental pollution have intensified interest in molded biofuels such as briquettes as alternatives to conventional wood, peat, and petroleum-based fuels. The simplicity of briquette production technology, combined with the utilization of waste biomass and reduced emission of harmful gases during combustion, makes this approach particularly attractive for sustainable energy development.

Phenol–formaldehyde (PF) resins are among the earliest synthetic polymers and remain widely produced on an industrial scale owing to their broad raw-material base, simple synthesis technology, and high chemical reactivity associated with methylol and hydroxyl functional groups. These features allow versatile modification, enabling the targeted tuning of performance properties. As a result, PF resins are used as binders and functional components in coatings, rubber and lubricant additives, concrete plasticizers, surfactants, laminated materials, adhesives, abrasive products, wood composites, casting resins, and briquettes [5].

Modification of PF resins using aromatic hydrocarbons is an effective strategy for improving compatibility with lignocellulosic matrices and enhancing both mechanical and thermal behavior.

From a materials science perspective, the incorporation of alkyl-aromatic fragments into the phenolic resin matrix is expected to enhance hydrophobicity, promote π–π interactions with lignin-rich biomass components, and increase the overall aromatic carbon content of the binder phase. These factors may contribute to improved mechanical integrity, controlled thermal degradation, and enhanced calorific value of the resulting briquettes. The incorporation of alkyl-aromatic fragments can increase hydrophobicity, interfacial bonding, and energy content of the binder, which is particularly beneficial for fuel briquette applications. Nevertheless, conventional synthesis routes for modified PF resins often rely on acidic or basic catalysts that require relatively harsh conditions and may lead to less controlled polycondensation.

In recent years, ionic liquids have attracted growing attention as environmentally benign catalytic and reaction media owing to their negligible vapor pressure, high thermal stability, tunable solvating ability, and strong interaction with biomass components [6-11]. The development of high-precision methods opens up new research opportunities, expanding the scope for studying functional materials and identifying their applications [12-15].

Ionic liquids have been successfully applied in lignocellulosic dissolution, cellulose processing, lignin valorization, and polymer modification. In phenolic systems, ionic liquids can promote controlled polycondensation and facilitate the incorporation of aromatic modifiers under milder and more selective conditions compared with traditional catalysts. At the same time, their application in the preparation of modified PF binders specifically designed for biomass fuel briquetting remains largely unexplored.

Recent studies have demonstrated that the molecular structure and aromatic composition of petroleum-derived materials significantly influence their physicochemical properties, confirming that such compounds, which possess a wide range of applications, can serve as promising components for the development of functional polymer systems [16, 17].

Despite the extensive development of biomass briquetting technologies, several critical limitations remain unresolved. Most reported systems require elevated processing temperatures (≥ 160 °C), additional thermal curing stages, significant binder loading, or energy-intensive drying of feedstock. Furthermore, the application of modified phenol–formaldehyde resins synthesized under controlled catalytic conditions, particularly using ionic liquids, for low-temperature densification of lignocellulosic biomass has not been systematically investigated. Therefore, there remains a need for binder systems capable of ensuring high mechanical strength and combustion efficiency at reduced processing temperatures and minimal additive content.

Beech and pine sawdust were used as fillers, and the influence of binder content and particle size on density, compressive strength, ignition behavior, combustion duration, calorific value, ash content, and storage stability was systematically investigated. The proposed approach provides an energy-efficient densification strategy that eliminates the need for high-temperature post-treatment while maintaining low binder content and improved combustion performance. This study aims to bridge the gap between polymer modification chemistry and practical biomass fuel engineering.

2. Experimental / materials and methods

2.1. Materials

Pine (Pinus spp.) and spruce (Picea spp.) wood residues were used as the primary biomass feedstock. The residues, consisting of sawdust and wood chips generated during industrial wood processing, were air-dried to a moisture content of 6–8 wt.% and stored under controlled laboratory conditions prior to use. The dried biomass was milled and sieved to obtain particle size fractions of 0.4, 1.6, and 2.0 mm.

The modified phenol–formaldehyde resin (MPFR) was synthesized via condensation of phenol and formaldehyde in the presence of an aromatic hydrocarbons contained in the light gasoline fraction of catalytic cracking. The reaction was carried out using N-methylpyrrolidinium hydrophosphate as an ionic liquid catalyst. The molar ratio of phenol to formaldehyde was maintained at 1 : 0.8, while the mass ratio of phenol to aromatic hydrocarbons was 2:1. The synthesis proceeded at 96–98 °C under continuous stirring for 4 h, yielding the modified resin, which was used without further purification [13].

In the briquette formulations, MPFR was introduced at concentrations of 0.5, 0.7, and 1.0 wt.% relative to dry biomass mass. In selected experiments, 0.5 wt.% pine charcoal was additionally incorporated to evaluate its influence on structural and combustion characteristics.

2.2. Briquette preparation

The dried biomass fractions were thoroughly mixed with the calculated amount of MPFR to ensure homogeneous binder distribution. Briquetting was performed using a laboratory hydraulic hot press at pressures ranging from 50 to 150 MPa and temperatures between 50 and 100 °C. The pressing time was 3–5 minutes, sufficient to obtain mechanically stable compacts.

Cylindrical briquettes with a mass of 10 ±0.2 g were produced. After pressing, the samples were allowed to cool to room temperature under ambient laboratory conditions prior to characterization.

2.3. Characterization

The density of the briquettes was calculated from the measured mass and geometrical dimensions, assuming a cylindrical shape. Mass was determined using an analytical balance with an accuracy of ±0.01 g, and dimensions were measured using a digital caliper.

Compressive strength was determined using a universal testing machine under uniaxial loading. The load was applied along the axial direction of the cylindrical briquettes at a constant crosshead speed of 2 mm min^{-1} until structural failure occurred. Compressive strength (MPa) was calculated as the maximum applied force divided by the initial cross-sectional area of the sample.

Combustion performance was evaluated using briquettes of identical mass. Ignition time and total burning duration were recorded under controlled laboratory conditions. The higher heating value (HHV) was determined on a dry basis using an isoperibol bomb calorimeter according to standard calorimetric procedures. Residual ash content was measured gravimetrically after complete combustion of the samples in a muffle furnace at 550 °C until constant mass was reached.

Moisture stability was assessed by storing briquettes at 25 ±2 °C and relative humidity of 50–60 % for a period of three months. Moisture uptake (%) was calculated relative to the initial dry mass of the samples based on periodic mass measurements.

All experimental measurements were conducted in triplicate, and the reported values correspond to the mean of three independent determinations.

3. Results and discussion

The experimental results demonstrate that the incorporation of modified phenol–formaldehyde resin (MPFR) significantly alters both the mechanical and combustion characteristics of wood-based briquettes. In this work, a modified phenol–formaldehyde resin (MPFR), synthesized by condensation of phenol and formaldehyde with aromatic hydrocarbons contained in the gas oil fraction of catalytic cracking using an ionic liquid catalytic system, was evaluated as a binder for wood-based briquettes.

In the present system, the binder is not merely a physical adhesive but a thermosetting aromatic network incorporating petroleum-derived alkyl-aromatic fragments. Therefore, the observed improvements in mechanical and combustion properties are expected to arise from both enhanced interparticle bonding and increased aromatic carbon content introduced into the composite structure.

The incorporation of MPFR at 0.5–1.0 wt.% relative to the filler markedly influenced the structural and combustion characteristics of the briquettes. For beech sawdust-based samples containing 0.5, 0.7 and 1.0 wt.% MPFR, the density increased to 1.4–2.0 g cm^{-3}, while the compressive strength reached 2.2–3.5 MPa. This improvement is associated with enhanced interparticle adhesion and the formation of a more compact structure, resulting from resin bridging between biomass particles.

The improvement in compressive strength observed at moderate pressing temperatures indicates effective softening and redistribution of the modified resin within the biomass matrix. Under applied pressure, the resin likely forms continuous interparticle bridges that, upon thermal activation, evolve into a rigid crosslinked aromatic network. The enhanced structural integrity suggests improved stress transfer between lignocellulosic particles, which can be attributed to increased interfacial compatibility between the aromatic-rich binder and lignin domains.

Simultaneously, the calorific value increased to 9300–11000 kcal kg^{-1}, indicating that the aromatic-rich MPFR contributes additional energy and promotes more efficient heat release during combustion. It is noteworthy that the calorific value approaches or exceeds that of several conventional solid biofuels, despite the minimal binder content. This suggests that the aromatic modification of the resin contributes not only structurally but also energetically to the composite fuel system. The combustion duration was extended to 16.3–26 min, demonstrating that the modified binder stabilizes the burning process and suppresses rapid mass loss at the ignition stage.

Compared with briquettes obtained by simple pressing without additives, MPFR-containing samples exhibited longer ignition and burning times, with increases of 1.96–30.7 % (0.7–4.7 min, depending on binder loading). This behavior suggests that MPFR not only reinforces mechanical integrity but also alters the thermal degradation pathway of the lignocellulosic matrix, resulting in a more controlled release of volatile products.

The increase in calorific value can be partially attributed to the incorporation of petroleum-derived aromatic hydrocarbons into the binder structure. Aromatic-rich materials generally exhibit higher carbon content and thermal stability, which contribute to prolonged combustion and more stable heat release. Thus, the modified resin functions not only as a structural binder but also as an energy-enhancing component of the composite fuel.

A similar tendency was also observed in briquettes obtained by adding 0.5 wt.% beech charcoal (HC) and 0.5 wt.% MPFR to pine and beech sawdust fillers. Under identical preparation conditions, the combustion duration reached 20.2 min for pine-based briquettes and 26 min for beech-based ones (table 1). The longer burning time of beech sawdust briquettes can be attributed to their higher structural density and better compatibility with the aromatic-modified PF binder, which enhances interfacial bonding and heat transfer inside the briquette body.

A consistent trend is observed with increasing MPFR concentration: ignition time decreases while total combustion duration increases. This behavior indicates improved heat propagation within the densified structure and reduced premature volatile release, suggesting a more controlled combustion mechanism.

In addition to the combustion performance, the incorporation of MPFR noticeably improved the mechanical integrity of the briquettes compared with additive-free samples, which facilitates handling and transportation. As a

Table 1

Combustion characteristics of briquettes prepared from beech and pine sawdust using MPFR as a binder

№	Briquette composition	Time to ignition, min	Full ignition period, min	Total combustion time, min
1.	Beech sawdust (without additives)	1.40	6.50	15.30
2.	Beech + 0.5 wt.% MPFR	1.15	5.2	16.0
3.	Beech + 0.7 wt.% MPFR	1.20	4.9	17.46
4.	Beech + 1.0 wt.% MPFR	1. 9	4.50	20.0
5.	Beech + 0.5 wt.% MPFR + 0.5 wt.% HC	0.28	4.0	26.0
6.	Pine + 0.5 wt.% MPFR + 0.5 wt.% HC	0.5	3.6	20.2
7.	Pine + 0.7 wt.% MPFR	1.5	5.4	17.3
8.	Pine + 1.0 wt.% MPFR	2.2	5.1	18.4
9.	Pine + 0.5 wt.% MPFR	1.3	5.8	16.9

continuation of the study on modified PF resins as binders, the influence of filler particle size on the combustion behavior of the prepared briquettes was also investigated using beech and pine sawdust.

For this purpose, the wood sawdust used as filler was sieved using standard screens (TGL 7354) with aperture sizes of 0.4, 1.6, and 2.0 mm to obtain fractions of different particle sizes. These fractions were then employed for briquette preparation, and their effect on ignition and combustion characteristics was evaluated.

As shown in table 1, the addition of MPFR reduced the ignition delay and prolonged the total combustion time compared with binder-free briquettes. In particular, beech -based briquettes containing both MPFR and beech charcoal exhibited the longest burning duration (up to 26 min), indicating a synergistic effect between the aromatic-modified binder and carbonaceous additive. Pine-based briquettes showed a similar tendency, although with slightly shorter combustion times, which can be related to differences in biomass structure and density.

The observed enhancement in combustion behavior can be attributed to improved particle packing and interfacial bonding induced by MPFR, leading to a more homogeneous heat distribution and controlled thermal degradation during burning.

The effect of MPFR addition on the mechanical and combustion properties of briquettes was systematically investigated using beech and pine sawdust as fillers. Briquettes prepared with MPFR exhibited significantly higher compressive strength compared with binder-free samples, which facilitates handling and transportation.

The combustion characteristics of briquettes prepared from different particle sizes and binder concentrations are summarized in table 2. The results indicate that the performance of briquettes depends on the type of wood, particle size, and binder content. For instance, the combustion heat of beech sawdust briquettes increased from 9500 kcal kg^{-1} to 10000 kcal kg^{-1} and the burning time from 20.43 to 25 min as the particle size increased from 0.4 to 2.0 mm at a constant 0.7 wt.% binder concentration. Compressive strength exhibited a corresponding increase with density, indicating improved structural integrity.

The particle size of raw materials used in briquette production is a key factor affecting compressive strength, moisture content and calorific value. Smaller particle sizes promote stronger inter-particle bonding when adhesives are applied, leading to higher mechanical strength and energy output [18-20]. The results indicate that particle size exerts a dual influence on briquette performance. While finer particles improve packing density and mechanical strength, intermediate particle fractions (1.0-1.6 mm) provide an optimal balance between structural integrity and combustion stability. Excessively fine particles may restrict internal air diffusion, whereas overly coarse fractions reduce contact area for effective binder interaction.

Briquettes prepared from modified phenol–formaldehyde resin (MPFR) and wood sawdust showed that both particle size and binder content significantly influence their combustion properties [21]. Reducing the binder content from 0.7 wt.% to 0.5 wt.% in briquettes made from 1.6 mm beech sawdust resulted in a calorific value of approximately 11000 kcal kg^{-1}, a combustion time of 26.0 min, and low residual ash (~1.05 wt.%).

Pine sawdust briquettes exhibited consistently high performance across different particle sizes, with densities

Table 2

Combustion properties of briquettes prepared from different particle sizes of beech and pine sawdust using MPFR as binder

№	Filler (wood type)	Particle size, mm	Binder content, wt.%	Briquette parameters					
				Density, g cm^{-3}	Compressive strength, MPa	Ignition time, min	Total combustion time, min	Calorific value, kcal kg^{-1}	Ash content, wt.%
1	Beech	0.4	0.7	1.2	3.0	0.27	20.43	9500	1.07
2	Beech	1.0	1.0	2.0	3.5	0.28	26.0	10000	1.2
3	Pine	2.0	0.7	2.0	2.5	1.18	25.4	9700	2.0
4	Beech	2.0	0.7	1.8	3.0	1.2	25.0	10000	1.2
5	Pine	1.6	0.7	1.9	2.8	1.18	25.4	9800	1.8
6	Beech	0.8	0.5	1.4	2.2	1.4	16.3	9300	1.4
7	Beech	1.5	0.5	1.9	3.5	0.28	26.0	11000	1.05

of 1.9-2.0 g cm^{-3}, compressive strengths of 2.5-2.8 MPa, ignition times around 1.18 min, total combustion times near 25.4 min, low residual ash (1.8-2.0 wt.%), and calorific values of 9700-9800 kcal kg^{-1} (table 2). These briquettes burned with minimal smoke and bright flames, indicating environmentally favorable combustion.

The results demonstrate that the quality of briquettes depends on the wood species, sawdust particle size, and binder content. Optimization of these parameters allows the production of high-performance, environmentally friendly solid biofuels with improved mechanical strength and combustion efficiency.

It is known that one of the critical requirements for fuel briquettes is their stability during storage, specifically their resistance to moisture absorption and shape deformation. Accordingly, briquettes prepared from wood residues with 0.7 wt.% MPFR as a binder were tested for moisture uptake under ambient conditions over 3 months (table 3).

The results show that after 3 months of storage at room temperature, the mass of briquettes made from pine sawdust increased by only 0.99%, while those made from spruce increased by 0.97%. No visible changes in shape or appearance were observed in either case, indicating good dimensional stability and resistance to moisture uptake.

An important technological advantage of the proposed approach lies in the moderate pressing temperature (50-100 °C), which eliminates the need for high-temperature post-treatment or carbonization stages commonly reported in conventional briquetting technologies. This significantly reduces energy consumption and simplifies process integration. From an industrial perspective, the combination of low binder content (0.5–1.0 wt.%) and absence of additional curing stages enhances the economic feasibility of scaling the process.

Compared to conventional briquetting technologies requiring temperatures above 160 °C and additional thermal curing stages, the present system achieves comparable or superior mechanical and combustion properties at significantly lower processing temperatures (50-100 °C). The absence of high-temperature post-treatment and the ultra-low binder loading highlight the potential of this approach for energy-efficient industrial implementation.

Table 3

Moisture uptake of briquettes under ambient storage

№	Briquette composition	Initial mass (g)	Mass after 3 months (g)	Mass increase (%)
1	Pine (1.6 mm) + 0.7% MPFR	10.201	10.302	0.990
2	Beech (1.6 mm) + 0.7% MPFR	10.077	10.175	0.973

Conclusions

The present study demonstrates that a phenol–formaldehyde resin modified with alkyl-aromatic fragments and synthesized in the presence of an ionic liquid catalyst can effectively function as an ultra-low-content binder (0.5-1.0 wt.%) for the production of high-performance wood-based fuel briquettes. The incorporation of petroleum-derived aromatic fragments into the resin structure enhances interfacial compatibility with lignocellulosic biomass, resulting in improved compressive strength (up to 3.5 MPa), high density (up to 2.0 g cm^{-3}), extended combustion duration, and low residual ash content (~1 wt.%). Importantly, the briquetting approach is based on the method described in the patented technology [22], which enables processing at moderate temperatures (50-100 °C) without high-temperature post-treatment or carbonization stages, thereby significantly reducing energy consumption. The results confirm that the modified resin acts not only as a structural binder but also as an energy-enhancing component due to its aromatic-rich composition. The proposed system represents an integrated petrochemical–bioenergy strategy, enabling simultaneous valorization of wood-processing residues and refinery-derived aromatic fractions. Overall, the developed patented approach offers a promising route toward scalable, energy-efficient production of composite solid biofuels with improved mechanical stability and combustion performance.

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