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Auto-Thermal Oxy-Steam Co-Gasification Of Multi-Layer Plastic Packaging Waste And Diverse Biomass For Hydrogen-Rich Syngas: A Simulation-Based Screening Study

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ABSTRACT

The non-recyclability of multi-layer plastic waste (MLP) and the underutilization of biomass present significant environmental challenges. This study developed and validated an Aspen Plus model for auto-thermal oxy-steam co-gasification of biomass-plastic mixture to produce hydrogen-rich syngas in a downdraft gasifier. Five different biomass-plastic mixtures involving MLP and five locally available biomass [Sawdust (SD), Rice Husk (RH), Palm Kernel Shell (PKS), Lemmon Grass (LG), Sugarcane Bagasse (SB)] were formulated and compared for feedstock selection using hydrogen yield under different MLP concentrations. Using the optimal feedstock, a parametric analysis was conducted to determine the ranges of Equivalent Ratio (ER) and Steam to Feedstock Ratio (SFR) that maximize hydrogen yield while maintaining thermal self-sufficiency. Among the fuel mixtures, the most promising was a mixture of MLP and LG. The results showed that hydrogen yield increases with higher MLP concentration and SFR, but decreases when the ER exceeds 0.15. The optimum operating ranges for co-gasification of LG-MLP were determined to be 2.6 to 3.0 for SFR and 0.1 to 0.2 for ER, corresponding to a hydrogen yield of 133.57 to 133.73 g/kg feed. This study offers a practical approach to screening various plastic-biomass mixtures for their hydrogen-production potential prior to laboratory testing and industrial applications.

INTRODUCTION

The persistent increase in plastic waste generation and the underutilization of biomass have intensified the effort toward sustainable waste management and renewable energy generation. The annual plastic production exceeds 400 million tonnes, of which a large proportion eventually turns to waste (UNEP, 2024). Packaging materials account for 40% of plastic waste generated (OECD, 2022). The multilayer plastic (MLP) is a composite material comprising polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET), polystyrene (PS), and sometimes thin aluminum layers. It is commonly used for packaging but is difficult to recycle due to its heterogeneous structure (Ciawi *et al*., 2025). Distinctly, biomass is an attractive material for energy generation due to its renewability, carbon neutrality, sustainability, and abundance (Adeleke *et al.,* 2024). However, the direct use of biomass for energy generation is limited by feedstock availability (Block *et al.,* 2019), variability in biomass composition (Yan *et al*., 2022), and corrosion arising from high oxygen content (Fan *et al.,* 2019). Thermochemical conversion via co-gasification of biomass and plastic is a promising route for producing hydrogen-rich syngas. Co-gasification of biomass and plastic, investigated through both experimental and modelling approaches, has been widely studied and consistently produces higher-quality products than gasification of the individual components alone (Ajorloo *et al.,* 2024; Tian *et al.,* 2024; A Erdem *et al.*, 2023; Rosha *et al.,* 2022). This process offers a solution by mitigating the drawbacks of each feedstock individually (Ajorloo *et al.,* 2022).

The reactive medium supplied to the gasifier to support the gasification process may consist of air, oxygen, steam, carbon dioxide, or any combination of these. The choice of gasifying agent substantially affects the compositions and the thermal potential of syngas. Oxy-steam gasification overcomes the challenges of nitrogen dilution in air gasification and the external heat requirement of steam gasification by supplying internal heat through partial oxidation. Hence, auto-thermal operation is achieved by supplying the heat required for endothermic reactions from exothermic reactions, making the system thermally self-sufficient.

Due to simplicity in design and operation, and suitability in decentralized applications, the use of the downdraft gasifier, among other gasifier configurations for syngas production, has been on the increase. Maintaining auto-thermal operation during biomass-plastic co-gasification in a downdraft gasifier requires careful control of key operating parameters, including the plastic content (PC) of the fuel blend, the equivalence ratio (ER), and the steam-to-fuel ratio (SFR). However, determining the optimal conditions experimentally is both time-consuming and costly.

Process simulation using Aspen Plus has become an important tool in predicting syngas compositions from biomass-plastic co-gasification. Cao et al. (2024) used a kinetic-based model in Aspen Plus to simulate the co-gasification of polyethylene (PE) and pine wood under various operating conditions, with steam serving as the gasifying agent. Erdem et al., (2024) used the thermodynamic equilibrium model of Aspen Plus to simulate the co-gasification of PE and biomass to investigate parametric studies on hydrogen production. Tian et al. (2024) developed an equilibrium-based Aspen Plus model with good prediction accuracy for the co-gasification of rice husk and PS. Rosha et al. (2022) developed an equilibrium-based Aspen Plus model for algae-biomass co-gasification to generate hydrogen-rich fuel. All the studies predicted with low error values. Unlike previous studies that mostly focused on co-gasification of biomass and plastics (PE, PP, PS, PET) to evaluate syngas compositions, this work integrates the most challenging plastic waste, MLP, with different biomass feedstocks to estimate hydrogen yield in an autothermal downdraft gasifier. A thorough review of existing studies indicates that the estimation of the hydrogen production potential of co-gasification of MLP with different biomasses using simulation studies is rarely reported in the literature.

In this study, an Aspen Plus model was developed to simulate oxy-steam co-gasification of MLP and biomass in an autothermal downdraft gasifier and to investigate the hydrogen yield of five different biomass-MLP fuel mixtures under varying PC, ER, and SFR. The objective is to identify the most suitable biomass for MLP and optimal operating windows that maximize hydrogen yield, while maintaining auto-thermal conditions. This study offers a practical approach to screen different plastic-biomass mixtures for their hydrogen production potential prior to laboratory testing and industrial applications.

METHODOLOGY

Feedstock Selection and Characterization

Based on availability, suitability for efficient gasification, and differences in physicochemical properties, five Nigerian biomass species, including sawdust (SD), rice husk (RH), palm kernel shell

(PKS), lemon grass (LG), and sugarcane bagasse (SB), were selected. MLP was considered due to its persistent increase in generation and limited recyclability. The biomass characterization was carried out using proximate and ultimate analyses data obtained from the literature. Due to the heterogeneity and inseparable laminated layers in MLP waste, a representative pseudo-component was adopted. The proximate and ultimate analysis results for the dominant packaging polymers (low-density PE, high-density PE, PP, PS, and PET) were obtained from the literature (Martinez-Narro et al., 2023) to determine the representative compositions of MLP by averaging each parameter. This approach was adopted to capture the contribution effect of constituent polymer layers. The proximate and ultimate results of the biomass and MLP used in this study are presented in Table 1. To prevent plastic agglomeration in a downdraft gasifier, co-gasification of biomass blended with up to 30% plastic is considered (Xiao *et al.,* 2007).

Simulation Model Development

The oxy-steam co-gasification of biomass and MLP waste was developed in Aspen Plus V11 to produce hydrogen-rich syngas. The Aspen Plus flowsheet of this process is shown in Fig. 1. The model assumed thermodynamic equilibrium and overlooked reactor hydrodynamics and tar formation (Erdem et al., 2024; Ranjan et al., 2023). The gases were assumed to behave as ideal gases (Cao et al., 2024). All nitrogen and sulfur in the feedstocks were assumed to convert to ammonia (NH_3) and hydrogen sulfide (H_2S), respectively (de Andrés, 2019). The char was considered pure carbon. The enthalpy and density of biomass, MLP, and ash, which were defined as non-conventional components, were calculated using the HCOALGEN and DCOALIGT models, respectively. The thermodynamic properties of carbon, specified as a solid, and other component species categorized as conventional components were calculated using the Peng-Robinson model with Boston-Mathias modifications (PR-BM).

The raw biomass (BIO) and MLP (PLT) were dried and decomposed into conventional components including C, H_2, O_2, N_2, H_2O, S, char, and ash in the yield reactors YRBIO and YRPLT, respectively, operating at 350 °C and 1 atm. The yield of each component in YRBIO and YRPLT is controlled by calculator blocks (BYRCAL and PYRCAL, respectively) embedded with nested FORTRAN code. The products of YRBIO (S1) and YRPLT (S2) were mixed in a mixer (MIX1) to produce stream S3. Next, the mixed stream (S3) was separated into the volatiles (S4), consisting of C, H_2, O_2, N_2, H_2O, and S, and the solid fraction (S5), comprised char and ash. The stream S4 was sent to the stoichiometric reactor (INGR), where NH_3 and H_2S were formed through the reactions (R9 and R10). The reactor products (S6) entered Gibbs' reactor (PYRR) to form pyrolysis products (S7). A fraction of the char (8% of the total produced) was considered unconverted char and separated along with ash in SEP4 as ASH-CHAR (Ranjan et al., 2023), while the remaining char was directed to the gasification reactor (GASF1) as stream S8. Depending on the predetermined SFR and ER, the mixture of steam (STM), oxygen, char (S8), and pyrolysis products (S7) was then fed into the gasifier (GASF1) modelled as an RGibbs reactor. The reactions in GASIF1 proceed according to R1-R8. The gasifier product stream (S10) was passed through a cooler (CL), cooled to 25 °C, producing a cooled stream (S11) that enabled separation of syngas from the remaining gasifier products (S12).

The performance evaluation of a gasification system for hydrogen production under varying operating parameters can be assessed using hydrogen yield, syngas composition, and the Lower Heating Value (LHV) of the syngas (Erdem et al., 2024). The hydrogen yield is expressed as the ratio of hydrogen mass flow rate (g/hr) to feedstock flow rate (kg/hr). The three operating parameters considered to have appreciable influence on the performance criteria are ER, SFR, and PC. Table 3 presents the base values and range for their variations. These values were chosen based on similar studies in the literature (Erdem *et al.*, 2024; Ranjan *et al.*, 2024; Cao *et al.*, 2024). The ER is calculated using Equation 1

$$ER = \frac{\text{Mass flow rate of oxygen supplied}}{\text{Mass flow rate of stoichiometric oxygen}} \quad 1$$

The SFR, which denotes the amount of steam supplied to the gasifier in relation to the feedstock, is given by Equation 2.

$$\text{SFR} = \frac{\text{Mass flow rate of the oxygen supplied}}{\text{Mass flow rate of the feedstock}} \quad 2$$

Table 4 Operational range of model parameters

Parameters	Base value	Range	Interval
PC (wt. %)	20	0 – 30	10
ER	1.4	1 – 3.8	0.4
SFR	0.21	0.05 – 0.4	0.05

Simulation model validation

The Aspen Plus model was validated by comparing its results with experimental data reported by Sandeep and Dasappa (2014). They conducted an experimental investigation on oxy-steam gasification of casuarina wood chips under varying conditions in a downdraft gasifier. The range of ER and SFR considered in this study was 0.75–2.7 and 0.18-0.3, respectively. Results for hydrogen yield and lower heating value (LHV) from five different experimental runs were used to assess the model's quantitative accuracy using Root Mean Squared Error (RMSE), as given in Equation 3.

$$RMSE = \sqrt{\frac{\sum (EXP_i - MOD_i)^2}{N}} \quad 3$$

where EXP_i are experimental data, and MOD_i are simulation results, and N is the number of data points.

Biomass-MLP Blend Formulation and Screening Methodology

A biomass-MLP blend was formulated by combining each biomass type with MLP at different MLP contents (PC) to determine the influence of MLP content on syngas quality and the most-performing feedstock for hydrogen production. Using the developed Aspen Plus model, the formulated blends (SD-MLP, RH-MLP, PKS-MLP, LG-MLP, SB-MLP) were simulated to produce syngas under identical operating conditions. The simulation results for each blend were compared with respect to hydrogen yield. For all simulations, the feed rate, ER, and SFR were kept at 1000 kg/hr, 0.21, and 1.4, respectively, while PC was varied between 0 and 30 wt.%. The best feedstock was identified and selected for further screening of operating parameters to determine the favourable operating conditions. ER and SFR were varied as screening parameters to evaluate hydrogen yield and identify the operating window that maximizes hydrogen yield while maintaining auto-thermal conditions.

RESULTS AND DISCUSSION

Model Validation

The validation results of the Aspen Plus model are presented in Fig. 2. The comparison of the predicted hydrogen yield and LHV of syngas to experimental data from the literature (Sandeep and Dasappa, 2014) is shown in Fig 2(a) and (b), respectively. The RMSE values of 4.69 in Fig. 2(a) and 0.98 in Fig. 2(b) demonstrate close agreement with the experimental data

Table 1: Proximate and ultimate analysis of biomass and plastic

	SD	RH	PKS	LG	SB	MLP
Ultimate Analysis (wt %)						
Carbon	47.40	40.75	44.39	41.12	47.69	81.06
Hydrogen	6.24	5.05	8.74	5.29	5.70	10.78
Oxygen	46.30	52.73	43.34	51.68	46.00	7.99
Nitrogen	0.05	1.38	2.36	1.75	0.57	0.17
Sulphur	0.01	0.09	1.18	0.06	0.04	-
Proximate Analysis (wt %)						
Moisture content	9.79	7.96	1.99	0.48	0.28	0.15
Fixed carbon	12.91	13.48	10.25	23.95	14.88	2.33
Ash content	0.05	22.58	4.56	9.88	1.30	2.31
Volatile matter	77.25	55.97	83.20	66.67	83.82	95.35
References	a	b	A	C	c	D

a = Osuolale et al., 2021; b = Luo et al., 2021; c = Onokwai et al., 2022; d = Martinez-Narro et al., 2023.

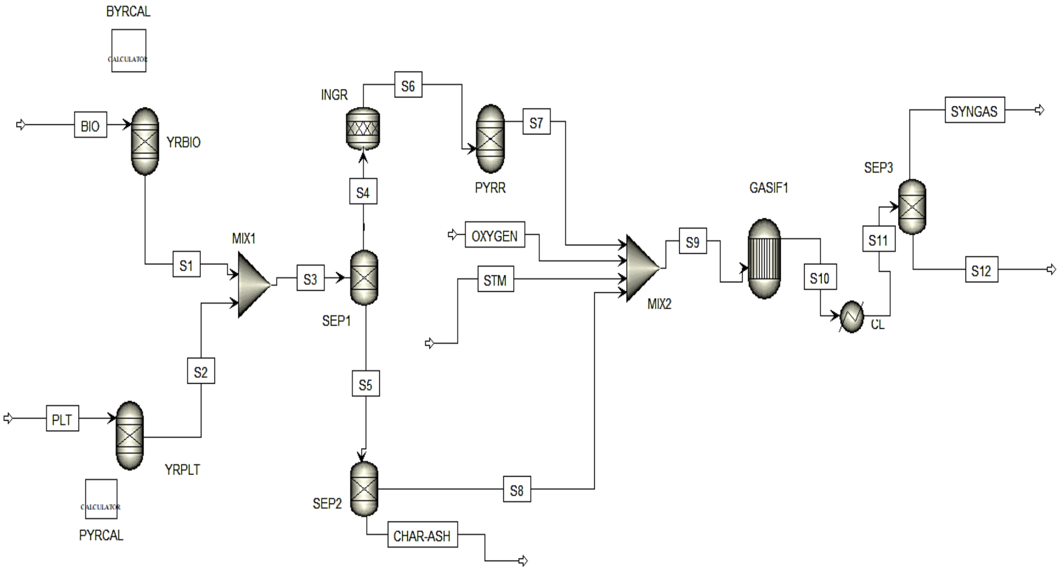


Fig.1: Flowsheet of the Aspen Plus model for biomass-plastic co-gasification

$$C(s)+0.5O_2(g)\leftrightarrow CO(g) \qquad -111\ kJmol^{-1} \qquad (R1)$$

$$CO(g)+0.5O_2(g)\leftrightarrow CO_2(g) \qquad -283\ kJmol^{-1} \qquad (R2)$$

$$H_2(g)+0.5O_2(g)\leftrightarrow H_2O(g) \qquad -242\ kJmol^{-1} \qquad (R3)$$

$$C(s)+H_2O(g)\leftrightarrow CO(g)+H_2(g) \qquad 131\ kJmol^{-1} \qquad (R4)$$

$$CO(g)+H_2O(g)\leftrightarrow CO_2(g)+H_2(g) \qquad -41\ kJmol^{-1} \qquad (R5)$$

$$CH_4(g)+H_2O(g)\leftrightarrow CO(g)+3H_2(g) \qquad 206\ kJmol^{-1} \qquad (R6)$$

$$C(s)+2H_2(g)\leftrightarrow CH_4(g) \qquad -75\ kJmol^{-1} \qquad (R7)$$

$$C(s)+CO_2(g)\leftrightarrow 2CO(g) \qquad 172\ kJmol^{-1} \qquad (R8)$$

$$S(s)+H_2(g)\rightarrow H_2S(g) \qquad \_\_ \qquad (R9)$$

$$N_2(g)+H_2(g)\rightarrow NH_3(g) \qquad \_\_ \qquad (R10)$$

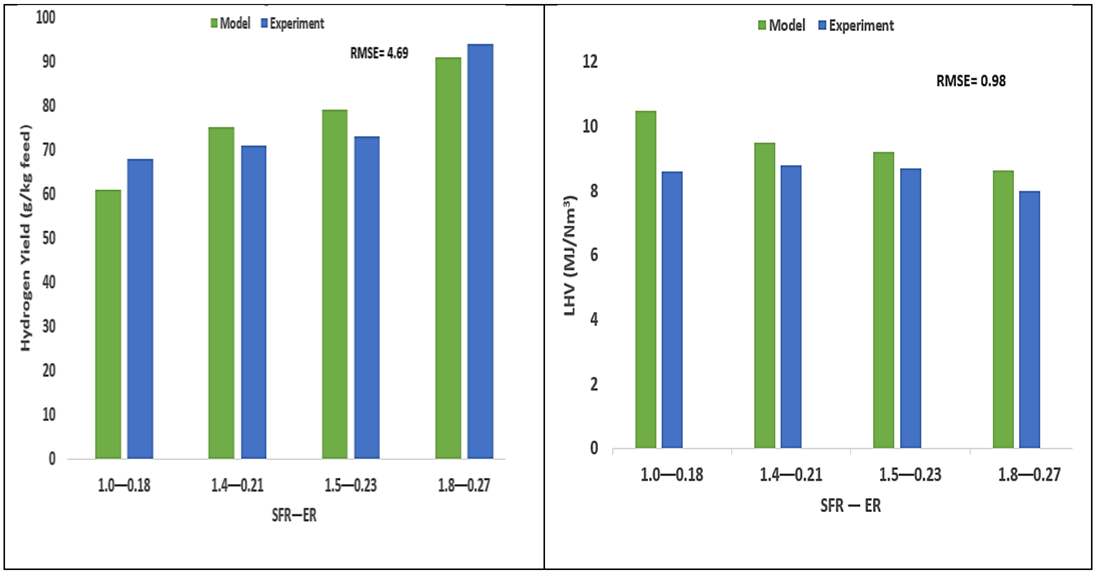
These small error values authenticate the model's suitability for screening applications. Singh *et al.* (2022) simulated biomass-plastic co-gasification in a fluidised bed gasifier using Aspen Plus. RMSE values in the range 3.101–6.775 were reported when predicted syngas compositions were compared with experimental values under identical operating conditions. Error values similar to this study were reported by Ranjan *et al.* (2023) and Ajorloo et al. (2022). Slight deviations in the model predictions are likely attributable to the assumptions made when developing the model.

Comparison of Hydrogen-Rich Syngas Production Systems for Various Feedstocks

The PC of the fuel mixture plays an essential role in determining syngas composition and yield in the biomass-plastic co-gasification system. Hence, the screening analysis was based on hydrogen yield estimates at different PC levels. The comparative analysis was carried out by varying PC, while SFR and ER were kept constant. As shown in Fig. 3, the hydrogen yield increases with increasing MLP concentration across all mixture ratios. For instance, the hydrogen yield of RH-MLP increased from 62.61 to 96.25 g/kg feed when the PC increased from 0 to 30%. This behaviour may be attributed to the high hydrogen content of MLP and polymer decomposition into H_2, CH_4, and other light hydrocarbons (Erdem *et al.,* 2024). Ahmed *et al.* (2011) reported an increase in hydrogen yield during the co-gasification of wood chips and polyethene. The increasing trend was also reported by Lopez *et al.* (2015). This implies that MLP, as a co-feedstock, enhances the concentrations of both H2 and CH4, thereby increasing the LHV of the syngas. As shown in Fig. 3, the highest hydrogen yield of 98.06 g/kg feed was obtained at PC of 30% for co-gasification of LG-MLP, while the lowest value of 62.61g/kg feed was obtained at PC of 0% for co-gasification of RH-MLP. In the Aspen Plus model development, char was assumed to be solid carbon and to actively participate in the gasification reactions. Therefore, the highest hydrogen yield of LG-MLP may be attributed to a high fixed carbon content, which makes solid carbon available for char-steam. (R4) and water-gas shift reaction (R5), enhancing hydrogen production. Compared to SD and PKS, which have higher hydrogen content, LG produced more hydrogen due to the dominant effects of FC, N, and S. Therefore, LG-MLP with 30% PC was selected for subsequent analysis.

Steam to fuel ratio

Fig. 4 illustrates the effect of SFR on hydrogen yield when the PC and ER were kept at 30% and 0.21, respectively. Increasing the SFR from 1.0 to 3.8 increased the hydrogen yield from 83.21 to 137.32 g/kg feed. The rate of increase in hydrogen yield slowed down and became less significant beyond an SFR of 3.0. The rapid change in hydrogen yield when SFR is less than or equal to 3.0 may be due to the strong participation of steam in steam-assisted reactions (R4, R5, and R6) to produce hydrogen. Beyond an SFR of 3.0, steam only acts as a thermal sink, resulting in only slight increases in hydrogen yield. This behaviour is consistent with the study of Wei *et al.*, (2007), who investigated the steam gasification of legume straw and pine sawdust. Lv *et al.*, (2003) also reported minor variations in H_2 composition as the SFR increased from 0.61 to 1.0, suggesting that beyond a certain threshold, further increases in steam flow may have a limited impact on H_2 yield. Therefore, an SFR within the range 2.6 to 3.0 was identified as the optimal range.



(a) (b)

Fig. 2: Model validation with experimental data for (a) hydrogen yield. (b) LHV

Equivalence Ratio

In Fig. 5, ER was varied from 0.05 to 0.4, while maintaining an SFR of 3.0 and a PC of 30%. At low ER (≤ 0.15), hydrogen yield increased as the ER rose, which can be attributed to the dominance of endothermic steam methane reforming (R6) and water gas (R4) reactions over the slightly exothermic water gas shift reaction (R5), which opposes an increase in hydrogen yield. Beyond 0.15, excessive oxygen promotes the direct oxidation of hydrogen, carbon, and sulphur, and the dominance of reaction R5 over R4 and R6, leading to a decrease in hydrogen yield as

ER increases. The change in H_2 yield with temperature was not significant, aligning with findings by Ranjan *et al.* (2023). Therefore, the ER of the range 0.1 to 0.2 could be considered the optimal range.

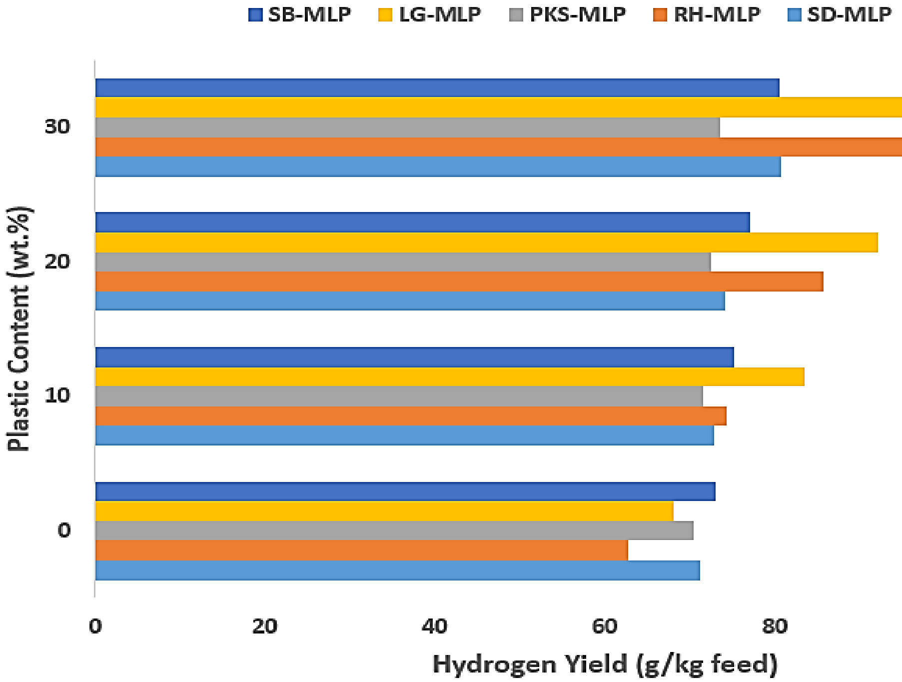


Fig. 3 Comparison of different biomass-plastic mixtures for hydrogen yield.

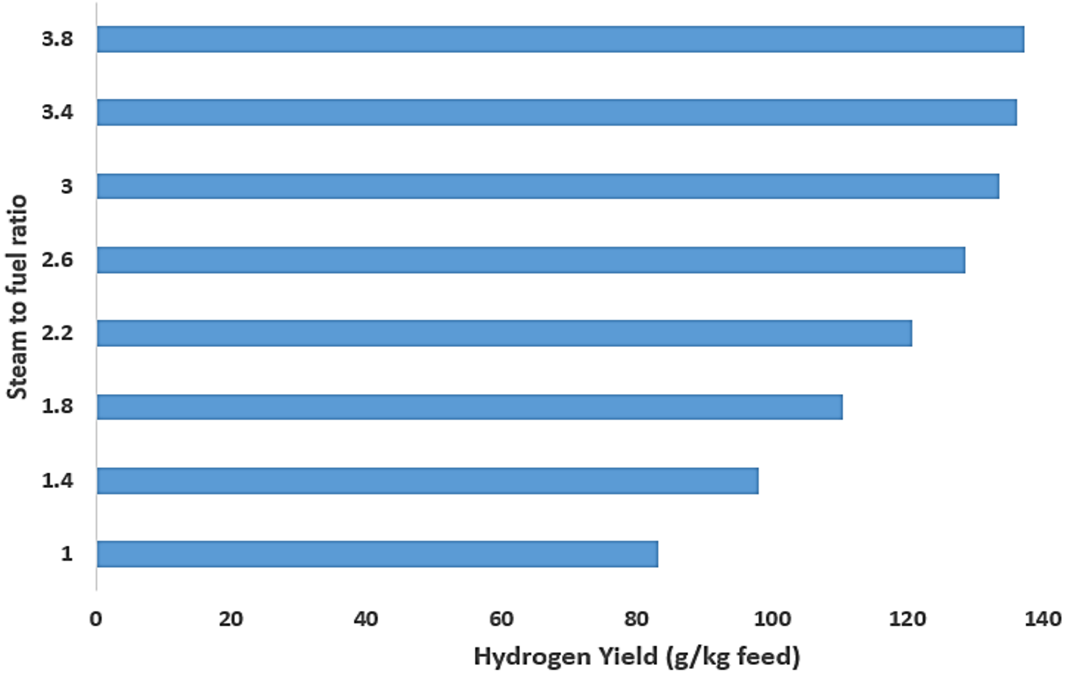


Fig. 4 Effect of the steam-to-fuel ratio on hydrogen yield (PC = 30%, FR = 0.21)

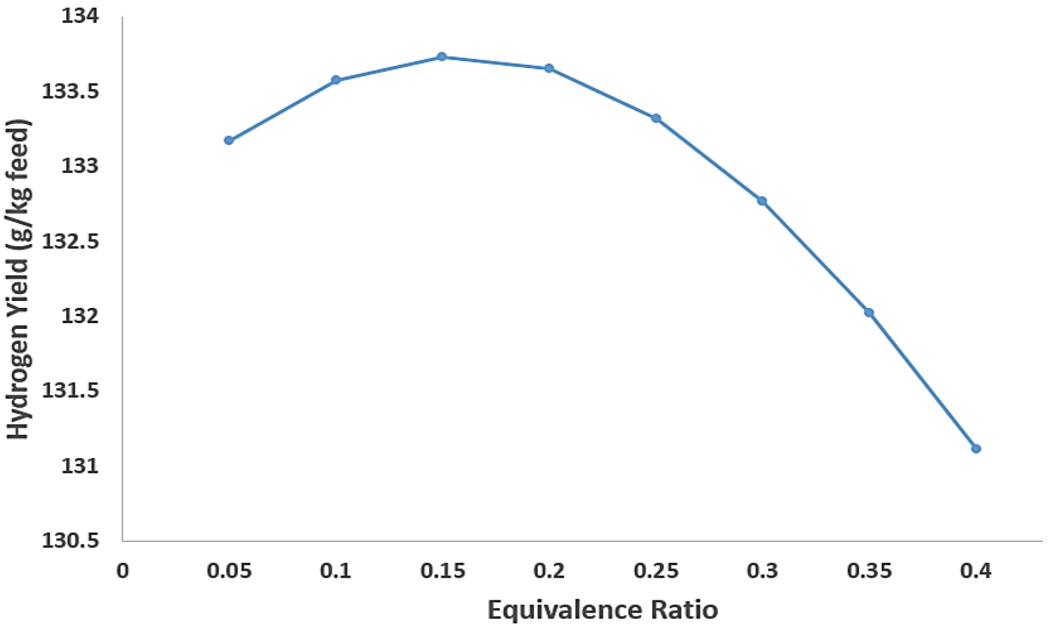


Fig. 5 Effect of equivalent ratio on hydrogen yield (PC = 30%, SFR = 3.0)

CONCLUSIONS

An Aspen Plus model for auto-thermal oxy-steam co-gasification of biomass-plastic mixture was developed to screen a mixture of MLP waste and five different locally available biomass for hydrogen-rich syngas production in a downdraft gasifier. The validated model was used to identify the best biomass for MLP waste under different PC for hydrogen production. The best fuel mixture was considered model's input, and the optimal ranges for SFR and ER were determined. For co-gasification of the five fuel mixtures (SD-MLP, RH-MLP, PKS-MLP, LG-MLP, SB-MLP), the hydrogen yield increased with increasing MLP concentration. The highest hydrogen yield of 98.06 g/kg feed was observed at a PC of 30% during co-gasification of LG-MLP, thereby identifying LG-MLP as the most suitable fuel mixture for subsequent analysis. The lowest value of 62.61g/kg feed was obtained at PC of 0% for co-gasification of RH-MLP. The SFR showed a positive correlation with hydrogen yield, whereas hydrogen yield decreased when the ER exceeded 0.15. The optimal operating range for the co-gasification of LG–MLP was identified as an SFR of 2.6–3.0 and an ER of 0.10–0.20, corresponding to a hydrogen yield of 133.57-133.73 g/kg feed. This study offers a practical approach to screen different plastic-biomass mixtures for their hydrogen production potential prior to laboratory testing and industrial applications. These findings are expected to contribute to the design and development of the downdraft gasifier. Further studies should consider experimental investigation, comprehensive performance indicators, kinetic modelling, uncertainty analysis and techno-economic analysis assessment of the process.

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