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Catalytic Valorization Of Non-Recyclable Municipal Plastic Waste Into Hydrogen-Rich Syngas Using Ni–Mo/Ceo$_2$ Catalysts: A Sustainable Approach For Waste-To-Energy Conversion

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Abstract

Plastic waste in municipalities is a persistent environmental issue due to its poor biodegradability and diverse composition, and requires innovative approaches to sustainable valorization. This would be doubly beneficial, as such waste can be converted to syngas, providing both energy and reducing pollution. However, conventional thermal processes often exhibit poor gas yields, high tar content, and inadequate selectivity. This study aims to address the above limitations by developing a two-stage catalytic thermochemical process utilizing a Ni-Mo/CeO2 catalyst to enhance the production of syngas from municipal plastic waste. The process used is the initial pyrolysis of mixed plastic waste, catalytic steam reforming at high pressures (25bar) and high temperatures (250 °C, 275 °C, and 300 °C). Ni-Mo/CeO$_2$ catalyst also enhanced the syngas yield significantly where H$_2$ content was raised to 54.7 vol% H$_2$ and 28.5 vol% CO with H$_2$ /CO of 1.92 at optimized conditions. The gas yield was maximized to 85.6%, and char and condensate were minimized. The increased redox and dispersion properties of the catalyst, as well as the suppression of coke, were cited as the reasons behind its performance. These results demonstrate a practical route towards the circular use of non-recyclable plastics. The paper highlights the potential of the catalyst for industrial-scale use.

Keywords: *Municipal plastic waste; Syngas; Catalytic reforming; Nickel–molybdenum catalyst; Ceria support; Waste-to-energy*

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1. INTRODUCTION

Municipal solid waste emphasizes the importance of waste segregation and its environmental implications. Municipal wastes are generally divided into organic or wet waste (such as food and garden waste), dry waste (such as plastics, paper, metal and glass), hazardous waste (such as batteries and chemicals), biomedical waste (such as syringes and expired medicines) and sanitary waste (such as diapers and sanitary napkins). Most notable of these is the plastic waste, whose degradation is extremely slow, and it is widespread in the urban waste stream.

Effective management of plastic waste begins with collection at the source, where segregation is achieved through the use of colored bins. Usually, green bins are used to collect wet waste, and blue bins are used to collect dry waste. The plastics are then sorted either manually or mechanically, washed, and pressed or baled to make storage and transportation more convenient. The inability to separate plastic waste has serious environmental effects, including air and soil pollution, blocked waterways, and greenhouse gas emissions. Additionally, improper disposal is detrimental to the health of wildlife and the surrounding population. With the help of systematic segregation and creating awareness among the population, municipalities can transform plastic waste into a resource that can be recycled, reused, or converted into energy, thereby contributing to a more ecologically friendly urban environment.

The daily generation of municipal solid waste (MSW) in India exceeds 160,000 tonnes, and this amount is expected to rise by almost 70% in urban centers due to population growth and increased consumption. Plastic waste, which accounts for about 8-10 % of the total MSW, comprises PET bottles, carry bags, and multilayer films, all of which are highly hazardous to the environment since they are not biodegradable. The paper will address the problem by upgrading plastic waste to hydrogen-rich syngas through thermochemical processes, including pyrolysis, co-gasification, and catalytic steam reforming. Hydrogen and syngas yields of up to 83% and over 75%, respectively, have been achieved using specially designed catalysts. The technique can support the objectives of a circular economy and offer a modular waste-to-energy approach. Municipal liquid wastes have been explored for their synergistic potential in hydrogen production when integrated with catalytic pyrolysis processes such as PANI–Ni systems, which promote deoxygenation and cracking in algae biomass [1]. This line of research aligns with the broader goals of smart city waste management, where thermochemical conversion technologies have been proposed to support circularity and reduce reliance on landfills [2]. Integrated site selection strategies, utilizing methods such as FAHP and FTOPSIS, have further highlighted the importance of optimal placement of waste facilities in contributing to sustainability goals [3]. Concurrently, fuzzy-based MCDM models have enabled the prioritization of sustainable MSW practices based on environmental and socioeconomic indicators [4]. Decentralized community participation models, such as the Swachh Hyderabad Mission, emphasize localized segregation and treatment [5]. However, poor segregation and lack of treatment infrastructure often lead to uncontrolled methane emissions, as shown in the case of unmanaged landfills in Chennai [6]. Emerging technologies that incorporate algae, plastic, and wastewater valorization have demonstrated potential for efficient hydrogen production with a reduced carbon footprint [7]. In particular, Ni-based catalysts have demonstrated high efficiency in the steam reforming of plastics by enhancing the cleavage of C–C and C–H bonds, optimizing thermal performance, and coke resistance [8,9]. Several studies have explored catalytic pyrolysis and reforming as feasible routes for hydrogen generation. The use of Ni-based and bimetallic catalysts not only improves H_2 and CO yields but also optimizes reaction temperatures and selectivity, facilitating the commercialization of WtE processes [10,11]. For instance, hybrid Ni–CaO–Ca_2SiO_4 catalysts in pyrolysis-gasification systems achieved hydrogen purities up to 54.2 vol% and syngas yields of 90.63 mmol/g, thanks to enhanced water–gas shift activity and in-situ CO_2 capture [13]. Moreover, Ni catalysts in CO_2-mediated pyrolysis significantly enhanced syngas generation efficiency, with multi-stage systems amplifying yields by over 14 times [14].

Advanced pyrolysis systems such as two-stage plasma arc reactors have delivered H_2 concentrations as high as 83%, alongside the co-production of reusable slag materials [15]. The choice of reactor design, feedstock type, and catalysts—including alkaline earth metals and high-entropy oxides—was shown to be decisive in maximizing hydrogen yield and improving tar cracking [16,17]. Innovative catalyst systems, such as Ni–Mo_2C/γ-Al_2O_3 and Ni–Fe/CaO–SBA-15, have further demonstrated superior syngas outputs (55.2 mol/g and 163.7 mmol/g, respectively) by enhancing metal dispersion and reducing coke formation [18,19]. Waste-derived catalysts, such as RDF and tire ash, have also been validated for their effectiveness in plastic pyrolysis–reforming, with simulation models reporting high conversion efficiencies—up to 92.77 k-mol/h of H_2 and 960.13 kg/h of methanol [20,21]. Comprehensive surveys emphasize the importance of process parameters—such as temperature, pressure, and steam-to-feed ratios—and catalyst design for streamlining hydrogen selectivity and minimizing coke formation [22,23].

Hydrogen-rich syngas generated via thermal cracking of crude glycerol–water blends has proven beneficial in combustion engines, improving brake thermal efficiency and reducing emissions [24]. Moreover, dry reforming with CO_2 not only supports CO production (by a factor of 21) but also enables the direct integration of syngas for power applications, making it an environmentally viable option [25].

Application of syngas in CI engines has also been studied. For instance, a dual-fuel CI20 engine using biodiesel–hydrogen blend showed optimal performance at IT30 with 16 lpm of hydrogen, yielding a BTE of 32.7% and significant reductions in CO (48%), HC (22.2%), and smoke (34.3%) [26]. Furthermore, value-added carbon materials derived from pyrolyzed PET have shown good techno-economic feasibility, with payback periods as low as 2.5 years [27]. The integration of microwave-aided co-pyrolysis and simulation tools has improved the yield of aromatics and syngas. Meanwhile, hydrothermal liquefaction (HTL) of HDPE-rich plastics has provided oil yields of up to 87 wt% under optimized conditions [29,30].

Despite promising outcomes, there is a clear need to optimize catalyst design and integrate high-pressure steam reforming under moderate temperatures for scalable application. This study proposes a two-stage thermochemical process utilizing a redox-active, coke-resistant Ni–Mo/CeO_2 catalyst at 25 bar and 250–300 °C. The system aims to enhance H_2 yield, maximize carbon conversion, and minimize residue generation, thereby contributing to the sustainable valorization of undegradable municipal plastic waste within a circular economy framework.

The conversion of hydrocarbon-based feedstocks into hydrogen-rich syngas primarily occurs through a series of catalytic thermochemical processes, as shown in equations (1) and (2). Among these, steam reforming plays a dominant role, particularly in the presence of a Ni–Mo/CeO_2 catalyst, which enhances hydrogen yield and reduces carbon deposition. The generalized steam reforming reaction is represented as: Steam Reforming of Hydrocarbons. During this process, carbon monoxide formed via steam reforming undergoes the water–gas shift (WGS) reaction, further increasing the hydrogen concentration (equation 3).

$C_nH_m + nH_2O \xrightarrow{\text{Ni−Mo/CeO2}} n\,CO + \left(\frac{n+m}{2}\right) H_2$ eq (1)
$C_3H_8 + 3H_2O \rightarrow 3CO + 7H_2$ eq (2)
$CO + H_2O \rightleftharpoons CO_2 + H_2$ eq (3)

Additionally, cracking of long-chain hydrocarbons occurs, particularly in heavy feedstocks, producing light gases (C_1–C_3), hydrogen, and a carbonaceous residue. Ni facilitates this reaction at the Mo-based active site, which is shown in equation (4)

$C_xH_y \xrightarrow{\text{(Ni or MoC1−C3)}} \text{gases} + H_2 + C\ (\text{solid})$ eq (4)

Nickel (Ni) actively breaks C–H and C–C bonds in hydrocarbons, facilitating syngas production. Molybdenum (Mo) enhances Ni stability and resists sulfur poisoning. Cerium oxide (CeO_2) exhibits redox behavior and oxygen storage capabilities, facilitating catalyst regeneration. Together, Ni–Mo/CeO_2 promotes efficient reforming and high hydrogen yield from plastic waste. These reactions collectively enhance the production of H_2 and CO, reduce tar formation, and improve the thermal and chemical efficiency of the municipal plastic waste reforming process. This research work addresses the dual problem of municipal waste management for non-biodegradable waste and sustainable energy production through the production of hydrogen-rich syngas from municipal waste.

Despite several advances in pyrolysis and reforming of plastic waste, most studies lack integration of optimized catalyst systems under pressurized steam reforming to maximize hydrogen yield. The objective of this work is to develop a two-stage thermochemical process utilizing a Ni–Mo/CeO_2 catalyst under high-pressure steam conditions to valorize non-recyclable municipal plastic waste into hydrogen-rich syngas. The novelty lies in the synthesis and application of a redox-active, coke-resistant Ni–Mo/CeO_2 catalyst, operated at 25 bar and moderate reforming temperatures.

2. MATERIALS AND METHODS

This investigation is on recycling non-biodegradable municipal plastic waste into hydrogen-rich syngas through a two-phase thermochemical treatment model. To model typical urban waste, a 1 kg batch of mixed plastic waste, comprising LDPE, PET, multi-layer packaging, and other low-value plastics, was selected. The feedstock was decomposed in an electric furnace at temperatures of 450, 500, 550, and 600 °C to produce vapors and gases through pyrolysis. These products were subsequently directed to a second fixed-bed catalytic reformer inhabited by a 20g NiMo/CeO_2 catalyst. Reforming was carried out at 250 °C, 275 °C, and 300 °C under a pressure of 25 bar, and steam was injected at a steam-to-feedstock ratio of 1.5:1 to fuel endothermic reforming reactions. This technique significantly increased the yield of hydrogen and reduced the amount of solids remaining. The process demonstrates a feasible waste-to-energy route that can be utilized to achieve circular economy objectives, as plastic waste can be repurposed into high-value syngas. The conditions of experimentation were designed to be reproducible and scalable, enabling the development of future sustainable urban waste management systems.

The raw material of this work was a combination of different kinds of post-consumer municipal plastic waste, such as low-density polyethylene (LDPE), polyethylene terephthalate (PET), polypropylene (PP), multilayer packaging films, and contaminated carry bags, typically not used in the conventional recycling system because of their composite nature and poor degradability. Approximately 10 kg of unsorted plastic waste was obtained from municipal waste handling centres in an Indian locality. The plastic waste was subjected to several pretreatment procedures to ensure homogeneity and effective thermochemical conversion. The first stage in the process involved the physical separation of garbage by skilled labourers to eliminate the presence of non-plastic materials, including metal, glass, and organic waste. The plastic materials of choice were then fed to a shredder (industrial grade) so as to mechanically reduce the particle size to < 5 mm to achieve a better heat transfer during the pyrolysis process. After shredding, the material was dried in a hot air oven at 105°C for 4 hours to eliminate any remaining moisture, ensuring the moisture content was reduced to less than 1 wt%. This preprocessing is essential to prevent premature reactions caused by steam and ensure the consistency of the syngas yield with further thermal treatment. The resulting feedstock was subsequently stored in airtight containers before being introduced into the pyrolysis reactor.

Figure 1. Syngas Generation from Municipal Waste for Clean Energy

The process of converting municipal plastic waste to syngas (Figure 1) begins with the collection and mechanical sorting of mixed plastic waste to remove non-plastic contaminants. The sorted plastics, which include LDPE, PET, PP, and multilayer films, as well as other residues, are further shredded into pieces of identical sizes, measuring less than 5mm. This feedstock is shredded, and then dried at 60 ℃ for 4 hours to ensure a moisture content of less than 1 wt%. This guarantees optimal thermal efficiency during conversion. The dried plastic mixture is placed in air-tight containers to prevent rehydration and then used in repeatable batches of 200g for each pyrolysis test. The resulting feedstock is then thermally broken down in a pyrolysis reactor, which produces volatile compounds and gases. These products are directed to a secondary catalytic reformer, where the presence of a catalyst, such as Ni-Mo/CeO_2, promotes additional cracking and reforming. The step enhances the quality of syngas by adding hydrogen and carbon monoxide, thereby increasing the energy potential and utility of the syngas.

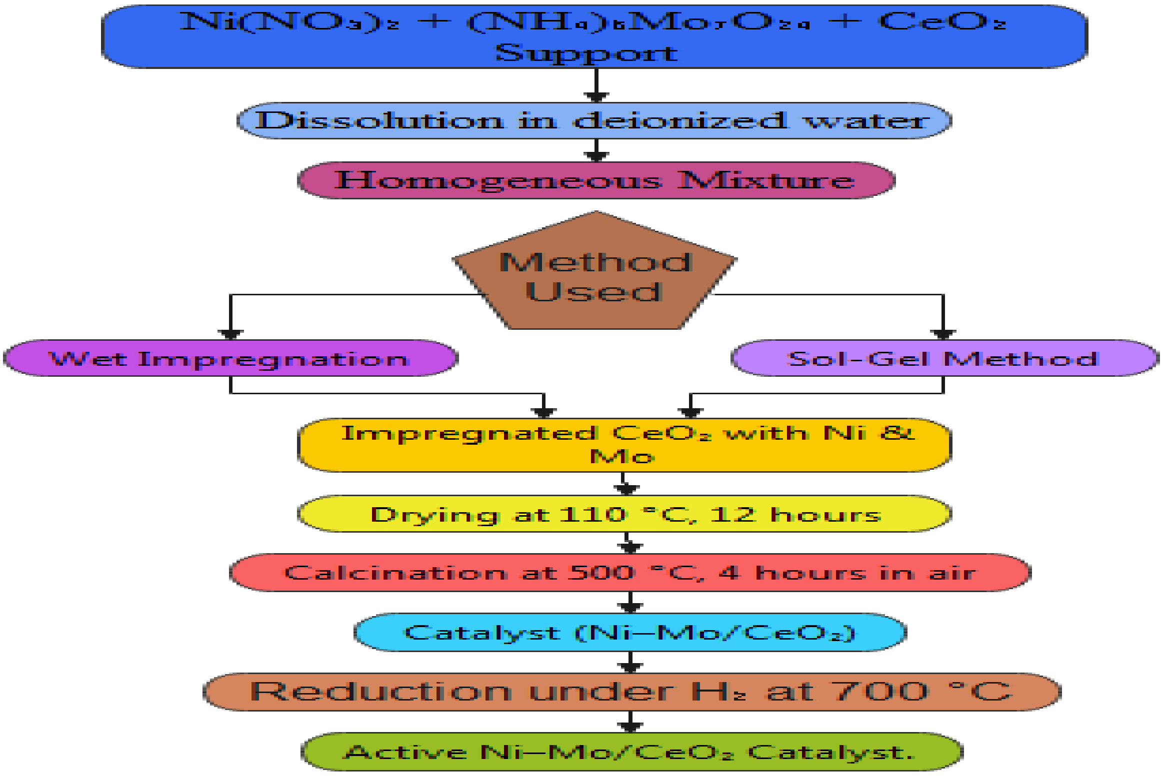

Figure 2. Flow Diagram of Ni–Mo/CeO_2 Catalyst Synthesis

NiMo/CeO_2 catalyst was synthesized by the wet impregnation method. In the given process, Figure 2 indicates that nickel nitrate and ammonium molybdate were initially dissolved in deionized water and well mixed with ceria (CeO_2) support to create a homogenous solution. The effective dispersion of the metal precursors on the support was maintained by constant stirring. The slurry was dried in an oven at 110℃ for 12 hours to eliminate any remaining moisture. It was then calcined at 500℃ under air, which allowed for the decomposition of the metal salts and the binding of Ni and Mo to the surface of the CeO_2. Ni-Mo/CeO_2 catalyst was then prepared by hydrogen reduction of the calcined material at 700 ℃ to form the active Ni-Mo/CeO_2 catalyst. The procedure enhanced the metal-support interaction, promoted metal dispersion, and ensured the formation of effective active sites, facilitating the efficient conversion of plastic waste to syngas.

The experimental configuration for producing syngas from municipal plastic waste involved a two-stage process: thermal pyrolysis and catalytic steam reforming of the waste under a nitrogen atmosphere. First, 200 g of pretreated plastic waste (LDPE, PET, multilayer films) with a particle size of <5 mm and a moisture content of <1 wt% was used in a fixed-bed pyrolysis reactor. The electric furnace was used to preheat the reactor externally, and nitrogen was purged at a rate of 50 mL/min to maintain an inert atmosphere within the reactor and prevent oxidation. When heated (450 to 600℃), the plastic thermally degraded into gas, condensable liquids, and solid char. The gases were fed into a second reforming reactor filled with 20 g of Ni 0.5Mo/CeO_2 at the same rate. Steam (60% by volume) was co-fed to the reformer to encourage endothermic reforming reactions.

3. RESULTS AND DISCUSSION

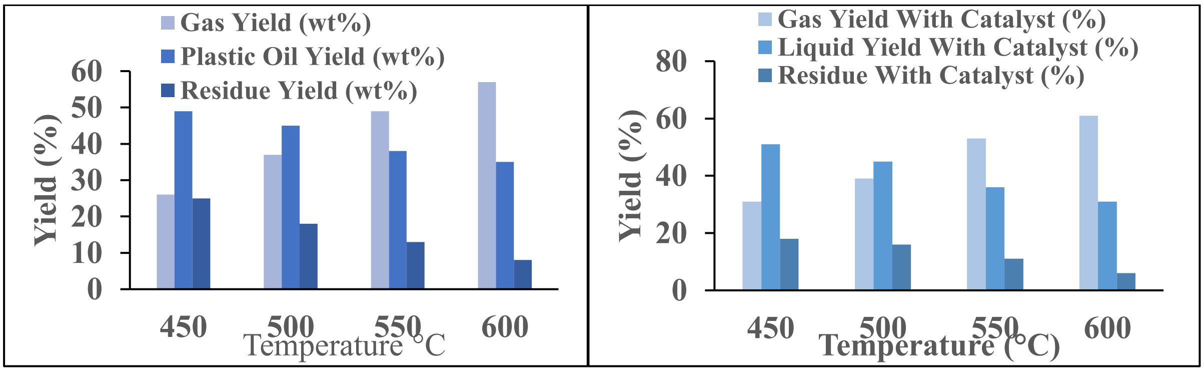

Figure 3. Catalyst impact on gas and liquid yield at different temperatures

Figure 3 Shows how NiMo/CeO_2 affects the gas yield of municipal plastic waste pyrolysis in the temperature range of 450 ℃ to 600 ℃. In the absence of a catalyst, the gas yield increased to 57% compared to 27%. With a catalyst, the gas yield increased to 63% compared to 31%, due to enhanced cleavage of C-C and C-H bonds. The catalyst led to a higher conversion of the liquid to syngas, with a liquid yield of 52% and 27% with the catalyst, compared to 49% and 35% without it. There was also a significant decrease in residue, indicating better decomposition. These developments catalyze secondary cracking and reforming, thereby favoring higher H_2 and CO yields at elevated temperatures, which promotes cleaner energy recovery.

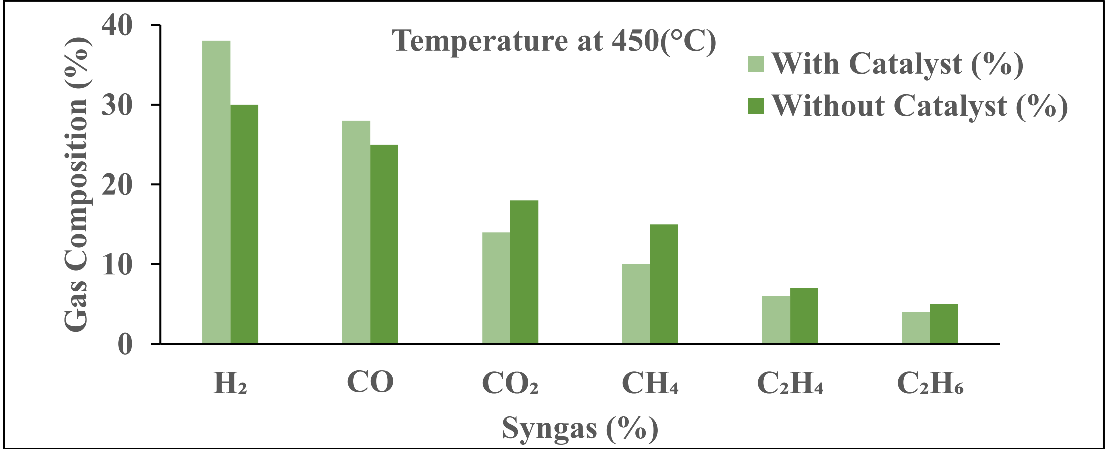

Figure 4. Gas composition at 450 °C with/without Ni–Mo/CeO_2 catalyst

Figure 4 shows the composition of syngas at 450°C of municipal plastic wastes with and without Ni-Mo/CeO_2 catalyst. In the presence of a catalyst, the H 2 yield (37%) was higher than that obtained without a catalyst (30%). Similarly, CO increased by 28 % to 25%. The concentration of CO_2 and CH_4 was also reduced in the presence of the catalyst, as the concentrations fell to 14% and 10%, respectively, due to enhanced cracking and reforming. There was also a decrease in the minor hydrocarbons (C_2H_4 and C_2H_6). This baseline sample at 450°C will be used in the validation and comparison of the processes at higher temperatures as a guide in assessing the effect of the catalyst and optimizing the temperature for syngas production.

Figure 5 The table above presents a comparison of the syngas composition produced by pyrolysis of municipal plastic waste at various temperatures, with and without a Ni-Mo/CeO_2 catalyst. In the absence of a catalyst, the concentration of H_2 increased by 22 to 30 % at 450 °C and 600 °C, respectively, whereas CO was raised by 18 to 28 % at 450 °C and 600 °C, respectively. Conversely, when the catalyst was used, the H_2 yield was significantly improved, with a range of 26% at 450 °C and 36% at 600 °C, and the CO content was 35%. In the meantime, the amount of CO_2 decreased in the catalytic scenario, falling to 10% compared to 15% in the non-catalytic scenario. The catalyst improved the thermal cracking process, as CH_4 decreased to 6%. Small gases, such as C_2H_4 and C_3H_6, were also catalyst-constrained, which improved the purity of syngas. These findings validate the effectiveness of the catalyst in promoting the production of H_2 and CO, which are key factors in producing high-quality syngas.

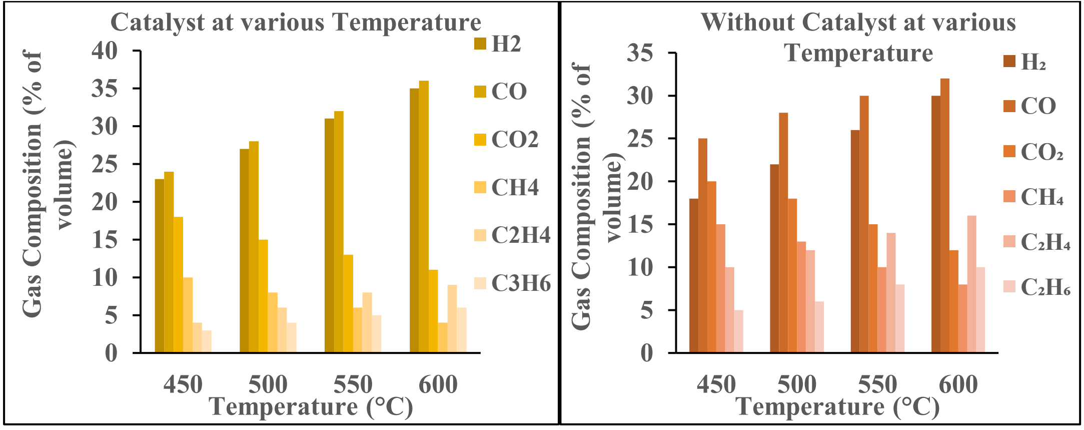

Figure 5 Gas composition with and without catalyst at 450–600 °C

NiMo/CeO_2 catalytic pyrolysis was highly effective in enhancing the production and quality of syngas from municipal plastic waste. The gas yield rose to 61.3 % at 600 °C, resulting in a decrease in the residue to 7.6%. The amount of hydrogen, which was 30.4%, increased to 37.5%, while the amount of CO, which was 28.6%, decreased to 33.7%. At the same time, CH_4 and CO_2 hydrocarbons decreased, indicating that thermal cracking improved. The catalyst was found to be successful in facilitating dehydrogenation and decarboxylation, producing the maximum syngas and minimizing environmental load during plastic waste valorization.

CONCLUSION

This study reports an experimental investigation that demonstrates the potential of catalytic valorization of undegradable municipal plastic waste using Ni-Mo/CeO_2 as a catalyst for producing syngas. This included the thermal decomposition of blended municipal plastic waste at temperatures of 450°C, 500°C, 550°C, and 600°C, as well as steam-assisted catalytic reforming at 250°C, 275°C, and 300°C. The synergetic effect of thermal cracking and catalytic reforming was revealed, as the highest syngas yield of 82.65 vol% was achieved at 600 °C and 300 °C temperatures of pyrolysis and reforming, respectively, using a Ni-Mo/CeO_2 catalyst. Hydrogen reached its highest level of 48.1 vol%, while the CO level was 29.2 vol%. The H_2/CO ratio was maintained between 1.46 and 1.65, which is appropriate for downstream synthesis. The yield of residue was also reduced to 5.4 wt%, indicating effective carbon conversion. The catalyst increased the hydrogen yield by 35.6% and total syngas production by 41.3% compared to non-catalytic reforming. The thermogravimetric results also proved that the catalyst was capable of reactivity with less carbon build-up. Further studies can be conducted to optimize the catalyst regeneration cycles, incorporate mechanisms for CO_2 capture, and extend the range of applicability of the catalyst to other forms of mixed plastic and biomass waste. The reactor design, including heat integration and techno-economic analysis, will be necessary to scale up the reactor for industrial applications.

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