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Research Article

Enhancing bio-char calorific value through catalytic pyrolysis: The role of magnesium oxide-zeolite based catalysts

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Abstract. The current research aims to improve the generation of bio-char with elevated higher heating values (HHVs) by utilizing magnesium oxide-zeolite-based catalysts across various temperature conditions. The exploration of biomass catalytic pyrolysis has intensified in the pursuit of sustainable energy solutions. Catalytic pyrolysis offers a technique to convert abundant and renewable biomass resources into valuable biofuels and bio-char, thereby improving energy security and reducing dependence on fossil fuels. The use of suitable catalysts in biomass catalytic pyrolysis is crucial for enhancing the yield of bio-char with higher calorific value. This investigation explores the impact of magnesium oxide-zeolite-based catalysts on the higher heating values of bio-char generated from coconut shells. The initial findings indicate a notable enhancement in the calorific value of bio-char. The HHV increased from 12.03 MJ/kg for untreated coconut shells to 20.06 MJ/kg with ZSM-5, ultimately reaching an impressive 38.11 MJ/kg with the MgO/ZSM-5 catalyst. The results demonstrate that the addition of magnesium oxide significantly improves the energy content of bio-char. Various combinations of magnesium oxide, such as MgO/ZSM-5, MgO/Y2O3/ZSM-5, and MgO/Mn3O4/ZSM-5, are evaluated for their effects on the pyrolysis process. The results demonstrate that the impregnation of metal oxides into zeolite catalysts enhances catalytic performance and facilitates the efficient conversion of coconut shells into high-energy bio-char. The findings highlight the promise of metal oxide-zeolite catalysts in improving bio-char quality and promoting the development of sustainable energy technologies.

Keywords: Catalytic Pyrolysis; Magnesium Oxide-Zeolite Catalysts; Bio-Char; Higher Heating Value (HHV); Sustainable Energy Conversion

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1. Introduction

The increasing need for energy and the need to address environmental issues require the advancement of renewable energy sources. Biomass catalytic pyrolysis is a feasible method for transforming agricultural waste into useful bio-char, which may be utilized as a sustainable energy source. Bio-char, which is obtained from biomass through thermal combustion in an oxygen-limited setting, is a carbon-rich substance (Lehmann and Joseph, 2015; Mohanty *et al.*, 2018). Bio-char possesses distinctive characteristics, including a substantial surface area, significant porosity, functional groups, high cation exchange capacity, and stability, which render it appropriate for diverse applications (Yaashikaa *et al.*, 2020). The advantages of bio-char include its rapid and simple manufacture, environmentally friendly properties, reusability, and cost-effectiveness (Gayathri *et al.,* 2021; Hemavathy *et al.*, 2020). The physiochemical properties of bio-char exhibit significant variation depending on factors such as the type of feedstock and the conditions of pyrolysis, including temperature and catalyst (Elbasiouny *et al.,* 2021). Generally, raising the temperature of pyrolysis has the effect of increasing the nutrient contents, specific surface area, and pH of bio-char, as well as the levels of ash and carbon. However, it results in low values of cation exchange capacity (CEC) and volatile matter content (Elbasiouny *et al.,* 2021; Rashid *et al.*, 2020; Tomczyk *et al.*, 2020). The rise in pyrolysis temperature results in an augmented carbon content, which is attributed to a greater degree of polymerization (Domingues *et al.,* 2017). Consequently, the bio-char exhibits more compact carbon structure (Lehmann & Joseph, 2009).

Furthermore, in the absence of oxygen, pyrolysis is a thermal degradation process that breaks down biomass into three distinct product fractions: solid residue (bio-char), condensable vapours that yield a liquid product fraction (bio-oil), and non-condensable gaseous products (gas) (Bianasari *et al.,* 2024). In order to achieve the desired result, it is essential to meticulously select the pyrolysis method, as each variant of pyrolysis yields various proportions of components (Gezae & Chandraratne, 2018). Catalytic pyrolysis is a pyrolysis technology that can be employed to optimize the composition and yield of the desired product. Several catalysts, such as alumina, Al-MCM-41, magnesium oxides, nickel oxides, and ZSM-5, have been found to positively impact the production of bio-char (Bianasari et al., 2024; Stefanidis et al., 2011). The biomass treated with ZSM-5 resulted in a higher bio-char production compared to the biomass treated with alumina oxide

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and sodium carbonate bio-char (Stefanidis *et al.*, 2011). Separately, according to prior research conducted by Pütün (2010), the addition of magnesium oxide in the raw material resulted in an enhancement of the oil's quality, as evidenced by an increase in its calorific value and H/C ratio (Pütün, 2010).

Nevertheless, numerous studies prioritize the utilization of pyrolysis as a means to generate bio-oil from biomass *(Isahak et al.*, 2012; Lachos-Perez *et al.*, 2023). Bio-oil, a liquid produced through pyrolysis, is considered to be one of the most promising biofuels, alongside bio-diesel and bio-ethanol. This is due to its carbon-neutral nature, making it very desirable for future use (Martínez *et al.*, 2014). Bio-oil produces much lower levels of sulphur dioxide and nitrogen dioxide emissions compared to conventional fossil fuels (Terry *et al.*, 2021). Additionally, it is important to explore the potential of bio-char, especially as a solid fuel with a high higher heating value that can function as a renewable and carbon-rich energy source. The production of bio-char via catalytic pyrolysis presents a cleaner and more sustainable option compared to fossil-derived solid fuels like coal, while also playing a crucial role in carbon sequestration (Afshar & Mofatteh, 2024; Zhou *et al.*, 2022). This positions it as a valuable asset in strategies aimed at mitigating climate change. The solid state facilitates straightforward handling, transportation, and storage, presenting clear benefits for rural electrification and decentralized energy systems. Beyond its energy applications, bio-char is recognized for its diverse functionalities. This substance serves as a soil amendment, enhancing fertility, water retention, and promoting microbial activity (Waheed *et al.,* 2025). The material demonstrates significant promise in the field of environmental remediation, attributed to its extensive surface area and impressive adsorption capacity (Beljin *et al.,* 2025).

The transformation of bio-char is in harmony with circular economy principles, as it turns waste into valuable products. In this context, the choice of raw materials is of utmost importance. The plentiful and often overlooked lignocellulosic biomass resources, including coconut shells, offer a compelling feedstock option due to their elevated carbon content, minimal ash, and extensive availability in tropical and subtropical areas (Preeti *et al.,* 2024; Umerah *et al.*, 2020). Agricultural residues are frequently viewed as waste, resulting in their disposal or open burning, which contributes to environmental pollution and suboptimal resource utilization. The transformation of these materials into bio-char serves to minimize waste and emissions while simultaneously fostering economic prospects for local communities via the establishment of bio-based industries.

Enhancing the quality of bio-char, particularly regarding calorific value, carbon content, and stability, necessitates a comprehensive grasp of the pyrolysis process and the selection of appropriate catalysts. This involves investigating how metal oxide–zeolite catalysts affect reaction pathways, enhance carbon retention, and optimize the energetic characteristics of the resulting char. Ongoing investigation into catalyst development and biomass utilization is crucial to fully harness the energy potential of bio-char. Prior studies have demonstrated that the use of ZSM-5 and MgO has a positive impact on the yield of bio-char (Bu *et al.*, 2021; Y. Zhang *et al.*, 2023). Additionally, the addition of MgO has the potential to enhance the calorific value of bio-char (Bianasari *et al.,* 2024; Pütün, 2010; Stefanidis *et al.*, 2011). MgO, known for its thermal stability as a basic metal oxide, is essential for maintaining catalytic activity in high-temperature pyrolysis environments (Lee *et al.*, 2025). The stability of this compound enables its persistence during the carbon generation process, facilitating deoxygenation reactions and reducing catalyst degradation. This leads to improved carbon retention and the generation of bio-char that possesses better structural integrity and energy content. ZSM-5, a microporous zeolite characterized by its strong acidity, plays a crucial role in regulating the thermal decomposition of biomass and affects the distribution of products (Karim *et al.*, 2024; Socci *et al.,* 2022). The integration of MgO and ZSM-5 establishes a harmonious interplay between basic and acidic catalytic roles, facilitating reaction mechanisms that mitigate excessive volatilization while enhancing the production of stable, carbon-rich bio-char with an elevated calorific value.

Recent studies have investigated the possibility of integrating extra metal oxides into dual-catalyst systems to enhance catalytic performance during biomass pyrolysis. Transition metal oxides like Mn_3O_4 provide redox functionality that can aid in the cracking of heavier organic molecules and enhance the formation of desirable pyrolysis products through improved deoxygenation reactions (Hubble et al., 2023; W. Li et al., 2022). Separately, rare-earth oxides such as Y_2O_3 are recognized for their ability to enhance catalyst stability and create basic surface sites, which can affect reaction pathways and product selectivity (Narasimharao & Al-Sultan, 2020; K. Xu et al., 2022). The anticipated modifications aim to improve carbon retention and affect the physicochemical characteristics of the resulting bio-char. Thus, incorporating these metal oxides into MgO-zeolite catalyst shows potential for enhancing the energy density and higher heating value (HHV) of bio-char obtained from lignocellulosic biomass. Therefore, this research aims to investigate the advantages of utilizing appropriate catalysts, particularly MgO-zeolite based catalysts, to improve the higher heating values (HHVs) of bio-char derived from coconut shell biomass. The objective is to facilitate the advancement of sophisticated catalytic systems that can improve the energy potential of bio-char and encourage sustainable waste-to-energy technologies.

2. Material and methods

2.1. Biomass Preparation

The coconut shell biomass was obtained, dehydrated, and pulverized in a grinder to get finely granulated particles with a consistent size for the pyrolysis procedure. The coconut shell powder was sieved using conventional sieves to obtain a uniform particle size less than 1 mm. It will serve as a feedstock in the pyrolysis reactor.

2.2. Catalyst Preparation

Magnesium oxide-zeolite based catalysts were synthesized using a wetness impregnation method. A 5% (w/w) solution of $Mg(NO_3)_2{\cdot}6H_2O$ was combined with 5 g of ZSM-5 catalyst at room temperature for 24 h, leading to the formation of MgO/ZSM-5. In order to produce MgO/Mn_3O_4/ZSM-5, a mixture of 5 g of $Mn(NO_3)_2{\cdot}4H_2O$ and $Mg(NO_3)_2{\cdot}6H_2O$ is prepared by diluting them in a 1:1 ratio to obtain solution with a concentration of 5% (w/w). Following that, the combined solutions were blended with 5 g of ZSM-5 for 24 h, resulting in the production of MgO/Mn_3O_4/ZSM-5. Likewise, the same procedure is used to synthesize MgO/Y_2O_3/ZSM-5. Further, the solution underwent a 5-hour drying process at 110°C following wet impregnation approach. Subsequently, the calcination procedure was conducted at 550°C for 5 h.

2.3. Biomass Catalytic Pyrolysis

A stainless steel fixed bed reactor was utilized to carry out the in situ pyrolysis of coconut shell. The reactor was placed inside a horizontally oriented tube furnace. This study was conducted to investigate the influence of temperature and catalysts on biomass catalytic pyrolysis. The conversion of 20 g of coconut shells into products was conducted at temperatures of 400°C, 500°C, and 600°C. In addition, the catalyst used for each run, namely MgO/ZSM-5, MgO/Y_2O_3/ZSM-5, and MgO/Mn_3O_4/ZSM-5, was 10% of the weight of the sample. The reaction was carried out for 30 min at a heating rate of 10°C/min. Nitrogen gas was chosen as the carrier gas during the experiment to remove impurities from the system and establish a non-reactive atmosphere. The flow rate of nitrogen gas was consistently maintained at 30 mL/min. The catalytic pyrolysis process produces biogas, bio-char, and bio-oil as its outputs. Equation (1) explains the determination of product yield by integrating the exact quantity of the outcome (bio-oil, bio-char, or biogas) with the initial weight of biomass (Fogler, 2016).

$$Yield\,(\%wt) = \frac{m_{desired\ product}}{m_{biomass}} \times 100\% \quad (1)$$

This equation measures the yield of a particular pyrolysis product (bio-oil, bio-char, or biogas) expressed as a percentage of the initial biomass input. The comparison involves the mass of the desired product to the initial mass ofthe biomass feedstock. This approach employs a fundamental mass-balance technique frequently utilized in biomass conversion investigations to evaluate the effectiveness of the pyrolysis process.

2.4. X-Ray Diffraction Analysis (XRD)

The X-ray diffraction (XRD) analysis was conducted using the Aeris fast XRD system manufactured by Malvern Panalytical Company. The system came with a Benchtop X-Ray Diffractometer. The system used Cu Kα radiation with an energy of 40 kilovolts and a current of 15 milliamperes. The scanning angle range extended from 5° < 2θ > 80°, with the measurement temperature fixed at 25°C. XRD data acquired will be examined with HighScore and OriginLab software.

2.5. Calorific Value Analysis

The biomass sample and bio-chars were analyzed to evaluate their higher heating values (HHVs) utilizing calorific value analysis using a bomb calorimeter, following the ASTM D 5468–02 technique (ASTM International, 2017). The sample's calorific value (Q) can be determined by applying Equation 4.

$$Qfuse\,(J/g) = mc \times qc \quad (2)$$

$$Qign\,(J/g) = mw \times qw \quad (3)$$

$$Calorific\ value,\ Q\,(J/g) = \frac{(\varepsilon.\Delta T)-(Qfuse+Qign)}{ms} \quad (4)$$

Where, $Qfuse$ is heat contributed from the cotton thread(J), $Qign$ heat contributed from the nichrome ignition wire (J)., Q calorific value of the sample (J/g), qc calorific value of cotton fuse (J/g), qw calorific value of nichrome wire (J/g), mc mass of cotton (g), mw mass of nichrome wire (g), ms mass of sample (g), ε calorific value of benzoid acid (J/K), ΔT $T_2 - T_1$ = final temperature – initial temperature (K).

This procedure assesses the calorific value by measuring the total heat released during the complete combustion of a known mass of sample in a pressurized oxygen environment. For each experiment, around 1 g of the dried sample was accurately weighed and positioned in a combustion crucible within the bomb calorimeter. A cotton thread served as the fuse, while a nichrome ignition wire was utilized to trigger combustion. The bomb was charged with pure oxygen at a pressure of 30 atm and submerged in a calorimetric vessel containing a known quantity of water. Following ignition, the recorded temperature increase (ΔT) was noted.

The calorific value was determined by considering the energy contributions from the cotton fuse and the nichrome wire. The total heat released was adjusted for these components and standardized to the mass of the sample. This method guarantees a precise assessment of the calorific value of bio-char by focusing exclusively on the energy derived from the sample itself, while eliminating any influence from external ignition sources. The resulting HHV is an essential parameter in evaluating the energy potential of the bio-char as a solid biofuel.

3. Results and Discussion

3.1. X-Ray Diffraction Analysis (XRD) Results of Catalysts

Wetness impregnation was used to produce magnesium oxide-zeolite based catalysts. It is necessary to run an XRD analysis to determine whether wet impregnation causes changes to or damage to the zeolite's crystallinity and structure. The XRD patterns of the ZSM-5, MgO/ZSM-5, MgO/Y_2O_3/ZSM-5, and MgO/Mn_3O_4/ZSM-5 catalysts are shown in Figure 1. It is apparent that for 2θ = 7.9°, 9.0°, 23.0°, 23.3°, 23.9°, 29.0°, and 30.0°, all catalysts show noticeable peaks. The fact that the maximum intensity of the distinct peaks occurs within the same peak range suggests that the structure and crystallinity of the ZSM-5 zeolite were not changed during the metal-oxide impregnation procedures.

The characteristic peaks of ZSM-5 are complemented by the confirmation of additional metal oxides. MgO/ZSM-5 exhibits a new diffraction peak at around 2θ = 43°, which aligns with the cubic phase of MgO (JCPDS No. 45-0946), thereby confirming the successful integration of MgO onto the ZSM-5 surface (Pei et al., 2010; Serhal et al., 2018). The XRD pattern for the MgO/Y_2O_3/ZSM-5 catalyst reveals additional peaks at 2θ = 29°, 48°, and 57°, corresponding to the characteristic diffraction pattern of Y_2O_3 (JCPDS No. 41-1105) (Hyun et al., 2021; Krsmanović Whiffen et al., 2012). The observed peaks demonstrate the effective incorporation of yttrium oxide while maintaining the integrity of the ZSM-5 framework, as evidenced by the clarity and prominence of the primary ZSM-5 peaks.

In a similar manner, MgO/Mn_3O_4/ZSM-5 catalyst exhibits multiple new peaks at approximately 2θ = 32°, 36°, and 72°, which are associated with the Mn_3O_4 spinel phase (JCPDS No. 24-0734) (Chen et al., 2019). This indicates the existence of manganese oxide, and its incorporation seems to preserve the structural integrity of the zeolite.

The XRD analysis clearly indicates that the loading of metal oxides through wetness impregnation leads to the effective deposition of metal oxides (MgO, Y_2O_3 and Mn_3O_4) onto the ZSM-5 support, while preserving the crystalline structure of the zeolite intact. The consistent presence of characteristic ZSM-5 peaks in all catalysts reinforces the notion that the framework is preserved following the impregnation process. Futhermore, the crystallinity of XRD diffraction patterns from 5° to 80° was assessed using OriginLab and presented in Table 1. Substituting

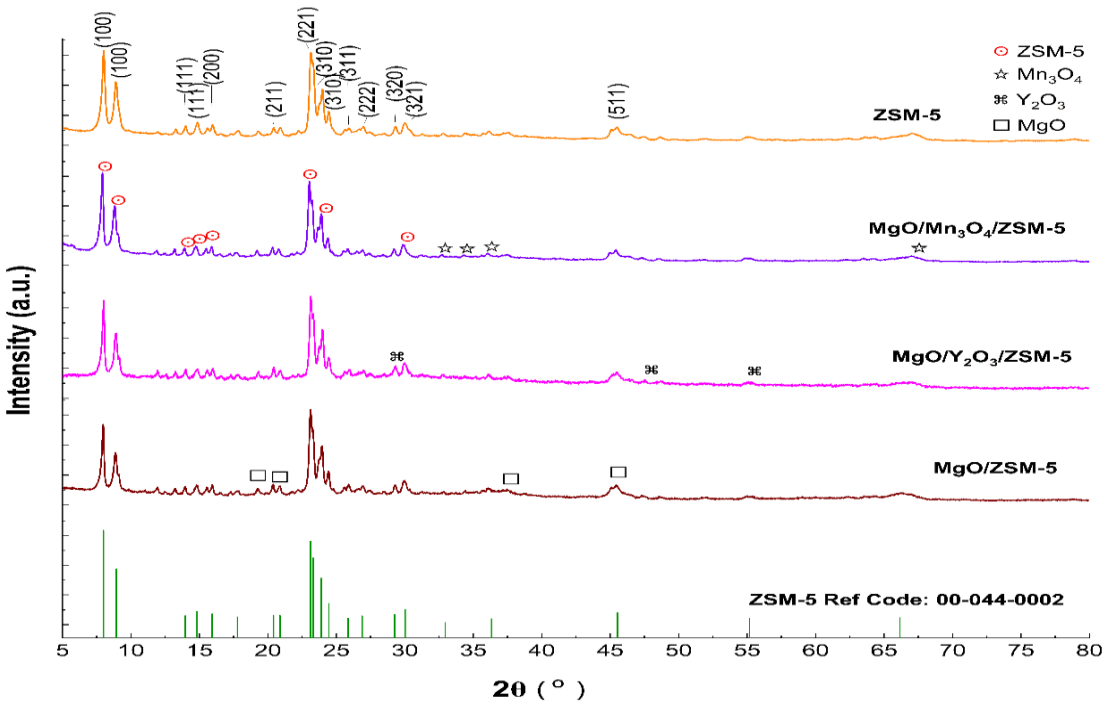

Fig 1. XRD patterns of magnesium oxide-zeolite based catalysts used in the coconut shell catalytic pyrolysis

ZSM-5 with MgO/ZSM-5, MgO/Y_2O_3/ZSM-5, and MgO/Mn_3O_4/ZSM-5 during pyrolysis revealed an enhancement in the catalyst's crystallinity. Crystallinity denotes t he degree to which a material displays a systematic, periodic organization of atoms or molecules within a crystalline structure (Rawat *et al.*, 2023). Materials exhibiting high crystallinity showcase well-defined structures, often leading to notable physical properties like precise melting points, uniform shapes, and improved mechanical strength (Kohutiar *et al.,* 2025; Murmu et al., 2022). Meanwhile, amorphous materials do not possess this long-range order and display a greater variability in properties, such as reduced mechanical strength and undefined thermal transitions (Rawat *et al.*, 2023; Vranic, 2004).

The addition of MgO to ZSM-5 with an appropriate amount through wet impregnation followed by calcination led to a 30.02% enhancement in relative crystallinity, rising from 35.72% to 46.44%. The improvement observed can be linked to the interaction between Mg^{2+} ions and the zeolite framework, which may help stabilise the structure under high-temperature conditions and inhibit framework degradation (Qu et al., 2020; Shahed et al., 2024; Yuan et al., 2019). Additionally, the modification of ZSM-5 with dual-metal oxides, specifically MgO and Y_2O_3, resulted in an increase in crystallinity to 41.31%, reflecting a 15.65% improvement. Y_2O_3, recognised for its robust Lewis acidity and structural enhancement, has the potential to work in conjunction with MgO to maintain the stability of the zeolite lattice throughout the calcination process (Hata et al., 2021; Lin et al., 2013). Separately, a significant enhancement in crystallinity (56.35%) was noted for MgO/Mn_3O_4/ZSM-5, with relative crystallinity increasing from 35.72% to 55.85%. The significant enhancement indicates that the addition of Mn_3O_4 strengthens the crystalline structure, likely due to its redox characteristics and its interaction with the zeolite framework (Kaczmarczyk et al., 2016; Kim et al., 2012). This interaction may contribute to preserving lattice alignment and preventing collapse during thermal treatment.

3.2. Product Distribution

The pyrolysis process for obtaining bio-char is influenced by several factors, including the type of feedstock, temperature, and catalysts (Elbasiouny et al., 2021; Fernández et al., 2014). Bio-chars derived from animal litter and solid waste feedstocks demonstrate reduced surface areas, carbon content, volatile matter, and elevated cation exchange capacity (CEC) in comparison to bio-chars derived from crop residue and wood biomass, even when subjected to greater pyrolysis temperatures (Tomczyk et al., 2020). The cause for this disparity is significant variance in lignin and cellulose content, as well as moisture content of biomass. Furthermore, biomass can be categorized into two different types: woody and non-woody.

Table 1
XRD crystallinity assessment of the catalysts used in this study

Catalyst	Treatment for Catalyst	Relative Crytallinity(%)
ZSM-5	Calcination on ZSM-5	35.72%
MgO/ZSM-5	Wet impregnation, calcination	46.44%
MgO/Y_2O_3/ZSM-5	Wet impregnation, calcination	41.31%
MgO/Mn_3O_4/ZSM-5	Wet impregnation, calcination	55.85%

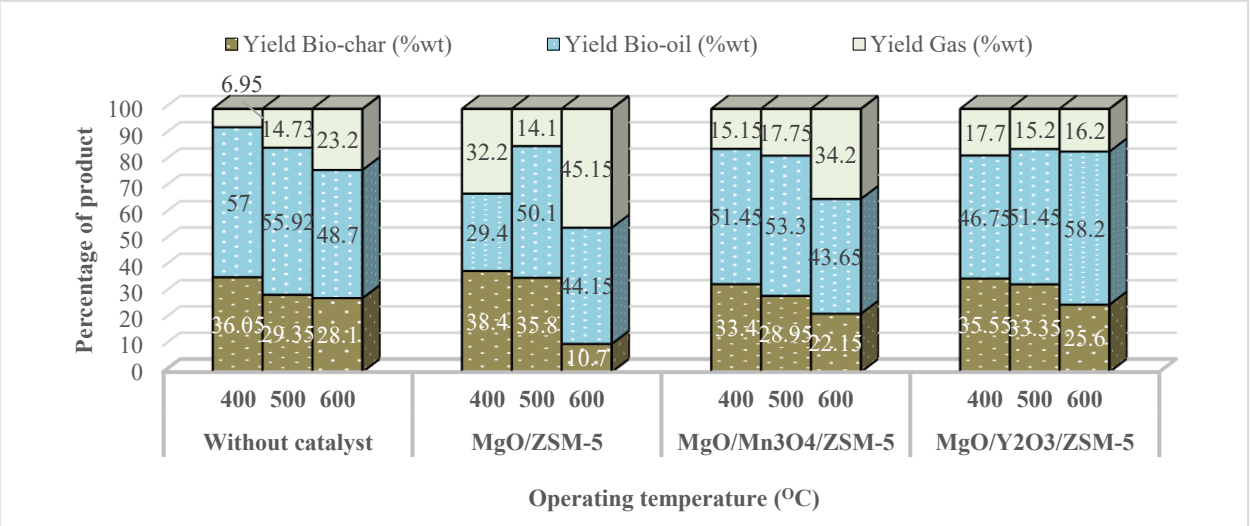

Fig 2. Comparison of bio-char formation with and without magnesium oxide-zeolite based catalysts at various pyrolysis temperatures

According to Jafri et al. (2018), woody biomass, primarily sourced from forestry and trees, exhibits characteristics such as low moisture content, low ash content, high calorific value, and high bulk density (Jafri et al., 2018). Non-woody biomass, such as agricultural products, animal waste, and urban and industrial solid waste, contains a significant amount of moisture, ash content, a lower calorific value, low bulk density, and a higher voidage (Jafri et al., 2018). The moisture content of the material affects the production of bio-char. It increases the amount of energy needed for pyrolysis and hinders the formation of (Kloss et al., 2012; Tripathi et al., 2016). Coconut shell, a woody biomass, was selected for this study. The coconut shell utilized in this experiment exhibits a relatively low moisture content, approximately 11.63%. The coconut shell's low moisture content makes it suitable for use as a fuel in combustion applications, which could potentially enhance productivity (Preeti et al., 2024).

Moreover, the results of pyrolysis include an oil (a mixture of hydrocarbons), synthetic gas (a combination of hydrocarbon gases), and bio-char (Verheijen et al., 2010). Two key variables that can impact the content and synthesis of bio-char are temperature and catalyst activity. Previous studies indicate that higher pyrolysis temperatures result in a decrease in the yield of bio-char formation (Devi, 2014; Mujtaba et al., 2023; Wystalska & Kwarciak-Kozłowska, 2021). In addition, the utilization of various catalysts at different temperatures for the production of bio-char can serve as a fascinating subject for research discovery. The present work employed numerous catalysts, specifically MgO/ZSM-5, MgO/Y_2O_3/ZSM-5, and MgO/Mn_3O_4/ZSM-5, at different temperatures of 400°C, 500°C, and 600°C. The objective was to investigate the impact of magnesium oxide-zeolite based catalysts on the production of bio-char, as depicted in Figure 2.

In general, Figure 2 demonstrates a negative correlation between temperature and bio-char production. This phenomenon arises from the breakdown of heteroatoms at low temperatures, leading to the formation of a dense tars composition (Guedes et al., 2018; Reza et al., 2023). Conversely, the significant breakdown of biomass species at elevated temperatures results in considerable molecular disorder and the formation of a wide range of chemicals (Akhtar & Saidina Amin, 2012; Guedes et al., 2018). At high temperatures (>500°C), both the primary and secondary pyrolysis processes modify the product composition. Enhancing the production of bio-oil and biogas can be achieved by increasing the reaction temperatures to optimize the conversion of char and secondary decomposition (Bianasari et al., 2024). Thus, bio-char is typically produced by biomass pyrolysis, with the maximum production being achieved at low operating temperatures (<500°C).

3.3. Impact of MgO/ZSM-5 Catalyst on Bio-Char Production

The data shown in Figure 2 demonstrates that the utilization of the MgO/ZSM-5 catalyst results in a greater production of bio-char when compared to pyrolysis without a catalyst, even at relatively low operating temperatures of 400°C and 500°C. It is the acidic nature of MgO and ZSM-5, together with the working temperature, that influences the transformation of biomass into end products. As shown in Figure 3, temperature and catalyst characteristics are potential determinants of bio-char generation.

Previous research findings indicate that acid and basic alterations of biomass led to a higher formation of bio-char (Yadav et al., 2019). Moreover, a previous study carried out by Smets et al. (Smets et al., 2013) showed a comparable pattern, where biomass conversion carried out using the ZSM-5 catalyst resulted in a higher production of bio-char. The inherent acidity of this zeolite catalyst determines its character, while the unique pore structure imparts certain attributes. Furthermore, research published by Chellappan et al. (2018) (Chellappan et al., 2018) has shown that an increase in temperature leads to an increase in pH level, which in turn causes a reduction in the quantity of bio-char. Magnesium oxide (MgO) maintains its acidic properties when used as a basic catalyst, as shown in the study conducted by Hata et al. (2021) (Hata et al., 2021). When MgO is mixed with ZSM-5, its acidic nature can cause the combined catalyst to exhibit an acidic character. This acidic condition is advantageous for the production of bio-char, as shown in Figure 3. Hence, the utilization of the MgO/ZSM-5 catalyst at low temperatures can enhance the impact of low temperatures, characterized by a reduced pH, in conjunction with the acidity of MgO and ZSM-5, resulting in a greater production of bio-char compared to pyrolysis without the catalyst.

Nevertheless, the conversion of biomass and bio-char into bio-oil persists as the temperature increases from 400°C to 500°C. Ultimately, the MgO/ZSM-5 catalyst achieves the greatest gas output at a temperature of 600°C. This is due to the

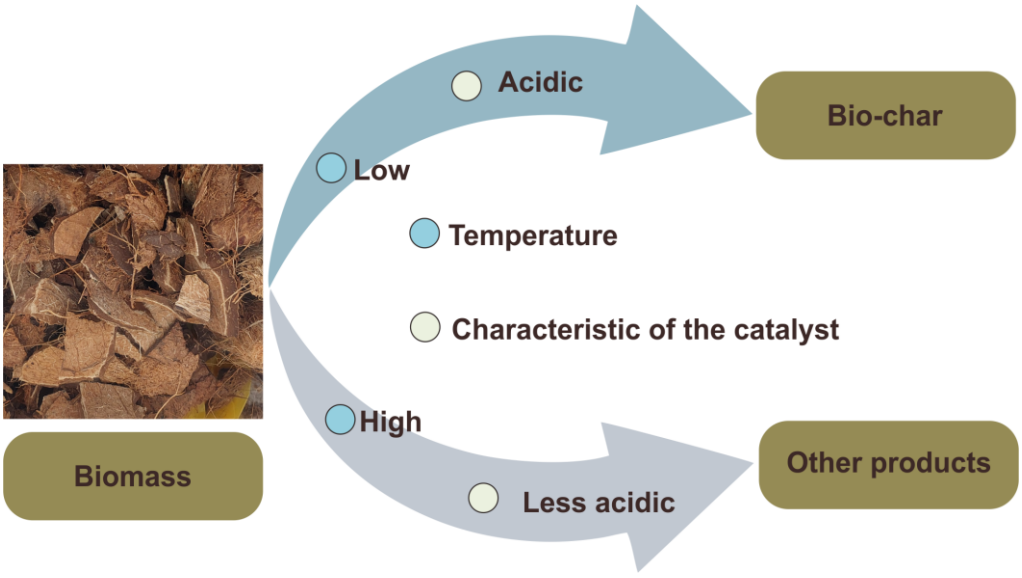

Fig 3. Factors affecting bio-char production in catalytic pyrolysis of biomass: temperature and charateristic of the catalyst

fact that the ZSM-5 catalyst's performance is improved by the addition of MgO, making it capable of effectively completing secondary reactions that transform biomass and bio-char into bio-oil and gas. The presence of metal in MgO has the potential to enhance secondary cracking and decomposition processes (Huang et al., 2015; Ji et al., 2017; Vichaphund et al., 2014). At higher temperatures, the product composition is modified by both the primary and secondary pyrolysis processes, resulting in the production of a greater amount of bio-oil and gas rather than bio-char. In contrast, temperatures exceeding 540°C are significantly elevated and quickly reduce the production of liquid fuel (Zheng et al., 2006). The occurrence is attributed to the phenomena of secondary breakdown of the volatiles at very high temperatures, resulting in an augmented gas generation (Guedes et al., 2018). The study undertaken by Ji et al. also demonstrated a comparable trend. The findings of the catalytic characterization suggested that the inclusion of MgO has a positive effect on the dispersion of NiO in the ZSM-5 (Ji et al., 2017). The observed phenomenon can be attributed to the catalyst's ability to augment secondary cracking and breakdown processes, namely decarbonylation and decarboxylation, of pyrolytic vapors (Huang et al., 2015; Vichaphund et al., 2014). By comparison to the treatment without catalyst, the bio-oil production decreased while the water production increased in the catalytic pyrolysis treatments, as depicted in Figure 2. This phenomenon occurs due to the deoxygenation capability of ZSM-5 zeolites and metal-modified ZSM-5 catalysts, which transfers oxygen into CO_2, CO, and H_2O (K. Wang et al., 2015; H. Zhang et al., 2011). Thus, the catalytic pyrolysis process demonstrated that MgO/ZSM-5 catalysts can improve secondary cracking and decomposition reaction, leading to an increase in the gas product although a drop in the liquid product yield, particularly at elevated temperatures.

3.4. Evaluating the Influence of Catalyst Combinations on Bio-char Production

The objective of this study was to investigate the impact of combining MgO/ZSM-5 with other metal oxides on the generation of bio-char. Figure 2 demonstrates that the utilization of MgO/Mn_3O_4/ZSM-5 results in a considerably lower production of bio-char in comparison to pyrolysis without a catalyst and MgO/Y_2O_3/ZSM-5. This phenomenon is depicted in Figure 3, where the properties of the catalyst, particularly its acidity, might impact the generation of bio-char. MgO/Mn_3O_4/ZSM-5 catalyst is formed by combining magnesium oxide (MgO), manganese oxide (Mn_3O_4), and ZSM-5 zeolite. This combination is specifically engineered to leverage the unique characteristics of each component in order to provide a synergistic impact, therefore augmenting the total catalytic performance. As previously described, the intrinsic properties of ZSM-5 and MgO cause the catalyst to function as an acid catalyst, resulting in the production of bio-char rather than bio-oil and gas at relatively low temperatures. Furthermore, zeolite ZSM-5 is renowned for its well-established acidic characteristics, mostly attributed to the inclusion of silica-alumina groups in its composition (Ghazimoradi et al., 2023; Shi et al., 2023). These groups encompass substantial Brønsted and Lewis acid sites, which have great utility in catalytic processes such as hydrocarbon cracking (Yi et al., 2022). However, the inclusion of Mn_3O_4 will enhance the properties of the catalyst mixture, resulting in a reduced formation of bio-char at low temperatures. The amphoteric behavior of manganese oxide in the form of Mn_3O_4 results in its ability to function as either an acid or a base, contingent upon the reaction conditions (Singh et al., 2020; Zhao et al., 2017). In addition, Mn_3O_4 exhibits redox characteristics, therefore enabling its involvement in oxidation-reduction processes (Gagrani et al., 2019; Kaczmarczyk et al., 2016; Pike et al., 2007). Mn_3O_4 is recognized for its high redox activity, which can accelerate the decomposition of biomass during pyrolysis, resulting in more effective transformation of organic matter into volatile compounds (bio-oil and gases) and, as a result, reduced residual bio-char concentration (de Boer et al., 2024; Gagrani et al., 2019; X. Li et al., 2009; Xiong et al., 2017). The redox properties of Mn_3O_4 enable oxidation-reduction processes that can enhance the breakdown of lignocellulosic organic matter into smaller molecules. Furthermore, manganese oxides with abundant redox chemistry have been extensively employed in (electro)catalysis for energy and environmental applications (Risch et al., 2017).

Separately, Y_2O_3 has weaker redox characteristics than Mn_3O_4, although it can still engage in redox reactions to some extent. This could potentially enhance the decomposition of biomass and its transformation into volatile compounds, hence decreasing the quantity of leftover bio-char. Nonetheless, the

redox activity of this product is typically less significant compared to manganese oxides, which implies that it may not facilitate as thorough biomass breakdown as Mn_3O_4. Furthermore, Y_2O_3 possesses unique properties intrinsic to its acidity. Y_2O_3 had higher Lewis acid strength than Ni when the catalyst was synthesized by the combination of Ni-NiOx-Y_2O_3 (K. Xu et al., 2022). Supplementary investigations have further confirmed that Y_2O_3 exhibits robust Lewis acid characteristics (Bhagat et al., 2017; Dong et al., 2022; Hussein & Gates, 1998; Lin et al., 2013). By operating the Y_2O_3/ZSM-5 catalyst at low temperatures, the impact of low temperatures, characterized by a reduced pH, can be combined with the acidity of MgO/Y_2O_3/ZSM-5 to produce a greater quantity of bio-char. Therefore, the bio-char yield achieved with MgO/Mn_3O_4/ZSM-5 catalysts is lower than that with MgO/ZSM-5 and MgO/Y_2O_3/ZSM-5 catalysts.

3.5. Comparative Analysis of Several Magnesium Oxide-Zeolite Based Catalysts Effect on HHV

The high heating value (HHV) is a crucial properties for bio-chars used as biofuels (Mujtaba et al., 2023). In order to guarantee significant and sustainable changes in the future supply of green clean energy, bio-chars demonstrate good fuel properties and potential (Abdullah et al., 2010; Kwapinski et al., 2010). The HHV of a fuel is the heat produced on complete combustion of a unit quantity of fuel, which includes the latent heat of vaporization of water vapor produced during combustion. High HHV fuel yields greater energy output (L. Xu & Yuan, 2015). The HHV of the fuel can be determined by experimental measurement using an adiabatic oxygen bomb calorimeter. The underlying concept is to precisely compute the difference between the reactant and the product before and after the reaction (Mujtaba et al., 2023). The impact of catalyst formulations on the calorific value of bio-char derived from coconut shell, along with data from literature-reported biomass, is detailed in Table 2.

The data shown in Table 2 clearly indicates that the caloric values of bio-char generated by catalytic pyrolysis were greater than those of coconut shells without any additional processing. Additionally, bio-char often possesses numerous characteristics that render it appropriate as a fuel source. A higher calorific value is a consequence of its higher carbon and hydrogen contents, and lower oxygen levels compared to the initial feedstock (Lee et al., 2013). Moreover, the carbon composition of bio-char can impact the current calorific value. During burning in biomass, carbon and hydrogen serve as the primary energy sources. Due to the high oxygen content in the biomass, its heating value is reduced since oxygen facilitates the full burning of the biomass, which acts as fuel (Y. Zhang et al., 2023). During biomass pyrolysis, combustion occurs, resulting in a rise in carbon content and a decrease in nitrogen, hydrogen, and oxygen contents of the bio-char structure (Sieradzka et al., 2022).

On the other hand, the calorific value of bio-char can be affected by several factors including its composition, processing conditions, and the catalysts employed in its production (Anand et al., 2023). The contribution of catalysts in enhancing the HHV of bio-char is illustrated in Figure 4. When mixed directly with bio-char, the catalyst may have no direct effect on its HHV. After the pyrolysis process, a catalyst added to the bio-char will usually not directly change the bio-char's calorific value. Yet, catalysts can exert an impact on the bio-char synthesis process during the pyrolysis stage. The primary factors influencing the HHV of bio-char are its chemical composition, including carbon, hydrogen, and oxygen content, as well as the structural features formed during pyrolysis (Hu & Wei, 2023; Mujtaba et al., 2023; Zhou et al., 2022). Although direct mixing may modify certain surface characteristics or chemical reactivity, it does not fundamentally affect the inherent energy content of the bio-char. The addition of catalysts during bio-char production process can alter the ash content and fixed carbon content of bio-char (Oladosu et al., 2022).

In this study, raw coconut shell demonstrated the HHV of 12.03 MJ/Kg, which is comparatively lower than other biomass types like dried cassava peel (18.30 MJ/Kg), cassava rhizomes (23.70 MJ/Kg), and palm fiber (16.60 MJ/Kg). Nonetheless, the application of catalytic formulations, specifically MgO/Y_2O_3/ZSM-5 and MgO/ZSM-5, led to significant improvements in the calorific value of the produced bio-chars. In particular, pyrolysis conducted at 500 °C utilizing MgO/Y_2O_3/ZSM-5 resulted in bio-char with a higher heating value of 29.08 MJ/Kg, whereas the combination of MgO/ZSM-5 produced a value of 28.08 MJ/Kg. The increases observed are 17.05 MJ/Kg and 16.05 MJ/Kg, respectively, when compared to the raw coconut shell. Comparable trends were noted in additional research. Egboisuba (2022) documented a rise from 18.30 to 23.77 MJ/Kg with the application of NiNPs in the pyrolysis of dried cassava peel, whereas Selvarajoo and Ooocith (2020) noted an increase from 16.60 to 26.60 MJ/Kg in palm fiber during thermal pyrolysis (Egbosiuba, 2022; Y. Zhang et al., 2023). In contrast, the non-catalytic pyrolysis of cassava

Table 2
Higher heating values (HHVs) of various biomass and bio-char obtained through catalytic pyrolysis with various catalysts at 500°C

Sample	Catalyst	HHV (MJ/Kg)	ΔHHV (MJ/Kg)	ΔHHV (%)	Temperature (°C)	Reference
Coconut shell	-	12.03			-	Present study
Bio-char of coconut shell pyrolysis	ZSM-5	20.06	+8.03	+66.7%	500	
Bio-char of coconut shell pyrolysis	MgO/ZSM-5	28.08	+16.05	+133.4%	500	
Bio-char of coconut shell pyrolysis	MgO/Y_2O_3/ZSM-5	29.08	+17.05	+141.7%	500	
Bio-char of coconut shell pyrolysis	MgO/Mn_3O_4/ZSM-5	26.08	+14.05	+116.8%	500	
Dried cassava peel (DCP)	-	18.30			-	(Egbosiuba, 2022)
Bio-char of DCP pyrolysis	NiNPs	23.77	+5.47	+29.9%	500	
Cassava rhizomes	-	23.7			-	(Rueangsan *et al.*, 2021)
Bio-char of cassava rhizomes pyrolysis	-	24.60	+0.90	+3.8%	500	
Bio-char of cassava rhizomes pyrolysis	Super dolomite	20.2	-3.50	-14.8%	500	
Palm fibre	-	16.6			-	(Selvarajoo and Oochit, 2020)
Bio-char of palm fibre pyrolysis	-	26.60	+10.00	+60.2%	500	

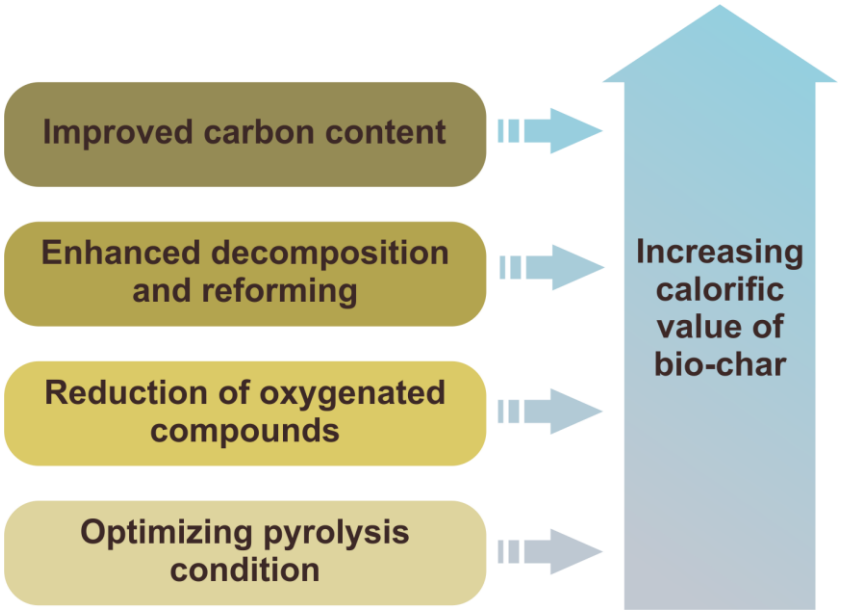

Fig 4. Effect of catalytic interventions on calorific value enhancement during bio-char production

rhizomes produced a modest HHV of 24.60 MJ/Kg bio-char, which is merely 0.90 MJ/Kg above its raw value. Furthermore, the use of Super Dolomite in the pyrolysis of cassava rhizomes led to a reduction in HHV to 20.20 MJ/Kg, suggesting a negative effect on the carbon retention and energy density of the resulting solid residue (Rueangsan et al., 2021).

Moreover, according to Table 2, the bio-char produced by the catalytic pyrolysis procedure utilizing $MgO/Y_2O_3/ZSM-5$ had the maximum calorific. The occurrence happens due to the direct impact of the individual characteristics of metal oxides and zeolites on the characteristics of the catalyst when combined. In a previous experiment carried out by Pütün (Pütün, 2010), it was shown that the inclusion of MgO significantly improved the calorific value. The addition of magnesium oxide to the raw material increased the oil's quality by increasing the calorific value and H/C ratio. Furthermore, MgO effectively reduced the oxygen content of the bio-oil (Stefanidis et al., 2016). The presence of oxygenates in biofuels, especially solid fuel, negatively impacts their heating value by reducing the capacity for oxidation (Demirbas, 2007; Gupta, 2018; Merckel et al., 2019; Pham, 2014; Rahim et al., 2023). Further, the MgO catalysts exhibited performance that was either equivalent to or better than that of the ZSM-5 catalyst often employed in industrial applications. Hence, the MgO/ZSM-5 catalyst can generate bio-char with reduced oxygen content and increased carbon content compared to the ZSM-5 catalyst. This leads to a higher calorific value of 28.08 MJ/Kg in comparison to bio-char produced by pyrolysis using ZSM-5 (20.06 MJ/Kg).

The data in Table 2 indicates that bio-char produced by catalytic pyrolysis with $MgO/Mn_3O_4/ZSM-5$ has lower HHV values compared to MgO/ZSM-5 and $MgO/Y_2O_3/ZSM-5$. This is due to the utilization of a 1:1:2 ratio of metal oxide 1: metal oxide 2: zeolite in this work. Concurrently, the metal oxide/zeolite ratio of MgO/ZSM-5 is 1:1. MgO concentration in MgO/ZSM-5 is higher compared to $MgO/Mn_3O_4/ZSM-5$. Mn_3O_4 exhibits enhanced redox capacity, resulting in increased bio-char production and reduced volatile matter. Nevertheless, MgO has a tendency to enhance the carbon content of bio-char by promoting deoxygenation and establishing a structurally more robust aromatic framework. As depicted in Figure 4, this can enhance the calorific value. The compounds MgO and Y_2O_3 have the capacity to improve decomposition and reforming in addition to their capacity to increase carbon content and reduce oxygenated compounds (J. F. Li et al., 2014; Narasimharao & Al-Sultan, 2020; Pawelczyk et al., 2022; Pelucchi et al., 2021; Santamaria et al., 2020; Y. Wang et al., 2014). Certain metal oxide catalysts can accelerate the breakdown of biomass and facilitate secondary reactions such as cracking and reforming of volatiles. Consequently, this can lead to the production of bio-char characterized by a more aromatic composition, which generally exhibits more energy content. Furthermore, the capacity to decrease the amount of volatile matter and enhance the concentration of fixed carbon leads to an elevated energy content in the bio-char (Hendrix et al., 2024; Tomczyk et al., 2020). Moreover, Y_2O_3 demonstrates outstanding thermal stability, strong chemical resistance, and remarkable mechanical strength, particularly when employed as a reinforcing material in composites, thereby augmenting the stability of the metal oxide surface (Guo et al., 2022; K. Xu et al., 2022). Moreover, according to a prior investigation, the presence of Y_2O_3 on the surface of $0.2Au-La_2O_3$ is the primary determinant for the improvement in the performance of oxidative cracking mechanism (Narasimharao & Al-Sultan, 2020). Hence, the inclusion of Y_2O_3 in $MgO/Y_2O_3/ZSM-5$ results in the generation of the greatest high-hate value (HHV) in bio-char produced by catalytic pyrolysis at moderate temperatures (500°C) when compared to alternative catalysts.

In general, the findings indicate that catalytic pyrolysis, especially utilizing $MgO/Y_2O_3/ZSM-5$ and MgO/ZSM-5, presents a viable method for transforming low-calorific biomass like coconut shell into high-energy-density solid biofuels. This catalytic approach presents an opportunity to enhance the utility of agricultural residues while promoting sustainable energy generation.

3.6. Effects of Temperature and Magnesium Oxide-Zeolite Catalysts on Calorific Value Enhancement

An analysis of the relationship between the employment of magnesium oxide-zeolite combination catalysts and the production of bio-char in biomass pyrolysis at various temperatures might provide valuable information on their

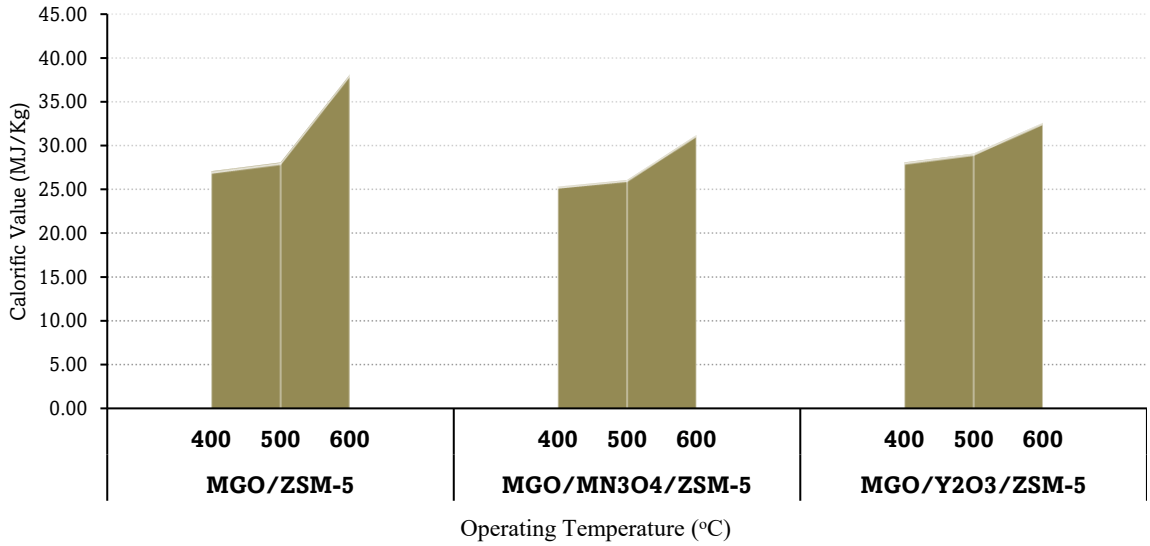

Fig 5. Effects of temperature and catalyst interaction on calorific value of bio-char

appropriateness and enhancement of the process. This investigation is carried out at temperatures of 400°C, 500°C, and 600°C. Figure 5 present the higher heating values (HHVs) or calorific values of bio-char obtained by catalytic pyrolysis using magnesium oxide-zeolite catalysts at different temperatures.

According to Figure 5, bio-chars from catalytic pyrolysis in every catalyst generate an increase in HHVs as the temperature rises. This happens as a result of bio-char's calorific value showing an upward correlation with rising temperatures. Further, the pH, fixed carbon, and specific surface area of bio-char all exhibited an upward trend with increasing temperature (Irfan et al., 2016; Qurat-ul-Ain et al., 2021; H. Zhang et al., 2017). Additionally, higher temperatures lead to the breakdown of cellulose and hemicellulose, as a consequence of which there is a higher concentration of pure carbon in the final product ((Majedi et al., 2016; Wardani et al., 2018). Nevertheless, the MgO/Y_2O_3/ZSM-5 catalyst can generate bio-char with a somewhat greater HHV value compared to alternative catalysts, particularly at temperatures of 400°C and 500°C. The stability of the complex catalyst can be attributed to the presence of Y_2O_3 (Cho et al., 2019; De Luca et al., 2021). In addition to that, Y_2O_3 can coordinate with MgO to enhance carbon concentrations and decrease oxygen concentrations in bio-char by promoting deoxygenation (Chueaphetr et al., 2023; Elangovan et al., 2017, 2018; Narasimharao & Al-Sultan, 2020; Stefanidis et al., 2016; Y. Zhang et al., 2023). At a relatively high temperature of 600°C, bio-char produced by catalytic pyrolysis with the MgO/ZSM-5 catalyst exhibits the highest HHV value, surpassing that achieved with MgO/Y_2O_3/ZSM-5. The prior section elucidated that the proportion of MgO in MgO/ZSM-5 exceeds that of MgO in MgO/Y_2O_3/ZSM-5 in this investigation. Therefore, the MgO character is more prominent in MgO/ZSM-5 compared to MgO/Y_2O_3/ZSM-5. MgO exhibits superior specific heat capacity and thermal conductivity in comparison to Y_2O_3 (Harris et al., 2013). By virtue of its superior specific heat, MgO demonstrates superior resistance to combustion on the catalyst and retains stability at high temperatures when compared to Y_2O_3. Furthermore, thermal conductivity plays a vital role in thermal interface materials as they must efficiently enable the transfer of heat between two surfaces (Chung, 2022). This study indicates that MgO/ZSM-5 exhibits greater heat transfer capacity to the feedstock and bio-char generated in comparison to Y_2O_3/ZSM-5 during the process of pyrolysis. Thus, the higher calorific value of bio-char produced using MgO/ZSM-5 compared to bio-char obtained using MgO/Y_2O_3/ZSM-5 and MgO/Mn_3O_4/ZSM-5 can be attributed to the superior thermal stability and temperature transmission capabilities of MgO/ZSM-5 in reducing oxygen content and producing a larger quantity of carbon.

4. Conclusion

This study demonstrates the significant benefits of using MgO-zeolite based catalysts in biomass catalytic pyrolysis for producing high-calorific-value bio-char. The combination of MgO with ZSM-5, Y_2O_3/ZSM-5, and Mn_3O_4/ZSM-5 shows promise in further enhancing the energy content of bio-char. Multiple findings are presented in this investigation. Employing magnesium oxide-zeolite-based catalysts (MgO/ZSM-5, MgO/Y_2O_3/ZSM-5, and MgO/Mn_3O_4/ZSM-5) at different temperatures reveals that the choice of catalyst can influence both the quantity and quality of bio-char. Typically, conducting pyrolysis at temperatures under 500°C appears to yield higher amounts of bio-char, while elevated temperatures tend to enhance the production of bio-oil and biogas. Furthermore, the bio-char production of MgO/Mn3O4/ZSM-5 is relatively lower when compared to the MgO/ZSM-5 and MgO/Y2O3/ZSM-5 catalysts, as well as in pyrolysis without any catalyst. The specific characteristics of the metal oxides and their interactions with ZSM-5 zeolite have a substantial impact on the catalytic performance. In addition, the utilization of MgO/Mn_3O_4/ZSM-5 led to a reduced HHV in comparison to MgO/ZSM-5 and MgO/Y_2O_3/ZSM-5. This is due to the robust redox characteristics of Mn_3O_4, which enhance the synthesis of bio-char but decrease its volatile content and, as a result, its calorific value. The combination of MgO and Y_2O_3 simultaneously improves the breakdown and reformation mechanisms, therefore facilitating the production of bio-char with increased energy content. Furthermore, both MgO/Y_2O_3/ZSM-5 and MgO/ZSM-5 catalysts are efficient in increasing the HHV of bio-char. The selection between them should take into account the desired operating temperature. MgO/Y_2O_3/ZSM-5 combined-catalyst is better suited for mild temperatures, but

MgO/ZSM-5 catalyst performs superiorly at higher temperatures because of its improved thermal characteristics and heat transfer capabilities. This study emphasizes the possibility of using tailored catalyst formulations to enhance biomass conversion processes for the purpose of sustainable energy generation.

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