Original Research Article

Catalytic Steam Gasification of Laminated Kraft Paper Packaging Waste for Hydrogen-Enriched Syngas over $NiO/La_2O_3/Al_2O_3$ Catalyst

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ABSTRACT

Laminated kraft paper (LKP) packaging waste and mixed plastic waste (MPW) are substantial contributors to landfill waste due to their heterogeneity, which limits the economic viability of conventional recycling. Furthermore, insufficient research has comprehensively explored the effects of catalyst loading on the steam gasification of composite waste. This study is the first systematic evaluation of the catalytic steam gasification of LKP packaging waste and its co-gasification with MPW using varied loadings of a $NiO/La_2O_3/Al_2O_3$ catalyst in a downdraft fixed-bed reactor at 800 °C. The effects of catalyst loading (0-2 g, C/F = 0-0.4 wt/wt) and plastic co-feeding (20 wt%) on syngas composition and hydrogen yield were evaluated. Under non-catalytic conditions, LKP gasification produced 1.64 L g^{-1} of hydrogen with an H_2/CO ratio of approximately 3.3. Catalyst addition significantly enhanced reforming and water–gas shift (WGS) reactions. At C/F = 0.2, hydrogen yield increased to 2.22 L g^{-1}, while at C/F = 0.4 it reached 2.42 L g^{-1}. The H_2/CO ratio increased to ~5.0 under catalytic conditions, indicating improved hydrogen selectivity and CO conversion. Light hydrocarbons (C_2–C_3) decreased substantially under catalytic conditions, demonstrating effective hydrocarbon cracking and reforming activity. However, increasing catalyst loading beyond 1 g did not significantly improve performance, indicating kinetic saturation. Co-gasification with 20 wt% MPW at C/F = 0.4 resulted in the highest hydrogen yield (2.56 L g^{-1}) without significant change in H_2/CO ratio. These findings demonstrate effective catalytic upgrading of laminated composite waste into hydrogen-rich syngas using Ni-based catalysis.

Keywords: Gasification; Laminated Kraft Paper Packaging Waste; Heterogeneous; Catalyst; Syngas; Hydrogen

INTRODUCTION

Thailand has experienced continuous industrial expansion over the past decade, driven by its strong petrochemical industry, the largest in ASEAN (1). The parallel increase in industrial waste generation has primarily been mitigated with landfilling and open dumping, which account for approximately 70-80% of Thailand's waste management practices (2). Heterogeneous wastes, namely laminated kraft paper (LKP) packaging, widely used for food packaging and the storage of bulk chemical powders, and mixed plastic

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waste (MPW) remain difficult to recycle due to their complex compositions and are therefore commonly disposed of in landfills (3). Landfills ultimately lead to severe environmental consequences, including ecosystem degradation and risks to public health. Consequently, alternative waste management and valorization approaches are necessary to effectively manage these wastes.

Among available technologies, gasification has attracted considerable attention due to its ability to convert carbon-containing materials into synthesis gas (syngas). Steam has been extensively preferred as a gasifying agent because it enhances hydrogen production through steam reforming and water–gas shift (WGS) reactions (4). Fixed-bed downdraft gasifiers provide stable operation and effective tar cracking under controlled conditions, making them suitable for laboratory-scale mechanistic studies. Parameters such as temperature, steam ratio and feedstock proximate composition significantly affect gas yield and hydrogen selectivity (5). Catalytic enhancement, particularly using Ni-based catalysts, is widely employed to reduce tar formation, an undesirable by-product of gasification, while improving syngas quality and hydrogen production efficiency (6, 7).

Despite extensive research on biomass gasification and plastic co-gasification, LKP packaging waste contaminated with industrial pigments and chemical residues has received limited attention. The multilayer structure and inorganic contaminants may significantly affect catalytic interactions and gasification behavior.

La_2O_3 promoters have been reported to enhance catalyst performance by improving the resistance of NiO/La_2O_3/Al_2O_3 catalysts to sintering and coke formation while maintaining catalytic activity (8, 9). Despite these findings, the effects of NiO/La_2O_3/Al_2O_3 catalyst loading during steam gasification of laminated composite waste on hydrogen yield and H_2/CO ratio have not been systematically investigated.

Accordingly, this study examines the catalytic steam gasification of LKP and its co-gasification with MPW in a downdraft fixed-bed reactor at 800 °C. The effects of catalyst loading (0-2 g; C/F = 0-0.4 wt/wt) on gas composition, hydrogen yield, and H_2/CO ratio are systematically evaluated. In addition, the influence of plastic co-feeding (20 wt%) on syngas quality is examined. The findings aim to analytically elucidate the role of catalytic enhancement in improving hydrogen-rich syngas production from laminated composite industrial waste.

MATERIALS AND METHODS

Feedstock Collection and Preparation

LKP packaging waste and MPW were obtained from a plastic compounding factory in Thailand. The LKP consisted of multilayer kraft paper laminated with polypropylene (PP) and polyethylene (PE) films and was contaminated with industrial pigments and additives. The MPW comprised heterogeneous polymer blends, including PP, PE, acrylonitrile butadiene styrene (ABS), polyoxymethylene (POM), and high-impact polystyrene (HIPS), containing various additives and exhibiting different physicochemical properties (10).

The LKP waste was manually cut into small fragments and subsequently shredded to obtain uniform particles. The shredded materials were homogenized and compressed into cylindrical pellets (diameter: 1 cm; height: 1 cm; mass: approximately 0.5 g per pellet) using a laboratory hydraulic press to ensure consistent feeding and reaction behavior.

For co-gasification experiments, MPW pellets were blended with LKP at a mass ratio of 80:20 (wt%). The mixture was homogenized prior to compression into cylindrical specimens under identical pressing conditions.

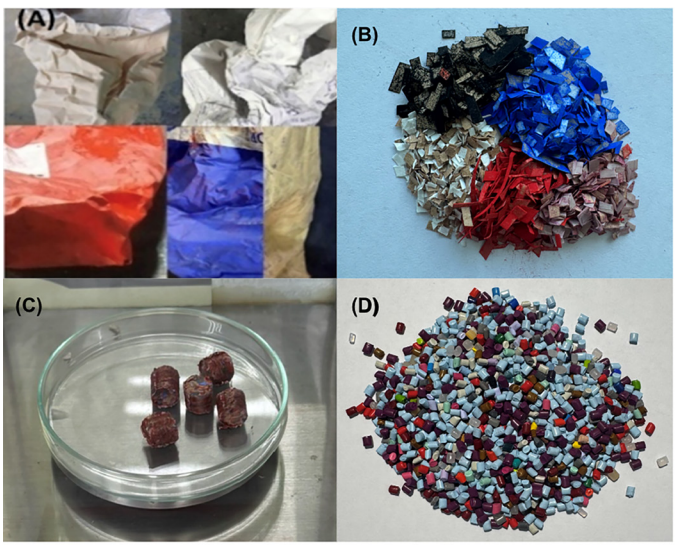

Figure 1. *Sample preparation procedure: (A) Collection of laminated kraft paper bags of pigments and chemicals (LKP); (B) Cut and shredded pieces of LKP; (C) Compression into standardized cylindrical LKP specimens; (D) MPW in pellet form prior to mixing and compression in preparation for mixed waste (80 wt.% LKP + 20 wt.% MPW).*

Feedstock Characterization

Proximate analysis (moisture, volatile matter, fixed carbon, and ash content) and ultimate analysis (C, H, N, S, Cl, and O content) were performed on a dry basis using standard analytical procedures. These analyses were conducted to evaluate fuel properties relevant to thermochemical conversion, including the H/C and O/C atomic ratios.

Catalyst Preparation

A commercial $NiO/La_2O_3/Al_2O_3$ catalyst (Sichuan Shutai Chemical Technology Co., Ltd.) was used in this study. The catalyst composition consisted of 18.6 wt% NiO, 3.2 wt% La_2O_3, 75.9 wt% Al_2O_3, and 2.3 wt% loss on ignition (LOI). The catalyst was crushed and sieved to obtain particles with a size range 150-212 µm to minimize mass transfer limitations.

Catalyst loading was varied to achieve catalyst-to-feedstock (C/F) mass ratios of 0, 0.2, and 0.4 (wt/wt), corresponding to 0 g, 1 g, and 2 g catalyst per experimental run, respectively. For co-gasification experiments involving LKP and MPW, the C/F ratio was fixed at 0.4 (wt/wt).

Steam Gasification Experimental Setup

Steam gasification experiments were conducted using a laboratory-scale fixed-bed downdraft gasifier (Figure 2.) equipped with: 1) a steam generator, 2) a fuel feeder, 3) a furnace chamber containing a vertical quartz tubular reactor, and 4) a tar trapping system. The quartz reactor had an outer diameter of 18 mm, an inner diameter of 14 mm, a total length of 500 mm, and heating zone length of 150 mm.

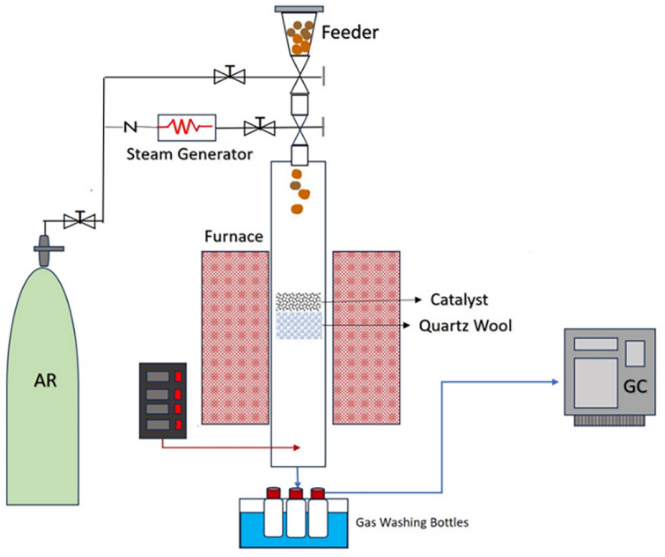

Figure 2. *Schematic diagram of a downdraft fixed bed gasifier.*

Prior to each experiment, the system was leak-tested using a gas leak detection solution. The reactor was heated to 800 °C under argon flow rate of 100 mL min^{-1}. Steam was generated at 300 °C with a water feed rate of 0.1 mL min^{-1} and continuously introduced into the reactor.

For each experimental run, ten cylindrical pellets (0.5 g each) were sequentially fed into the reactor, corresponding to approximately 5 g of total feedstock. All experiments were conducted under identical operating conditions and continued until no further gas evolution was observed and only minimal char residue remained. Tar yield was not quantified as the study focuses primarily on syngas yield and composition.

Gas Collection and Analysis

Product gases were collected in 5 L gas sampling bags. Gas composition was analysed using a Shimadzu GC-2014 gas chromatograph equipped with Porapak N, Porapak Q, and molecular sieve 13X columns. Hydrogen (H_2), carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4), ethane (C_2H_6), ethylene (C_2H_4), and propane (C_3H_8) were quantified. Each experiment was conducted in duplicate, and the average values and error bars were reported.

Calculation of Gas Yield and H_2/CO Ratio

The yield of each gas component (Yi, L g^{-1}) was calculated using the following equation:

$$Yi = Vi / m$$

where Vi represents the total collected volume of gas species i (L), and m represents the total mass of feedstock (g).

Gas composition (%) was determined based on GC analysis. The H_2/CO ratio was calculated from the volumetric composition of the product gas. The percentage change in gas yield due to catalyst addition was calculated relative to the non-catalytic condition. Similarly, the effect of plastic co-feeding was evaluated relative to LKP-only gasification under identical catalytic conditions.

RESULTS AND DISCUSSIONS

Feedstock Characterization

Proximate and ultimate analyses (Table 1) provide insight into the observed gasification behavior and syngas composition. Both LKP and MPW exhibited high volatile matter contents (>80 wt%, dry basis), indicating strong devolatilization during thermal conversion. The high volatile fraction promoted the formation of gaseous intermediates and hydrocarbons, which subsequently underwent steam reforming and cracking reactions, thereby contributing to enhanced gas and hydrogen production (11).

LKP contained higher oxygen and fixed carbon contents than MPW. This has likely facilitated the formation of oxygen-containing gaseous intermediates during thermal conversion, thereby favoring WGS reactions and contributing to the comparatively high H_2/CO ratio observed during LKP gasification (Figure 6). Additionally, the higher fixed carbon content may have promoted char-steam gasification reactions ($C + H_2O \rightleftharpoons CO + H_2$), further enhancing hydrogen production (12).

In contrast, MPW exhibited higher carbon and hydrogen contents with extremely low oxygen content, resulting in a lower O/C ratio and a more hydrocarbon-rich composition. Consequently, co-feeding MPW promoted the formation of light hydrocarbons, particularly C_2H_4 and C_3H_8, while suppressing CO_2 formation, as shown in Figure 5. The low nitrogen, sulfur, and chlorine contents in both feedstocks minimize the risk of harmful emissions and catalyst poisoning, supporting their suitability for catalytic steam gasification.

Gas Composition

Steam gasification involves a complex network of heterogeneous and homogeneous reactions, including char gasification, steam reforming of hydrocarbons, and water–gas shift (WGS) reactions. The principal hydrogen-forming reactions are as follows:

$$C + H_2O \rightleftarrows CO + H_2$$
$$CH_4 + H_2O \rightleftarrows CO + 3H_2$$
$$CO + H_2O \rightleftarrows CO_2 + H_2$$

Table 1. *Proximate and ultimate analysis of LKP and MPW waste measured on dry basis.*

Characteristics	LKP	Method	MPW	Method
Proximate Analysis (wt%, dry basis)				
Moisture content	0	ISO 18134-1 and 3	0.00	ASTM E790-15
Volatile Matter	85.6	ISO 18123	82.89	ASTM E897-88
Ash Content	4.34	ISO 18122	13.75	ASTM E830-87
Fixed Carbon	10.06	Calculation	3.36	Calculation
Ultimate Analysis (wt%, dry basis)				
Carbon (C)	44.22	ASTM D 5373-16	75.57	ISO 16948
Hydrogen (H)	5.95	ASTM D 5373-16	10.38	ISO16948
Nitrogen (N)	0.53	ASTM D 5373-16	0.23	ISO 16948
Sulfiu (S)	0.20	ASTM D 4239-17	0.06	ASTM E775-15, B
Chlorine (Cl)	0.79	ASTM D 6271-01	<0.01	ASTM E776-16 (mod.)
Oxygen (O)	43.96	Calculation	0.01	Calculation

Note: The wt% of fixed carbon was calculated on a dry basis as 100 – (volatile matter + ash content). The wt% of oxygen for LKP and MPW was calculated by difference as 100 – (C + H + N + S + Cl + ash content) on a dry basis. Due to the distinct physical and chemical compositions of the two feedstocks proximate and ultimate analyses were conducted by two accredited laboratories, each applying standardized testing frameworks appropriate for its respective material class. LKP was analysed at the KU Biomass laboratory, using standards for solid biofuels, while MPW was analysed at Intertek Testing Services (Thailand) Ltd using frameworks for refuse-derived fuels and recovered plastics.

The product gas consisted primarily of H_2, CO, and CO_2, with minor amounts of CH_4 and C_2–C_3 hydrocarbons (Figure 3A and 3B). Under non-catalytic conditions, LKP gasification produced 1.64 L g^{-1} of hydrogen, representing 55 vol% of the total gas, with an H_2/CO ratio of approximately 3.3. This ratio suggests WGS reactions promoted by the presence of steam at 800 °C.

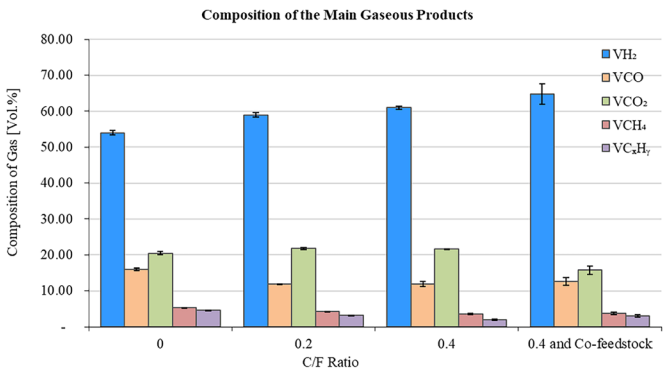

Figure 3A. *Composition (vol. %) of the main gas species obtained from steam gasification of LKP under different catalyst loadings and co-feeding with MPW. Data points represent the mean values, and error bars represent the range from duplicate experiments (n=2). VC_xH_y and VC_xH_y are the sum of vol. % of C_2H_6, C_2H_4 and C_3H_8.*

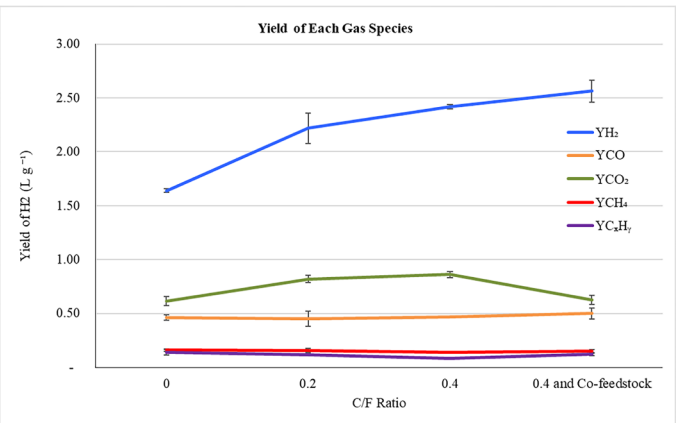

Figure 3B. *Yield (L g^{-1}) of the main gaseous products obtained from steam gasification of LKP under different catalyst loadings and co-feeding with MPW. Data points represent the mean values, and error bars represent the range from duplicate experiments (n=2). YC_xH_y and YC_xH_y are the sum of yield of C_2H_6, C_2H_4 and C_3H_8.*

Under catalytic conditions at C/F ratios of 0.2 and 0.4 (wt/wt), the hydrogen yield increased to 59 and 62 vol% of the total gas, respectively, while the H_2/CO ratio increased to approximately 5.0. Co-gasification with 20 wt% MPW at a C/F ratio of 0.4 further increased the hydrogen yield to 65 vol% without a significant change in the H_2/CO ratio.

Effect of $NiO/La_2O_3/Al_2O_3$ Catalyst

As no catalyst characterization was performed in the present study, the mechanistic interpretations presented herein are inferred from and supported by previously reported findings (7, 9, 13, 14).

Figure 4 illustrates the relative changes in gaseous product yields obtained using the $NiO/La_2O_3/Al_2O_3$ catalyst at C/F ratios of 0.2 and 0.4. At C/F = 0.2 (1 g catalyst), hydrogen yield increased by approximately 35–40% compared to the non-catalytic condition. Concurrently, CO_2 production increased by roughly 35–38%, while CO showed a slight decrease. This trend indicates enhanced water–gas shift (WGS) activity and improved reforming during gasification. Similar catalytic enhancement of hydrogen production over Ni/Al_2O_3 systems during steam gasification has been widely reported (7, 13).

Nickel active sites facilitate C–H bond cleavage and steam reforming of methane and light hydrocarbons, generating additional CO and H_2. The simultaneous increase in H_2 and CO_2 suggests that CO produced during primary devolatilization and char gasification was subsequently converted via the WGS reaction ($CO + H_2O \rightleftharpoons CO_2 + H_2$). The decrease in CO concentration is therefore consistent with catalytic promotion of this equilibrium pathway (7).

La_2O_3, a rare earth oxide, was used as a promoter in the Ni/Al_2O_3 catalyst system to enhance catalytic activity, particularly by strengthening the metal-support interaction and suppressing carbon deposition (9). The strong Ni-La interaction may modify the electronic structure of the active sites, thereby inhibiting carbon nucleation. Figure 4 suggests enhanced coke suppression behavior at a C/F ratio of 0.4, where notable reductions in CH_4 and light hydrocarbons were observed alongside increased H_2 and CO_2 production compared to the C/F ratio of 0.2.

At higher catalyst loading (C/F = 0.4; 2 g), hydrogen enhancement approached approximately 45–50%, while CO_2 production remained elevated. In contrast, light hydrocarbons (C_2H_6, C_2H_4, and C_3H_8) exhibited significant reductions of approximately 20-40%. This

decline confirms enhanced steam reforming and hydrocarbon cracking activity at a higher available active metal surface area. The reduction in CH_4 yield further supports steam methane reforming ($CH_4 + H_2O \rightleftharpoons CO + 3H_2$), followed by secondary WGS conversion (7).

However, increasing catalyst loading from 1 g to 2 g did not proportionally increase hydrogen production. The plateau in performance suggests kinetic saturation or mass transfer limitations within the fixed-bed configuration. Additionally, higher catalyst loading may promote localized heat accumulation and potential sintering, thereby reducing effective active surface area. This observation is consistent with previous studies reporting an optimal catalyst loading beyond which further improvement diminishes (14).

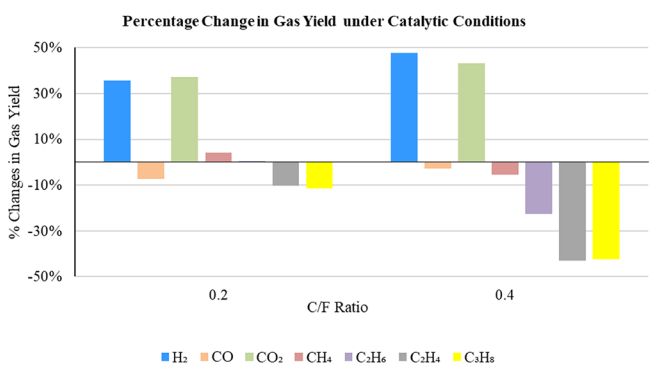

Figure 4. *Effect of increasing catalyst-to-feedstock (C/F) ratio as percentage change in gas yield from LKP steam gasification, compared to the non-catalytic baseline. Values represent the difference in the average yield of each gas species.*

Effect of MPW Co-Feeding

Co-gasification of LKP with 20 wt% MPW under catalytic conditions (C/F ratio = 0.4) resulted in the highest hydrogen yield (2.56 L g^{-1}). Notably, CO_2 production decreased, while C_2–C_3 hydrocarbon yields increased.

The reduction in CO_2 can be attributed to the lower intrinsic oxygen content of plastics, which limits oxygen availability for oxidation pathways during gasification. Simultaneously, the elevated hydrocarbon yield suggests incomplete reforming of plastic-derived volatiles. The presence of hydrocarbon intermediates indicates that steam reforming reactions may be kinetically limited under the applied steam-to-feed ratio.

Although hydrogen yield increased only modestly compared to catalytic LKP-only gasification, the overall syngas quality improved due to lower CO_2 concentration and higher calorific-value gaseous components. Bakhtiar *et al.* similarly reported that the addition of plastics during steam gasification of woody biomass enhances syngas quality. The co-feeding strategy modifies the H/C and O/C balance of the feedstock, thereby influencing product distribution (15).

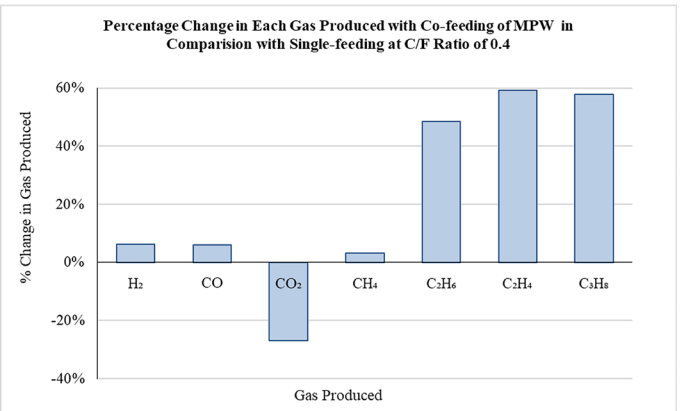

Figure 5. *Effect of 20 wt% MPW co-feeding as percentage change in gas produced under catalytic steam gasification of LKP, relative to single-feeding of LKP at a constant C/F ratio of 0.4. Values represent the relative difference in the output of each gas component.*

H_2/CO Ratio

Figure 6 shows the variation of the H_2/CO ratio under different catalytic and co-feed conditions. In the absence of catalyst (C/F = 0), LKP gasification produced an H_2/CO ratio of approximately 3.3. This value is consistent with reported ranges for non-catalytic steam gasification of biomass, typically between 1.8 and 3.5 under comparable steam conditions (16). The moderate ratio reflects partial steam reforming and limited water-gas shift (WGS) activity at an elevated temperature.

The introduction of $NiO/La_2O_3/Al_2O_3$ catalyst significantly increased the H_2/CO ratio to nearly 5.0 at C/F = 0.2, indicating enhanced reforming and WGS reactions. Nickel active sites promote methane and light hydrocarbon steam reforming, generating additional CO and H_2, while the WGS reaction further converts CO

into H_2 and CO_2. The concurrent increase in H_2 yield and reduction in CO concentration confirm the catalytic promotion of these pathways (7,13).

Increasing catalyst loading from 1 g (C/F = 0.2) to 2 g (C/F = 0.4) did not substantially increase the H_2/CO ratio, which stabilized at approximately 5.0–5.1. The limited additional enhancement at higher catalyst loading suggests that the system may have approached an equilibrium-limited state under the investigated steam gasification conditions at 800 °C (14). Similarly, Artetxe *et al*. reported comparable behavior during steam reforming of phenol, used as a biomass tar model compound, over a Ni/Al_2O_3 catalyst. Increasing catalyst mass from 1.5 g to 2 g at 750 °C resulted in relatively constant carbon conversion and H_2 potential (14).

Co-feeding 20 wt% MPW at C/F = 0.4 did not significantly alter the H_2/CO ratio compared to catalytic LKP gasification alone. Although plastics contribute hydrogen-rich volatiles, the overall ratio remained largely equilibrium-controlled. The limited variation suggests that hydrogen formation was constrained by steam availability and WGS equilibrium rather than hydrocarbon availability alone. Comparable behavior has been observed in biomass–plastic co-gasification systems, where syngas composition remained relatively stable despite increased hydrocarbon input (16).

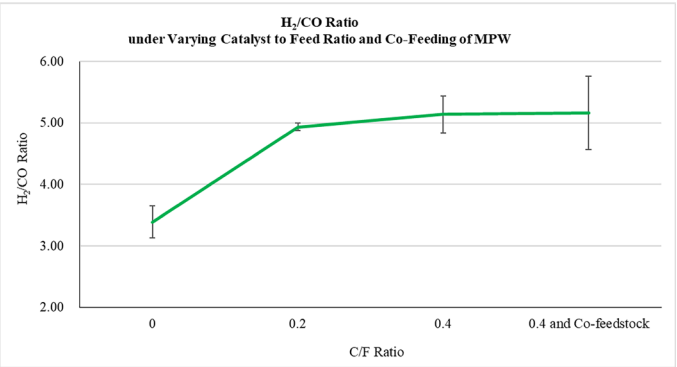

Figure 6. *Effect of catalyst loading and 20 wt% MPW co-feeding on H_2/CO ratio during steam gasification of LKP: C/F ratio of 0, 0.2 and 0.4 (wt/wt) and LKP+MPW with C/F ratio of 0.4 (wt/wt). Data points represent the average values determined from H_2 and CO volumetric compositions in each gasification condition. Error bars represent the range of duplicate measurements (n=2). The relatively wide error bar observed for the C/F=0.4 and co-feedstock condition is attributed to the inherent heterogeneity of the MPW.*

CONCLUSION

This study investigated the steam gasification of laminated kraft paper (LKP) packaging waste and its co-gasification with mixed plastic waste (MPW) using varied loadings of a $NiO/La_2O_3/Al_2O_3$ catalyst in a downdraft fixed-bed reactor at 800 °C. Under non-catalytic conditions, LKP gasification produced 1.64 L g^{-1} of hydrogen with a H_2/CO ratio of approximately 3.3. The introduction of the catalyst significantly enhanced hydrogen production and increased the H_2/CO ratio to approximately 5.0, confirming the promotion of steam reforming and water–gas shift reactions. Concurrently, C_2–C_3 hydrocarbons yields decreased, indicating enhanced cracking and reforming of light hydrocarbon species. Increasing catalyst loading from C/F = 0.2 to 0.4 resulted in further hydrogen enhancement; however, the incremental improvement was limited. This suggests that the system approached an equilibrium-limited state under the investigated gasification conditions at 800 °C, where syngas composition became less dependent on catalyst quantity. Co-feeding 20 wt% MPW increased hydrogen production to 2.57 L g^{-1}, while the H_2/CO ratio remained relatively unchanged. This observation indicates that syngas composition was governed predominantly by reaction equilibrium rather than feedstock hydrogen content. Overall, the $NiO/La_2O_3/Al_2O_3$ catalyst demonstrates effective performance for converting laminated composite waste into hydrogen-rich syngas. Further optimization of catalyst loading and operating conditions may improve process efficiency and syngas quality.

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CONFLICT OF INTEREST

The author declares that there are no conflicts of interest related to this work.

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