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Physicochemical Enhancement of Low-Rank Coal via Low-Temperature Co-Torrefaction with Coconut Shell Charcoal

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ABSTRACT. This research investigates physicochemical and structural upgrading of Indonesian low-rank coal through low-temperature co-torrefaction with coconut shell charcoal. Coal-char mixtures containing 5, 25, and 45 wt% coconut shell charcoal were co-torrefied at 200 °C and characterized by thermogravimetric proximate analysis, bomb calorimetry, and X-ray diffraction (XRD). Linear regression was analyzed to evaluate relationships between coconut shell charcoal content and proximate properties. Increasing the charcoal fraction reduced the moisture content from 30.06% in raw lignite to 8.46% at 45 wt% charcoal and fixed carbon increased from 27.69% to 59.94%. Fixed carbon showed a strong positive relationship with charcoal content with R^2 = 0.9249, while volatile matter decreased with increasing charcoal addition with R^2 = 0.9161. The calorific value increased from 4823.46 to 6786.28 kcal/kg, demonstrating substantial energy upgrading. XRD patterns showed changes in the broad carbon-related diffraction features, indicating limited modification of the short-range structural ordering of the carbonaceous matrix after co-torrefaction. These research results show the potential of solid fuel for combining biomass charcoal as a renewable energy source with low-rank coal through low-temperature co-processing.

INTRODUCTION

Low-rank coal from Indonesia is a solid fuel that should be utilized more efficiently as an energy source. The high moisture content and presence of oxygen-containing functional groups contribute to its low calorific value. The oxygen-containing groups, including hydroxyl and carboxyl groups, also contribute to the hydrophilic nature of coal (Srivastava *et al*., 2025). In addition, oxygen-containing compounds are partly associated with volatile matter, which can be released when coal is heated. Moisture also increases energy consumption during combustion because water must be evaporated before the fuel can burn effectively. These characteristics contribute to the relatively low calorific value of low-rank coal. Since moisture and some oxygen-containing compounds can be removed through low-temperature treatment, thermochemical processing is an initial step toward increasing the carbon content and improving coal quality.

Torrefaction is part of the pyrolysis process held in low tempeature rang 200 – 300 ^{o}C under limited oxygen conditions, whereas, promotes dehydration, deoxygenation, and partial cabonization (Rizkiana *et al*., 2024) that release some part of moisture and volatiles. Due to the use of 2 feed stoct form coal and biomass. This process is termed thermochemical processing, exactly co-torrefaction. Biomass, as one of the feedstocks, can provide carbon enhancement and a renewable aspect for solid fuel. The incorporation of biomass-derived carbonaceous materials can modify the composition and thermal behavior of coal–biomass mixtures, potentially improving fixed carbon content and energy density. Recent studies have demonstrated the potential of coal–biomass thermochemical processing using various biomass precursors and operating conditions (Yadav *et al*., 2025).

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However, the physicochemical response of low-rank coal to co-torrefaction with pre-carbonized coconut shell charcoal under low-temperature conditions remains insufficiently understood. In particular, quantitative relationships between coconut shell charcoal fraction, proximate properties, energy content, and carbon structural ordering have received limited attention (Adeleke *et al*., 2024). Previous studies have predominantly focused on biomass torrefaction, coal-raw biomass mixtures, or fuel performance, whereas the role of pre-carbonized coconut shell charcoal as a carbon-rich component in low-temperature coal upgrading has been less systematically investigated. This knowledge gap limits understanding of whether increasing coconut shell charcoal content can systematically improve the physicochemical and structural characteristics of low-rank coal under mild processing conditions (Adeleke *et al*., 2024).

Coconut shell charcoal is particularly suitable for investigating this approach because of its high fixed carbon content, low ash content, and availability as an agricultural by-product, making it an attractive renewable carbon source for upgrading solid fuels (Sousa *et al*., 2024). Its incorporation into low-rank coal enables the combination of a carbon-rich, biomass-derived material with a moisture-rich fossil feedstock. Accordingly, this study hypothesizes that increasing the proportion of coconut shell charcoal during low-temperature co-torrefaction will decrease the content of moisture and volatile while fixed carbon increases and then calorific value also rose, accompanied by detectable changes in the short-range ordering of the carbonaceous structure.

Accordingly, this study is designed to (a) quantify the influence of coconut shell charcoal fractions of 5, 25, and 45 wt% on the moisture, volatile matter, ash, and fixed carbon contents of low-rank coal; (b) evaluate the associated enhancement in calorific value; and (c) examine changes in the carbonaceous structure using X-ray diffraction. Linear regression analysis is further employed to evaluate the relationships between coconut shell charcoal fraction and the resulting proximate properties. Through these analyses, the study evaluates low-temperature coal-char co-torrefaction as a potential pathway for upgrading low-rank coal while utilizing coconut shell-derived carbonaceous residues.

RESEARCH METHODS

Preparation of Coconut Shell Charcoal and Low-Rank Coal

Coconut shells from a local market in Padang were cleaned to remove adhering impurities and dried at room temperature for 24 h. The dried coconut shells were then converted into charcoal by partial combustion under atmospheric conditions using a conventional carbonization method. The shells were burned until red-hot glowing charcoal was obtained, after which the combustion was stopped. The charcoal was allowed to cool to room temperature while minimizing further oxidation. The resulting charcoal was ground and sieved to obtain a particle size of 60 – 70 mesh. Low-rank lignite coal was obtained from a coal supplier in the Jambi region. The coal was also dried at room temperature for 24 h and crushed and sieved to obtain a particle size of 60 – 70 mesh.

Co-Torrefaction Procedure

The pulverized low-rank coal and coconut shell charcoal were manually blended until a uniform mixture was obtained. Coconut shell charcoal was used as an additive at mass fractions of 5, 25, and 45 wt%, and the resulting mixtures were labeled C5, C25, and C45, respectively. Raw low-rank coal and coconut shell charcoal were labeled B100 and C100, respectively, and used as reference materials. Cassava starch (Manihot esculenta) was added as a binder at 10 wt% of the total coal–charcoal mixture to form briquettes (Nonsawang *et al*., 2024). The mixtures were manually molded into spherical briquettes with diameters of 2 – 3 cm. A 50 g portion of the briquettes was placed in a porcelain crucible and subjected to co-torrefaction in a laboratory oven at 200 °C for 1 h under oxygen-limited conditions. After heat treatment, the briquettes were cooled to room temperature before being removed for characterization.

Physicochemical and Structural Characterization

Proximate analysis was performed using LECO TGA701 thermogravimetric analyzer in accordance with ASTM D7582-15. Approximately 0.85 g of each sample was analyzed to quantify the moisture, volatile matter, and ash contents under the instrument's standard operating conditions. Fixed carbon content was determined by difference based on the results of proximate analysis. Each sample was analyzed in duplicate, and the reported values represent the average of the duplicate measurements. X-ray diffraction analysis was conducted using a PANalytical X'Pert Pro PW3040/60 diffractometer fitted with Cu Kα radiation with λ= 1.5406 Å. The powdered samples were scanned over a 2θ interval of 10 – 80º. The diffraction patterns were analyzed to compare the carbon-

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related structural characteristics of raw low-rank coal, coconut shell charcoal, and co-torrefied samples, with particular emphasis on the broad (002) and (100) reflections associated with turbostratic carbon. The interplanar spacings d_{002} and d_{100} were determined using Bragg's law (Li *et al*., 2024), while variations in peak position and peak broadening (FWHM) were used to comparatively evaluate changes in the short-range structural ordering of the carbonaceous matrix (Love-Baker *et al*., 2024), following co-torrefaction. The calorific value of each sample was measured using a LECO AC500 automatic bomb calorimeter in accordance with ASTM D5865/D5865M-19. Approximately 1 g of each sample was analyzed according to the standard operating procedure. All Measurements were performed in duplicate, and the reported values represent the average of the duplicate determinations. The calorific value was used to assess the enhancement of fuel energy content after blending with coconut shell charcoal and subsequent low-temperature co-torrefaction.

RESULTS AND DISCUSSION

Co-Torrefaction Behavior of Low-Rank Coal and Coconut Shell Charcoal

Figure 1 shows the mass flow of hybrid coal production, starting from raw material preparation until the production of co-torrefied briquettes. The process was conducted in batches, and the mass was measured at each stage to determine the mass balance. Low-rank coal was air-dried before grinding because its moisture content could affect the grinding process. Similarly, raw coconut shells were carbonized before grinding because their fibrous and elastic structure made them more difficult to grind than charcoal, which is more brittle. Low-rank coal and coconut shell charcoal were then pulverized to obtain a particle size of 60 – 70 mesh. The two materials were blended to obtain a uniform mixture. The prepared mixture was then combined with cassava starch as a binder and formed into briquettes to improve particle contact during thermal treatment. The mass flow diagram shows material losses during several stages, particularly during coconut shell carbonization and co-torrefaction. The initial briquette mass before co-torrefaction was 49.5 g, while the final mass decreased to 33.5 g after treatment, resulting in a mass yield of 67.7%. This indicates that approximately 32.3% of the initial mass was lost during thermal treatment. The mass loss was likely associated with moisture evaporation and the release of volatile compounds during heating.

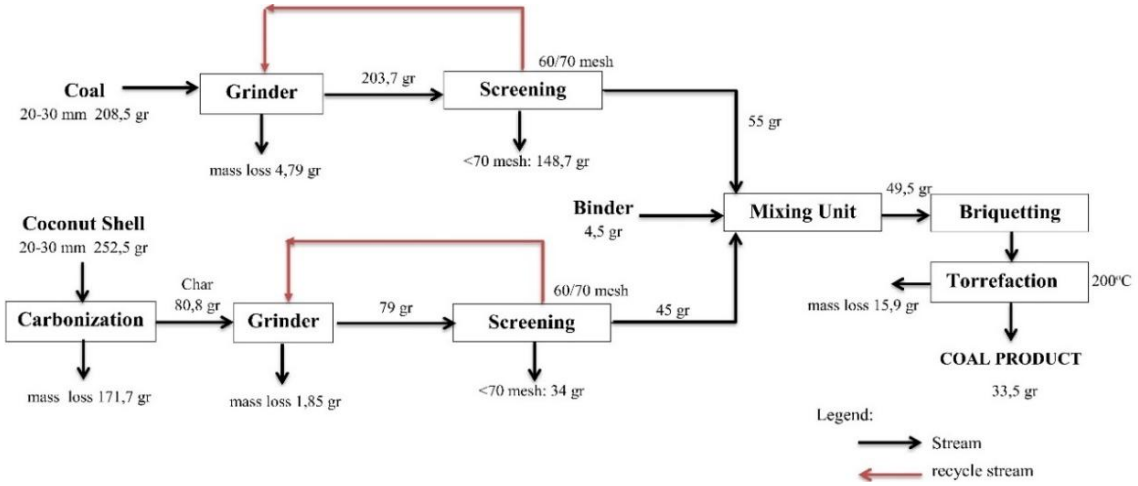


Figure 1. Flow diagram for low-temperature co-torrefaction of low-rank coal with coconut shell charcoal.

At a torrefaction temperature of 200 °C, dehydration is the main reaction, followed by the decomposition of oxygenate groups, such as hydroxyl and carboxyl groups. These reactions release H_2O and CO_2, as well as a small amount of CO, which reduces the oxygen content and increases the relative carbon content of the solid fuel. Although the temperature is not high enough for extensive carbonization, some deoxygenation still occurs during the process, resulting in a hybrid fuel with a higher relative carbon content. Similar mass loss during mild torrefaction has also been reported in biomass briquettes, where dehydration and the release of volatile compounds contribute to the improvement of fuel properties (Paulo *et al*., 2023). The use of pre-carbonized coconut shell charcoal also supports the upgrading process because it already has a high fixed carbon content and low moisture content. The coconut shell charcoal not only acts as an additive in the briquette, but also contributes to carbon enrichment. This condition can be related to the improvement in proximate composition, calorific value, and carbon structural ordering, which are discussed in the following sections.

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Thermogravimetric and Proximate Analysis

Thermogravimetric analysis was used to determine the proximate composition of raw coal, coconut shell charcoal, and hybrid coal samples. The results are presented in Table 1. Raw low-rank coal B100 had a high moisture content of 30.06% and volatile matter of 40.06%, while its fixed carbon content was relatively low at 27.69%. These values are characteristic of Indonesian lignite. As the proportion of coconut shell charcoal increased, the hybrid briquettes showed a decrease in moisture and volatile matter, while the fixed carbon content increased. Among the samples, C45 had the lowest moisture content of 8.46% and the highest fixed carbon content 59.94%. These results indicate that the addition of coconut shell charcoal improved the proximate composition of the briquettes after low-temperature co-torrefaction. Because the cassava starch binder was incorporated at a constant proportion of 10 wt% in all briquette formulations, its contribution to the proximate composition and calorific value was assumed to be identical for all samples. Therefore, the observed differences in fuel properties were primarily attributed to the increasing proportion of coconut shell charcoal and the co-torrefaction treatment rather than variations in binder content.

The marked decrease in moisture content is largely attributed to dehydration during torrefaction and to the inherently lower moisture content of the pre-carbonized coconut shell charcoal. At approximately 200 °C, physically adsorbed water is readily removed, while weakly bound water associated with oxygen-containing functional groups is partially eliminated. The addition of pre-carbonized biomass, therefore, not only dilutes the high-moisture lignite but also promotes the production of a more hydrophobic solid fuel. comparable decrease in moisture content during coal–biomass torrefaction has been documented by Rizkiana *et al*. (2024), confirming that mild thermal treatment effectively improves the storage stability and fuel quality of low-rank coal.

A pronounced decrease in volatile matter was also observed as coconut shell charcoal content increased, from approximately 40% in raw coal to 29.49% in the C45 sample. The reduction in volatile matter is related to the decomposition of oxygen-containing functional groups, such as hydroxyl, carbonyl, and carboxyl groups, which release H_2O, CO_2, and CO during torrefaction (Zhang *et al*., 2025). These reactions remove oxygen from the solid product and cause changes in its chemical structure, resulting in a lower oxygen-to-carbon ratio. Similar changes have been reported during biomass and coal torrefaction, where the release of oxygen-rich compounds can improve fuel stability and combustion properties.

In contrast, the fixed carbon content increased from 27.69% in raw lignite to 59.94% in the C45 hybrid briquette. This increase is related to the removal of moisture and volatile matter during heating, which increases the relative proportion of fixed carbon in the remaining solid. In addition, coconut shell charcoal, which has a fixed carbon content of approximately 74.5%, contributes carbon-rich material to the hybrid briquettes. Therefore, the combination of low-rank coal and coconut shell charcoal through co-torrefaction improved the fixed carbon content and overall fuel quality. The fixed carbon content of C45 was also higher than the value reported for Ombilin Basin bituminous coal, with fixed carbon at 48.78% (Algifari *et al*., 2023). This result shows that the addition of coconut shell charcoal can improve the proximate composition of low-rank coal even at a relatively low treatment temperature. Meanwhile, the ash content remained relatively stable, ranging from 2.1% to 2.8%. This indicates that the addition of coconut shell charcoal did not cause a substantial increase in the ash fraction of the briquettes. Maintaining a low ash content is beneficial because excessive ash can reduce combustion efficiency and contribute to slagging and fouling during fuel utilization.

Table 1. Proximate analysis results of solid fuel samples.

Sample	Moisture (%)	Volatile Matter (%)	Ash (%)	Fixed Carbon (%)
C5a	10.41	47.83	2.83	38.93
C5b	10.42	46.87	2.82	39.89
C25a	13.66	39.19	2.51	44.64
C25b	13.63	38.09	2.43	45.85
C45a	8.48	29.43	2.11	59.98
C45b	8.44	29.55	2.11	59.90
C100a	6.09	17.25	2.20	74.46
C100b	6.11	17.14	2.21	74.54
B100a	30.11	40.99	2.04	26.86
B100b	30.01	39.12	2.34	28.53

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Figure 2 shows the relationship between the coconut shell charcoal content and the proximate composition of the hybrid briquettes. As the proportion of coconut shell charcoal increased, moisture and volatile matter generally decreased, while fixed carbon increased. Linear regression showed that fixed carbon had the strongest relationship with charcoal content R^2= 0.9249, followed by volatile matter R^2= 0.9161. These results indicate that the proportion of coconut shell charcoal was closely related to the changes in fixed carbon and volatile matter during the process. In contrast, moisture R^2= 0.4365 and ash content R^2= 0.2087 showed weaker relationships with charcoal content. This suggests that these two parameters were less strongly associated with the proportion of coconut shell charcoal in the samples.

The decrease in moisture and volatile matter, together with the increase in fixed carbon, indicates that the combination of low-rank coal and coconut shell charcoal affected the proximate composition of the briquettes during co-torrefaction. These changes may be related to moisture removal and the release of volatile compounds during heating, as well as the contribution of carbon-rich coconut shell charcoal. The changes in proximate composition are also relevant to the calorific value of the hybrid briquettes, which is discussed in the following section.

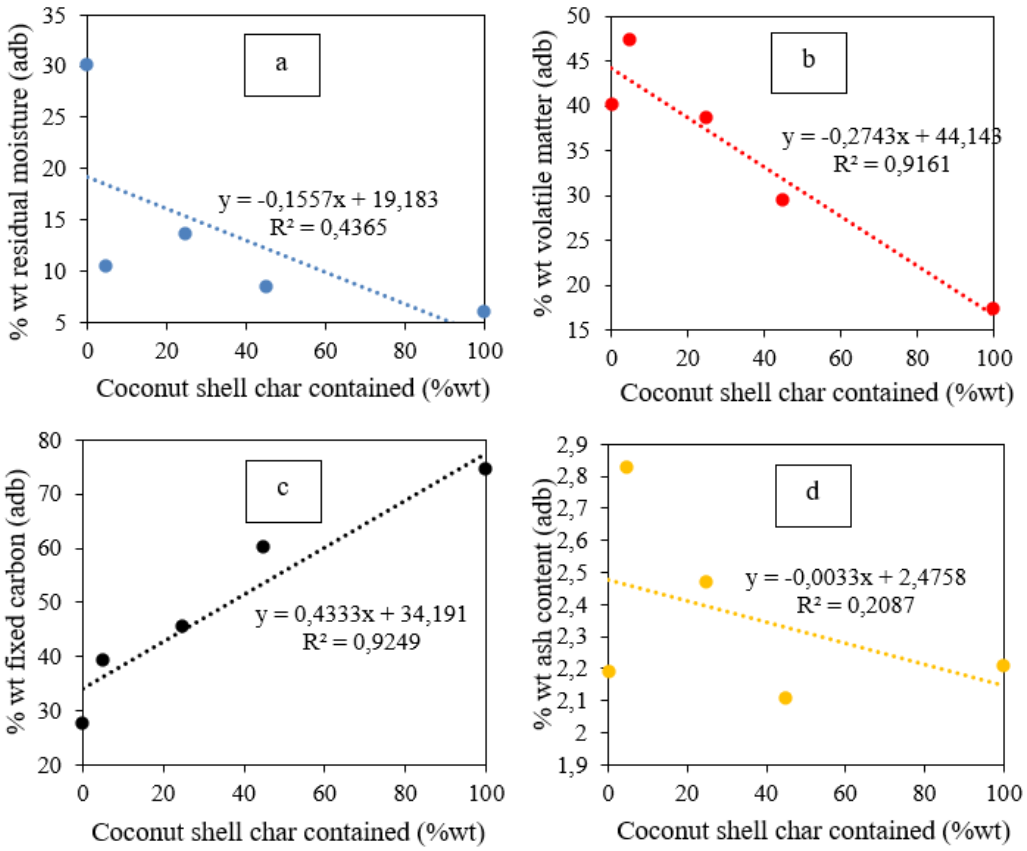


Figure 2. Effect of coconut shell charcoal content on (a) moisture, (b) volatile matter, (c) fixed carbon, and (d) ash content of the solid fuel.

Combustion Performance, Energy Densification, and Fuel Upgrading

The combustion performance of the hybrid coal briquettes was evaluated using bomb calorimetry, and the results are presented in Table 2. The calorific value increased markedly from 4823.46 kcal/kg for the raw lignite to 6786.28 kcal/kg for the C45 hybrid coal, representing an increase of approximately 40.7%. This value approaches the calorific value of the reference bituminous coal 7340 kcal/kg (Algifari *et al*., 2023), demonstrating that low-temperature co-torrefaction effectively upgraded the energy quality of the low-rank coal.

The increase in calorific value is closely associated with the physicochemical transformations identified by the proximate analysis. As discussed in the previous section, torrefaction decreased the moisture and volatile matter contents significantly while increasing the fixed carbon fraction. Selective removal of water and oxygen-rich volatile compounds concentrates the remaining carbonaceous matrix, thereby increasing the energy stored per unit mass of fuel. In addition, incorporating pre-carbonized coconut shell charcoal, which has an elevated fixed

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carbon content and a comparatively high calorific value, further enhances the hybrid fuel. Similar relationships between fixed carbon enrichment and calorific value have confirmed that carbon enrichment is the primary factor governing the energy upgrading of coal–biomass composite fuels (Sofán-Germán *et al*., 2025).

The effectiveness of the upgrading process can be further evaluated using the energy densification ratio and energy yield (Soria-Verdugo *et al*., 2024). Based on the calorific value of the C45 sample, the calculated EDR reached 1.41, indicating that the energy density of the torrefied hybrid coal increased by approximately 41% compared with the raw lignite. Considering the measured mass yield of 67.7%, the corresponding energy yield was approximately 95.5%. This high energy yield indicates that although about one-third of the initial mass was removed during torrefaction, most of the original energy was retained within the solid product. The preferential removal of moisture and low-energy volatile compounds, therefore, resulted in efficient energy concentration rather than significant energy loss.

Unlike the substantial changes observed in moisture, volatile matter, and fixed carbon, the ash content remained nearly constant. The low ash content is advantageous for practical combustion because it minimizes slagging and fouling tendencies while maintaining high combustion efficiency. Furthermore, the increased fixed carbon content and reduced volatile matter are expected to improve combustion stability by promoting longer and more uniform burning characteristics while reducing the release of volatile organic compounds during combustion (Song *et al*., 2024). The combustion results demonstrate that co-torrefaction with coconut shell charcoal provides an effective strategy for upgrading Indonesian low-rank coal into a higher-quality solid fuel. The simultaneous improvement in calorific value, energy density, and energy yield reflects the combined effects of low-temperature torrefaction and the incorporation of pre-carbonized coconut shell charcoal. These findings are consistent with the proposed deoxygenation pathway, in which dehydration and devolatilization increase the relative carbon concentration while preserving the predominantly low-rank carbon structure.

Table 2. Calorific value analysis for some organic materials.

Parameter	Brown Coal	Hybrid Coal (C45)
Calorific value (kcal/kg)	4823.46	6786.28
Energy Densification Ratio	1.00	1.41
Mass Yield (%)	–	67.7
Energy Yield (%)	–	95.5

X-ray Diffraction Analysis

X-ray diffraction analysis was conducted to characterize the carbonaceous material of low-rank coal before and after low-temperature co-torrefaction with coconut shell charcoal. The diffraction patterns of raw lignite B100, coconut shell charcoal C100, hybrid coal C45, and reference coal ranks are presented in Figure 3. All samples showed broad diffraction patterns instead of sharp crystalline peaks, suggesting that the carbonaceous materials were predominantly composed of disordered turbostratic carbon. Such broad diffraction characteristics are typical of lignite, biomass-derived char, and other low-temperature carbonized materials, in which the carbon layers exhibit limited stacking order and remain far from the highly crystalline structure of graphite.

The broad diffraction pattern was observed at approximately $2\theta = 25 – 26°$ aligns with the (002) reflection plane of turbostratic carbon and is associated with aromatic carbon stacking (Tian *et al*., 2026). A second broad reflection at approximately $2\theta = 43 – 47°$ is assigned to the (100) plane, representing the in-plane ordering of aromatic carbon (Tian *et al*., 2026). The broad nature of both reflections indicates that the carbonaceous structure remained predominantly turbostratic and poorly ordered after co-torrefaction. This behavior is consistent with the relatively low treatment temperature at 200 °C, which is insufficient to induce graphitization. Although no new crystalline carbon phases were detected, the (002) diffraction feature of the hybrid coal appeared slightly more pronounced than that of the raw lignite while remaining characteristically broad. These observations suggest that co-torrefaction promoted limited structural rearrangement and a slight increase in short-range carbon ordering.

Nevertheless, the absence of sharp graphite reflections confirms that the structural evolution remained limited to early-stage carbonization rather than the formation of highly ordered graphitic structures. The observed structural evolution is supported by the results of the proximate data and calorific value measurements. During co-torrefaction, dehydration and devolatilization selectively removed oxygen-rich functional groups, while the addition of pre-carbonized coconut shell charcoal increased the relative carbon concentration of the hybrid fuel. Similar structural evolution during biomass and coal thermochemical treatment has been observed, with carbon

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enrichment accompanied by increased aromatic ordering while maintaining predominantly turbostratic carbon structures (Li *et al*., 2024).

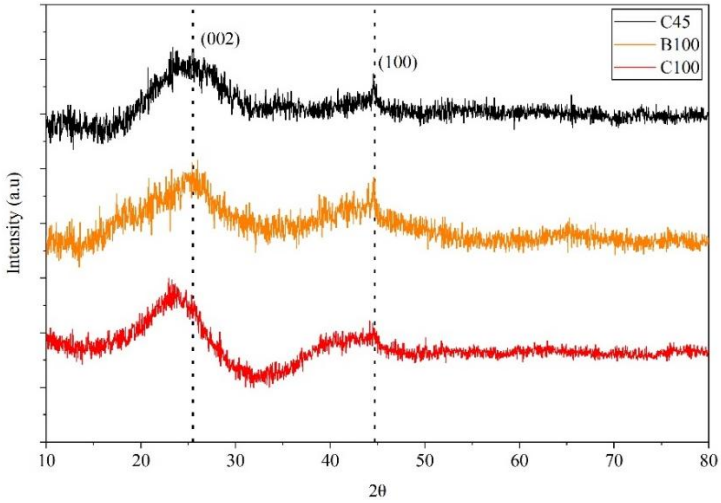


Figure 3. X-ray diffraction patterns of raw lignite B100, coconut shell charcoal C100, and co-torrefied hybrid coal C45.

The comparison with the reference coal ranks further illustrates the extent of structural modification during co-torrefaction. The hybrid coal retained a diffraction pattern more closely resembling lignite than bituminous coal or anthracite, indicating that low-temperature co-torrefaction did not produce extensive structural ordering or graphitization (Li *et al*., 2024). Although the position of the (002) reflection and the corresponding interlayer spacing d_{002} remained essentially unchanged in Table 3, the hybrid coal exhibited a slightly more pronounced (002) diffraction feature than the raw lignite. A decrease in the FWHM of both the (002) and (100) reflections was also observed. However, because the FWHM values were obtained from comparative peak analysis using Origin software rather than from full-profile peak fitting or crystallite size analysis, these changes should be interpreted qualitatively. Therefore, the observed peak narrowing is considered indicative of limited short-range structural rearrangement instead of definitive evidence of increased crystallinity or graphitization (Sousa *et al*., 2024). The improvement in fuel quality is primarily attributed to carbon enrichment associated with dehydration and devolatilization during co-torrefaction rather than to the formation of highly crystalline graphite. These structural observations are consistent with the simultaneous increase in fixed carbon content and calorific value.

Table 3. XRD peak parameters of low-rank coal, coconut shell charcoal, and hybrid coal samples.

Sample	$2\theta_{(002)}$	d_{002} (nm)	$FWHM_{002}$ (°)	$2\theta_{(100)}$	$FWHM_{100}$ (°)
C100	25.34	0.3512	0.25	44.48	0.23
B100	25.39	0.3505	0.17	44.61	0.56
C45	25.39	0.3505	0.07	44.55	0.06

*FWHM values were obtained from peak analysis in Origin software and are presented for comparative evaluation of peak broadening.

CONCLUSION

This study showed that low-temperature co-torrefaction with coconut shell charcoal improved the physicochemical properties of Indonesian low-rank coal. As the proportion of coconut shell charcoal increased, the moisture content decreased from 30.06% to 8.46%, while the fixed carbon content increased from 27.69% to 59.94%. The calorific value also increased from 4823.46 to 6786.28 kcal/kg, indicating an improvement in the energy content of the solid fuel. X-ray diffraction analysis showed a slight increase in short-range carbon ordering, although the hybrid coal still retained the predominantly amorphous structure of the original lignite. These results indicate that coconut shell charcoal can be used as a carbon-rich additive to improve low-rank coal through low-temperature treatment. The improvement in fuel properties was mainly related to moisture removal, the release of volatile matter, and the addition of carbon-rich material, rather than major changes in the carbon structure. However, this study was limited to proximate analysis, calorific value, and X-ray diffraction, without ultimate analysis or detailed kinetic analysis. Further studies should include ultimate analysis, Van Krevelen analysis, kinetic modeling, and scale-up experiments to better understand the co-torrefaction process and its potential for industrial application.

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CONFLICT OF INTEREST

The authors declare no conflict of interest with any party. This work was supported by institutional funding.

AUTHOR CONTRIBUTION

AA: Conceptualization, Methodology, Investigation, Data Analysis, Visualization, Manuscript Drafting, and Editing; FN: Investigation, Data Curation, and Laboratory Experiments; SRY: Data Analysis, Validation, and Visualization; HP: Supervision, Project Administration, and Manuscript Review; and JJ: Resources and Manuscript Review.

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DECLARATION OF GENERATIVE AI

During the preparation of this article, ChatGPT was used as an auxiliary tool to support drafting, paraphrasing, scientific development, and English-language refinement. The experimental work and data processing were conducted by the author. The author reviewed the AI-assisted content, assessed the scientific interpretations against the experimental results and relevant literature. The authors take full responsibility for the accuracy, originality, and integrity of the article.

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