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Sensitivity Analysis and Optimization of Parameters for the Gasification of High Ash Indian Coal

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Abstract

Gasification of high ash coals and controlling resulting emissions is a challenging task. Effective utilization of these coals depends on proper selection of operating conditions. This paper is aimed at comprehensive analysis of effect of operating conditions on the gasification of a high ash Indian (Jharia) coal with two-fold objectives of maximizing syngas heating value and reducing CO_2 emissions. An ASPEN Plus model is developed to simulate the gasification process and simulation results are validated with experimental data available in literature. The simulated results are in good agreement with experimental results with less than 10% error.

In the first stage the sensitivity analysis of four operating conditions viz., Equivalence Ratio (ER), Steam-to-Coal Ratio (SCR), Gasifier Temperature, and Gasifier Pressures is performed. A design specification Limiting Air Flow Rate (LAFR) is used for effective carbon conversion and reduced emissions. Gasification temperature is the most influencing parameter among all parameters, followed by ER and SCR. Gasification pressure has no significant effect on syngas composition. The results clearly indicate that a lower ER and SCR are favorable for formation of maximum amount of combustible gases (H_2, CO) and minimum amount of CO_2. Different combination of ER and SCR in the specified ranges were examined. In all cases, the maximum efficiency is attained in a temperature range of 600 ^oC to 900 ^oC. In the second stage these parameter are optimized with an objective of maximizing the heating value of syngas and minimizing CO_2 emissions. Both normal and constraint based optimization is performed. In constraint based optimization, a constraint of CO_2 mole fraction less than 1% is imposed. Optimum parametric domains for maximum efficiency are presented.

Keywords: Coal gasification, high ash coals, sensitivity analysis, optimization, equivalence ratio, steam to coal ratio, limiting air flow rate, minimum steam flow rate

Nomenclature

ER	Equivalence Ratio
SCR	Steam to Coal Ratio
HHV	Higher Heating Value (MJ/Nm^3)
CGE	Cold Gas Efficiency
M	Moisture (wt.%)
VM	Volatile Matter (wt.%)
FC	Fixed Carbon (wt.%)
ΔHo	Enthalpy of formation (kJ/mol)
GCV	Gross Calorific Value of coal (MJ/kg)
LAFR	Limiting Air Flow Rate

INTRODUCTION

Gasification is process of converting carbonaceous fuels into high calorific value gases like H_2, CO and CH_4. It is most efficient and eco-friendly process of utilizing fossil fuels and biomass fuels [1]. The product gas of gasification process is called syngas consisting of mainly H_2, CO and other gases like CH_4, CO_2, SO_x and NO_x in smaller quantities. This syngas is used for power generation and for producing petrochemical products. Indian coals contain high amount of ash and makes them difficult to use for direct combustion. Gasification is a good alternative to exploit these resources for power generation. One of the metrics of gasification process is the gasification efficiency which primarily depend on amount of fuel gases H_2 and CO present in syngas. Gasification involves complex chemical reactions and depends on several factors like gasifier design, fuel properties, gasifying agent, operating conditions like flow rate of gasifying agent, gasification temperature, pressure, equivalence ratio, etc.

In the past decade many researchers worked on understanding the effect of various parameters and operating conditions on gasification process. Majority of the work is based on biomass fuels. Seldom work is done on understanding gasification process of coals especially low rank coals available in India. The presence of excess ash and other unwanted constituents and releasing of pollutants like CO_2, SO_x and NO_x, etc., makes these coals an unattractive option for gasification [2]. However, with careful analysis and by proper selection of critical parameters these drawbacks can be overcome.

Yaji Haung et al [3] studied the effect of operating conditions and coal properties and reported that high rank coals like anthracite are more suitable for gasification than low rank bituminous coals. V. Satyam Naidu, et al [4] studied the gasification kinetics of low rank coals and concluded that coals with higher inorganic matter has higher reactivity. Maan Al-Zareer et al [5] studied effect of gasification parameters using kinetic based model and Gibbs free energy model on three coals. They reported that kinetic based gives accurate results than Gibbs free energy based model, but later one is more suitable due to its compatibility to all coal types. They concluded that syngas composition is highly dependent on air and steam flow rates. Vijay Kumar. K et al [6] conducted experimental study on high-ash Indian coal in bubbling fluidized bed using air and steam. They have varied equivalence ratio from 0.25 to 0.35 and steam-coal-ratio from 0.25 to 0.35 and studied their effect on syngas composition. They concluded that steam flow rate is has positive effect on calorific value and gasification efficiency. Neeraj Singh et al [7] conducted mathematical modeling of gasification high-ash Indian coals in moving bed gasification system. They have reported that steam-coal-ratio is dependent on amount of oxidizing agent and that for air based system SCR is 0.4 and for O_2 based system it is 1.5. According to them operating pressure has significant effect on gasification performance and pressure of 8 bar and above is suitable for high ash coals. Nourredine Abdoulmoumine et al [8] studied the effect of temperature and equivalence ratio on mass balance and energy analysis on biomass gasification. They reported that as the temperature and ER increases the mass balance increases. The increase in temperature has positive effect on cold gas efficiency and hot gas efficiency. In contrast the efficiencies are decreased with increase in equivalence ratio. Zhi-Hua Wang et al [9] investigated characteristics of Shenhua bituminous (SH) and Baorixile lignite coal (BRXL) using both a thermogravimetric analyzer and tubular furnace. They reported that both the gasification temperature and atmospheric composition greatly gasification. They concluded that reactivity is lowest under CO_2 gasification atmosphere and increased continuously with increase in the H_2O partial pressure. They emphasized that there will be a particular combination of gasification atmosphere and temperature which is specific to a given coal. Haibin Li et al [10] developed an integrated gasification fuel cell with zero emission and studied the effect of gas flow rate, pressure, pre-heating temperature, heat losses. They reported that as pressure increases lower heating value is reduced and reaction temperature is increased. O_2 preheating has greater influence on reaction temperature and lower heating value when compared to CO_2 preheating.

In the present work, the effect of four parameters viz., ER, SCR, Gasifier Temperature and Gasifier Pressure on gasification of Jharia coal is studied in a comprehensive manner. Gasification process consists of complex chemical reactions. Identification of influencing parameters and understanding their synergetic effect is very important. In this work, the interdependency of the parameters and their effect on efficiency is assessed. The optimum parametric domains are presented for maximum gasifier efficiency and minimum emissions, which was not done in previous works.

MODEL DESCRIPTION:

The model is developed based on Gibbs free energy principle and corresponding flow sheet is given in Figure.1 for simulation. Table.1 presents the details of components along with functionality and also operating conditions. The model consists of two reactors, DECOMP for decomposing the wet coal into it primary components and GASIFIER to carry out the chemical reactions. The main reactions for different processes along with the enthalpy values are shown in Table.2. The yield of the DECOMP reactor is calculated using FORTRAN sub-routine from the ultimate analysis on dry basis. The output of yield reactor is then sent to GASIFIER where the chemical reactions will take place based on minimization of Gibbs free energy principle. Oxygen is produced from air using air separation unit and steam is produced from heater. The heat required for converting water into steam is supplied from gasifier. The product gas is then sent to cyclone separator where the ash is separated and clean syngas is sent for further process.

The following assumptions and model settings are made for conducting the analysis

- The system is operating under steady state conditions
- Coal supplied in the form of particles and the particles have uniform temperature
- There is no pressure drop in the heat exchanger, splitters and cyclone separator
- Coal heat capacity is based on Kirov's correlation (ASPEN option: HCOALGEN)
- Density calculation based on IGT (Institute of Gas Technology) equations (ASPEN option: DCOALIGT)
- Thermodynamic property method used for real components and mixture (ASPEN option: RK-SOAVE)
- Tar formation is neglected

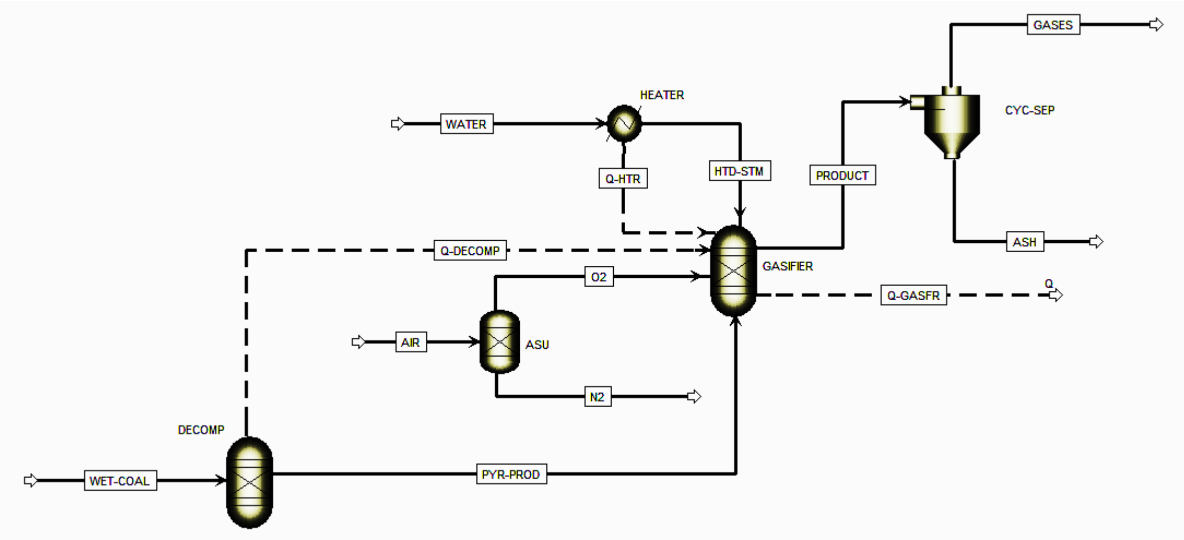

Figure 1: Flow sheet of gasification plant for simulation

Table 1: Components of gasification plant and representation in ASPEN Plus

Block/ Stream	**Type**	**Function**	**Parameters**
DECOMP	RYield Reactor	Decompose the coal into primary form	1 atm, 25 ºC
Air Separation Unit (ASU)	Component Separator	Separate O_2 from Air	1 atm, 25 ºC
GASIFIER	RGibbs Reactor	React all component inputs based on Gibbs free energy principle	1 atm, 300 to 1500 ºC, Rigorous with all possible components
HEATER	Heat exchanger	Convert water into steam at specified temperature and pressure	1 atm, 300 ºC
CYC-SEP	Cyclone Separator	Separate ash from product gas	1 atm, Gasifier Temperature

Table 2: Main reactions taking place in the RGibbs reactor

Process	**Reaction**	**Reaction #**
Oxidation	$C + 0.5O_2 \rightarrow CO\ , \Delta H^{\circ} = -268\ kJ/mol$ $C + O_2 \rightarrow CO_2, H^{\circ} = -\ 406\ kJ/mol$ $H_2 + 0.5O_2 \rightarrow H_2O, \Delta H^{\circ} = -258.8\ kJ/mol$	R1 R2 R3
Water Gas Shift Reaction	$C + H_2O \rightarrow CO + H_2, \Delta H^{\circ} = +131\ kJ/mol$	R4
Boudouard Reaction	$C + CO_2 \rightarrow 2CO\ , \Delta H^{\circ} = +172\ kJ/mol$	R5
Shift Reaction	$CO + H_2O \leftrightarrow CO_2 + H_2, \Delta H^{\circ} = -42\ kJ/mol$	R6
Methanation	$C + 2H_2 \rightarrow CH_4, \Delta H^{\circ} = -75\ kJ/mol$ $CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O, \Delta H^{\circ} = -206\ kJ/mol$	R7 R8

Table 3: Properties of Illinois #06 coal [11]

Proximate Analysis				**Ultimate Analysis**					
M	FC	VM	ASH	C	H	N	Cl	S	O
0.2	58.01	26.46	15.53	73.9	6.24	0.71	0.37	1.77	1.32

Model Validation

The model is validated with the experimental data from the work done by Wen et al., [11] for the Illinois #06 coal. They have investigated the effect of various parameters on syngas composition and compared the experimental results with the Texaco entrained flow gasifier plant data. The ultimate and proximate analysis of Illinois No.6 coal is given in table 3. The proposed model is given input for three runs and the gas components are compared with the experimental results. Table.4 shows the input conditions and output gas components. Since model input streams are different from those of experiment, while giving inputs for O_2 and SCR, the air and steam flow rates are adjusted by using applicable factors to match the inputs of experiment. The operating conditions are gasifier pressure of 24 atm, and steam temperature of 490 ºC for all cases.

Simulation Methodology

The proximate and ultimate analysis of Jharia coal is presented in Table.5. The wet coal is fed to the DECOMP reactor which decomposes it into its primary components. The heat required for the DECOMP reactor (Q-DECOM) is supplied from the Gasifier. A constant coal feed rate (10kg/s) is maintained and feed rates of air and steam are varied. The air feed rate is adjusted such that the equivalence ratio is maintained between 0.1 and 0.6 (step size 0.1). Steam rate adjusted such that SCR is varied between 0.2 and 2.0 (step size 0.2). The stoichiometric air quantity and equivalence ratio are calculated using Eq. (1) and (2) respectively. In order to study the effect of each parameter individually, a separate sensitivity analysis of each parameter is conducted and syngas composition is analyzed from the product gas. The results indicate that as the air flow increases the combustion increases due to presence of excess O_2 resulting in generation of more CO_2 and excess heat in the gasifier (Q-Gasifier). This excess heat has to be recovered by other means. Therefore in order to control the combustion such that the reactions are self-sustained and the heat requirement of gasification plant are met, a design specification for the GASIFIER block LAFR is introduced. The design specification controls the air flow such that gasifier produces just enough heat to meet the plant requirements while keeping the Q-Gasifier and CO_2 formation to a minimum value. The steam flow rate and gasifier temperature are then varied and syngas composition is analyzed. The HHV and CGE for different combinations of parameters are studied and optimum values of parameters are identified. CGE and HHV are calculated using Eq. (3) and (4) respectively.

$$\left({}^{Air}/_{Fuel}\right)_{Stoich} = 4.23 * [2.67C + (8H - O) + S] \quad (1)$$

Where C, H, O and S are wt.% of Carbon, Hydrogen, Oxygen, and Sulfur taken from Ultimate analysis.

$$Equivalence\ Ratio\ (ER) = \frac{\left({}^{Air}/_{Fuel}\right)_{Actual}}{\left({}^{Air}/_{Fuel}\right)_{Stoichiometric}} \quad (2)$$

$$Cold\ Gas\ Efficiency = \frac{Syn\ gas\ flow\ rate\ in\ m^3/s \times Gas\ HHV\ in\ MJ/m^3}{Coal\ feed\ rate\ in\ kg/s \times Coal\ HHV\ in\ MJ/kg} \times 100 \quad (3)$$

$$HHV_{Syngas} = \frac{V_{CO} \times CV_{CO} + V_{H_2} \times CV_{H_2} + V_{CH_4} \times CV_{CH_4}}{100} \quad (4)$$

Where V is the volumetric percentage and CV is the calorific value of individual gas species.

Table.4: Input conditions, and validation of simulation data of Illinois #06 coal [11]

Run No		Input Conditions			Product Gases (g/s)		
		Coal (g/s)	O_2 (g/s)	SCR	H_2	CO	CO_2
Run-1	Experiment [9]	76.66	66.38	0.241	6.01	123.77	9.985
	Model				5.68	113.87	9.38
	%Error				5.49	7.98	6.35
Run-2	Experiment [9]	81.18	62.34	0.314	6.23	112.92	17.2
	Model				6.04	104.52	15.6
	%Error				3.04	7.43	8.99
Run-3	Experiment [9]	82.20	66.82	0.309	6.211	121.5	19.26
	Model				5.77	109.73	17.61
	%Error				7.10	9.68	8.56

Table 5: Properties of Jharia coal [12]

Proximate Analysis				Ultimate Analysis					
M	FC	VM	ASH	C	H	N	Cl	S	O
6.94	36.4	28.5	28.2	49.3	4.81	1.11	0.08	0.31	9.25

RESULTS AND DISCUSSION

Gasification temperature is the most influencing parameter followed by SCR and ER. Gasification pressure has a less significant effect. For each SCR there exists a limiting air flow rate (LAFR) which warrants maximum carbon conversion and minimum emissions. This air flow rate is estimated by a design specification. The effect of each parameter and their interdependency is presented below;

Effect of Equivalence Ratio

Figure. 4(a) to 4(d) show the variation of individual gas components with respect to equivalence ratio. The graphs are plotted at four different steam flow rates (2, 6, 10, and 14 kg/s) at a temperature of 700 ºC. Figure. 4(a) depicts the H_2 flow rate with equivalence ratio at all SCRs. It indicates that as the equivalence ratio increases the H_2 is decreasing. This is due to increase in quantity of O_2 which converts some H_2 into H_2O. This is evident from Figure. 4(d) in which there is an increase in H_2O with increase in equivalence ratio. It is also observed from Figure. 4(a) that the rate of decrease in H_2 formation depends on SCR. Figure. 4(b) represent the variation of CO with equivalence ratio. It can be observed from the figure that as equivalence ratio increases CO is decreasing. This is due to the fact as equivalence ratio increases, O_2 increases. The excess O_2 tend to react with CO and convert into CO_2. This can be seen from Figure. 4(c) in which the CO_2 increase with increase in equivalence ratio. It is evident from the results that an ER of less than 0.4 is favorable for higher H_2 and CO formation irrespective of SCR.

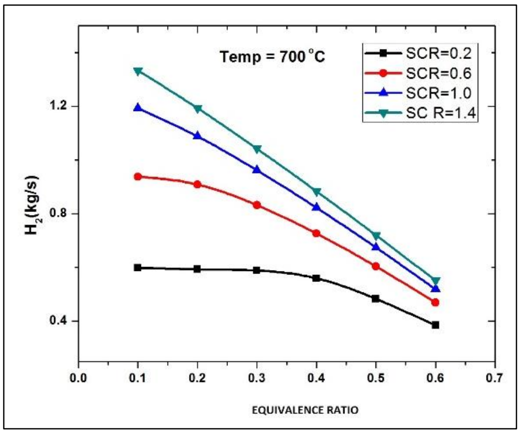

Figure 4(a): Equivalence Ratio Vs H_2 formation

Effect of Steam-Coal-Ratio

Figures 5(a) to 5(d) shows the effect of SCR on syngas components. The curves are plotted with equivalence ratio of 0.3 at four different temperatures ranging from 600 ºC to 900 ºC. From Figure. 5(a) it is observed that as SCR increases H_2 increases at all temperatures. Up to a certain SCR H_2 yield will increase due to decomposition of steam into hydrogen and oxygen. After that the rate of H_2 formation becomes slower. Figure. 5(b) represent the variation of CO with SCR at different temperatures. It is observed that as SCR increases, the CO is decreasing. This is due to fact that as SCR increases O_2 increases. This oxygen will react with CO forming CO_2. This is evident from the Figure. 5(c). A similar trend is show in H_2O formation which shown in Figure.5 (d). From the results it is seen that the favorable range of SCR is 0.2 to 0.6.

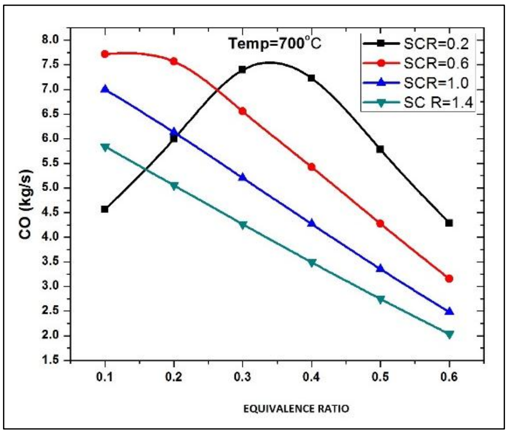

Figure 4(b): Equivalence Ratio Vs CO formation

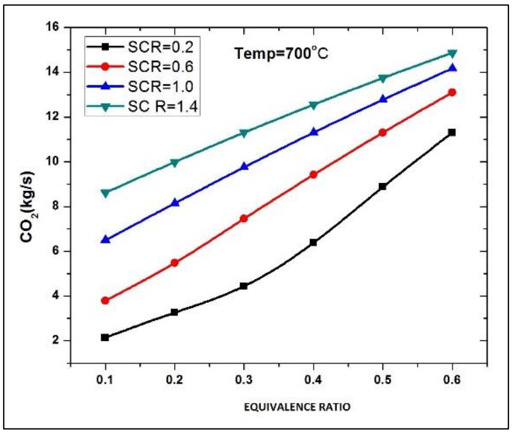

Figure 4(c): Equivalence Ratio Vs CO_2 formation

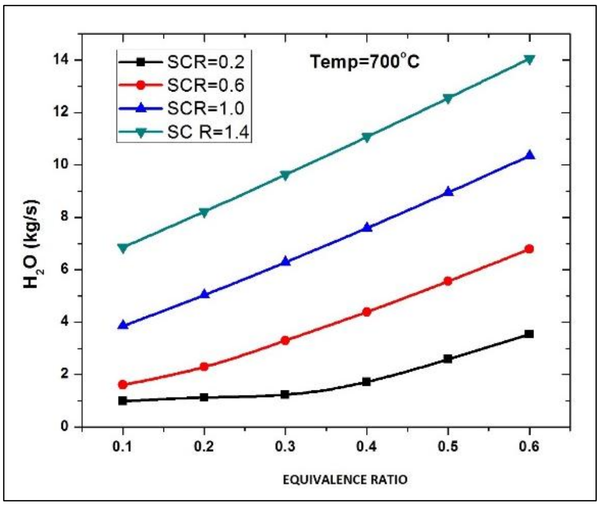

Figure 4(d): Equivalence Ratio Vs H_2O formation

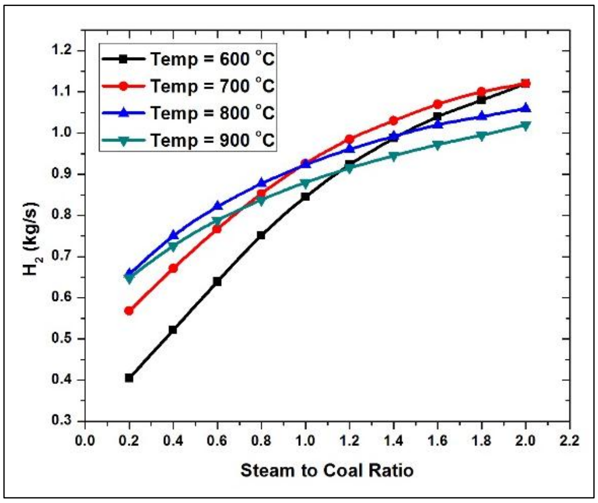

Figure 5(a): Steam-to- Coal ratio Vs H_2 formation

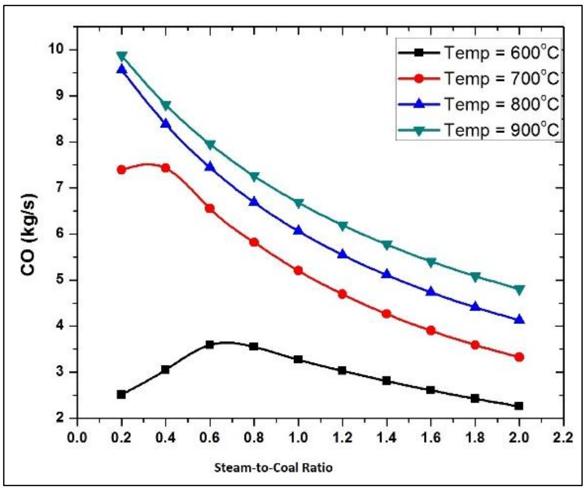

Figure 5(b): Steam-to- Coal ratio Vs CO formation

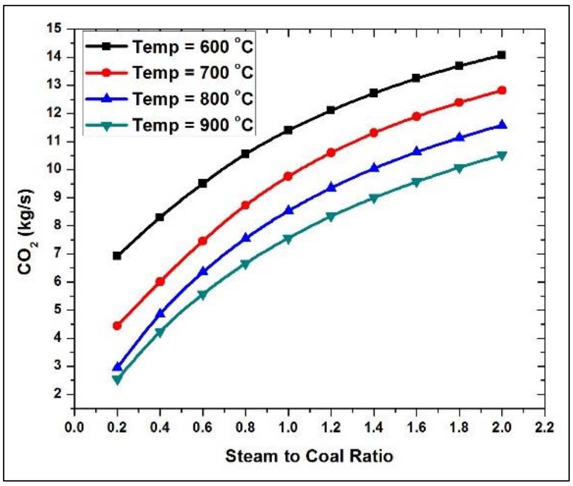

Figure 5(c): Steam-to- Coal ratio Vs CO_2 formation

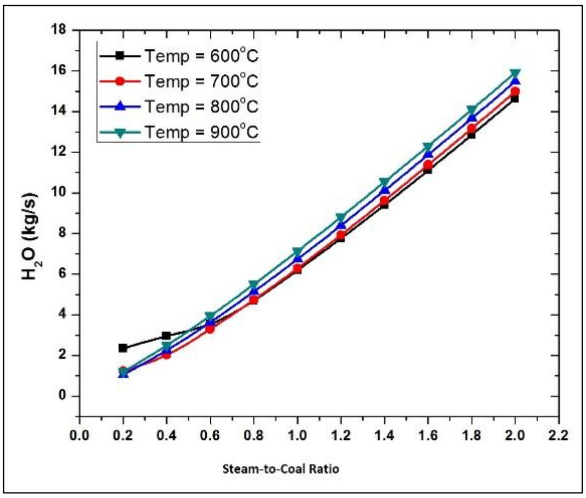

Figure 5(d): Steam-to- Coal ratio Vs H_2O formation

Effect of Gasifier Pressure

Effect of gasifier pressure on syngas components is shown in Figure. 6(a). The curves are plotted at an equivalence ratio of 0.2, SCR of 0.2 and a temperature of 600 ºC. The results show that H_2 and CO decreases with increase in pressure whereas CO_2, H_2O and CH_4 increases. The N_2 remains constant as it is only limited by the quantity present in the coal. Even though CH_4 is increasing with increase in gasifier pressure, H_2 and CO are decreasing. This results in decreasing of the HHV of syngas. Figure. 6(b) shows the variation of HHV with increase in gasifier pressure. Thus a lower pressure is suitable for higher HHV.

Effect of Gasifier temperature

Most significant parameter is gasification temperature because all reactions are temperature dependent. Figure. 7(a) to Figure.7 (f) are plotted at different SCR values ranging from 0.2 to 1.4.

Equivalence ratio is maintained in such a way that the temperature remains in a specified value. Figure. 7(a) shows the variation of H_2 with temperature. It indicates a sharp increase in quantity of H_2 initially followed by slow and gradual decrease beyond a certain temperature. Figure. 7(b) represents the variation of CO with temperature. It indicates that after a threshold temperature, CO increases steeply and reaches its maximum value. After that it remains almost stable. As the SCR increases the amount of CO decreases indicating that H_2 and H_2O formation is predominant than CO. The favorable temperature range for maximum amount of H_2 and CO is 600 to 900 ^oC.

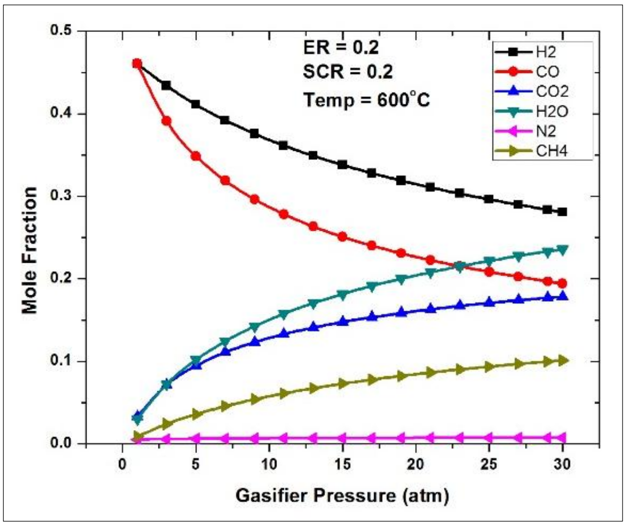

Figure 6(a): Gasifier Pressure Vs Syngas Components

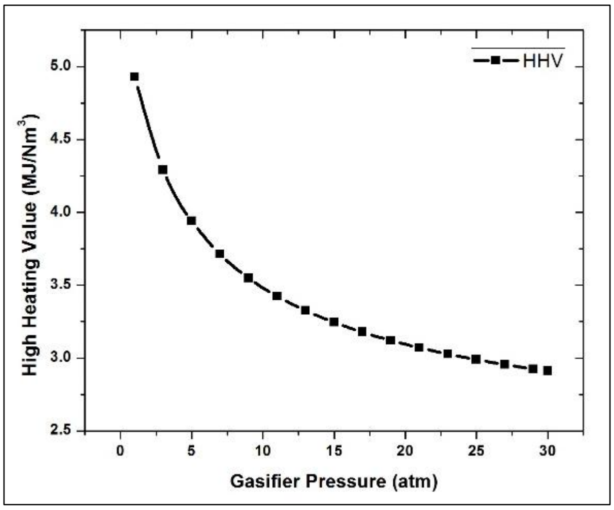

Figure 6(b): Gasifier Pressure Vs HHV

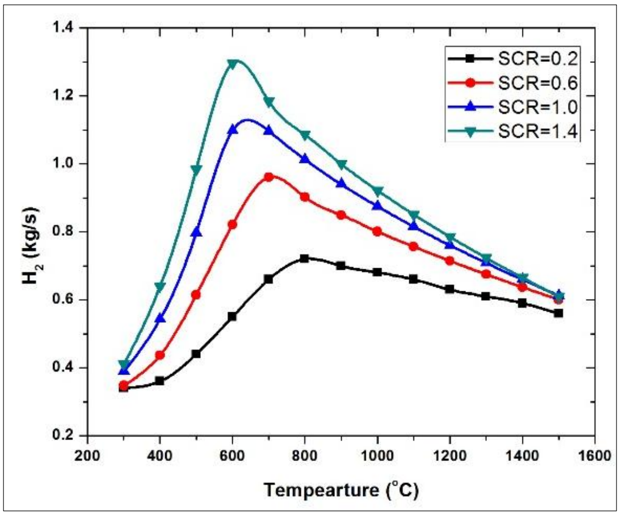

Figure 7(a): Temperature Vs H_2 formation

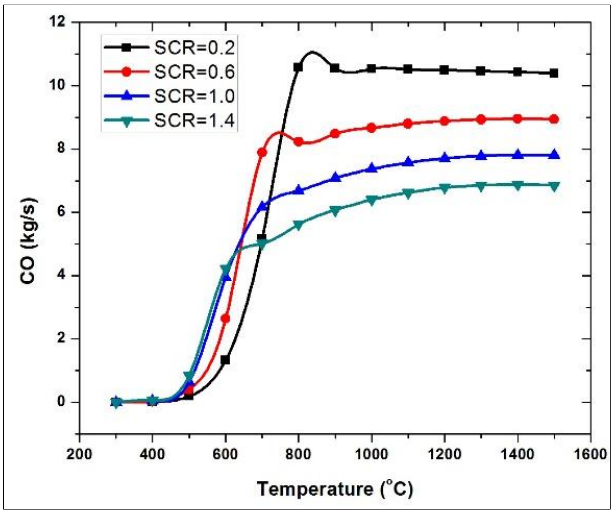

Figure 7(b): Temperature Vs CO formation

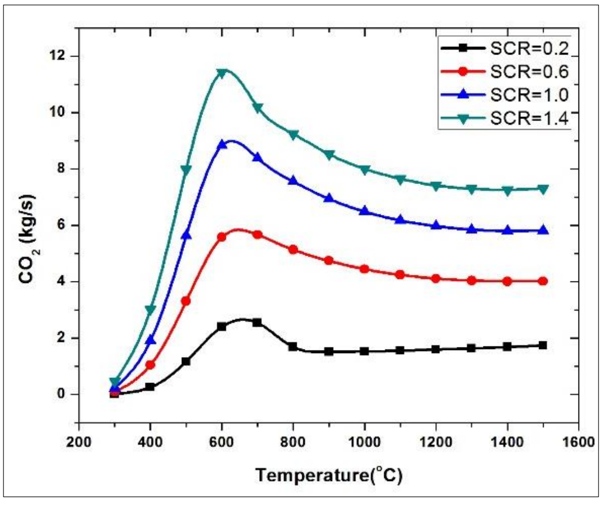

Figure. 7(c): Temperature Vs CO_2 formation

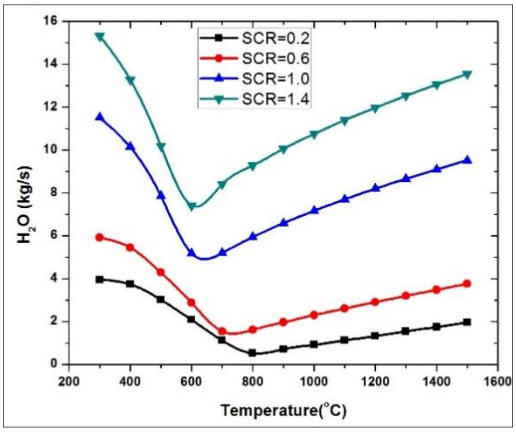

Figure 7(d): Temperature Vs H_2O formation

Figure. 7(e) depicts the char formation with temperature. Presence of char in syngas indicates incomplete carbon conversion. For maximum efficiency the char must be minimum / zero. It is clearly seen that a char formation occurs till a particular temperature and becomes zero after that. This temperature is threshold temperature of zero char formation and depends on SCR. Threshold temperature increases with decrease in SCR. However beyond 900 °C, char formation is zero irrespective of SCR.

Figure.7 (f) shows the variation of gasifier efficiency with temperature. Syngas HHV and gasifier efficiency are given in Eq. (5) and (6). The heat required for generating steam is supplied from gasifier only. Hence the total heat output of the system is the sum of syngas HHV and Q-Heater. The system efficiency is minimum at low temperature and raises steeply till a particular temperature and then starts decreasing gradually. It is also observed that as the SCR increases the maximum efficiency decreases which indicates that a lower SCR is favorable for higher efficiency.

$$HHV = (H_2 * 30.52 + CO * 30.18 + CH_4 * 95) * 4.1868\ MJ / (Nm^3) \quad (5)$$

$$Gasifier\ Efficiency = \frac{HHV_{Product\ gas} + Q_{Heater}}{GCV_{fuel} * \dot{m}_{fuel}}\ x\ 100 \quad (6)$$

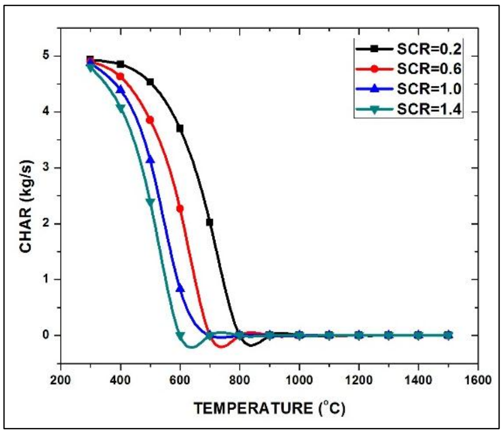

Figure 7(e): Temperature Vs Char formation

Process optimization

Optimization is performed on parameters air flow rate, steam flow rate, gasifier temperature and pressure. The objective is to maximize the HHV of syngas. The objective has been tested with and without a constraint of reduced CO_2 emissions. Table.6 shows the manipulated variables and their ranges used. The constraint of CO_2 mole fraction less than 1% is used. In both the cases equivalence ratio of 0.1 yielded maximum HHV. Table.7 shows the optimization results. Table. 8 shows the optimum parameter ranges for different objectives (higher combustible gases and lower emissions).

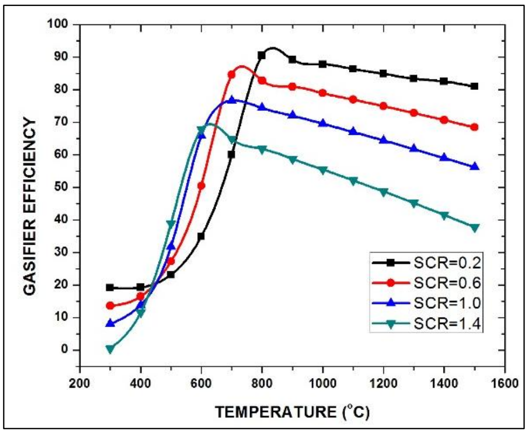

Figure 7(f): Temperature Vs Efficiency

Table 6: Optimum values for operating variables

Manipulated Variable	Range	Optimum Values	
		Without Constraint	With Constraint (CO_2 Mol Frac. < 1%)
ER	0.1 - 0.6	0.1	0.1
SCR	0 – 2	0.438	0.417
Gasifier Temp. (°C)	500 – 1500	918.006	1085.23
Gasifier Pres. (atm)	1 – 30	1	1

Table7: Optimization results

Objective	**Optimum Values**	
	Without Constraint	**With Constraint**
HHV (MJ/Nm^3)	6.1075	6.0805
CO_2 (Mole Frac.)	3.3167 x 10-2	8.22 x 10-3

Table 8: Optimum parametric domains for different objectives

	H_2 (↑)	CO (↑)	CO_2 (↓)	Gasfr. Eff. (↑)
ER	0.1 – 0.3	0.1 – 0.3	0.1 – 0.3	0.1 – 0.3
SCR	0.8 – 1.2	0.2 – 0.4	0.2 – 0.8	0.4 – 0.6
TEMP (°C)	600 – 800	600 - 800	800 – 1200	600 – 900

CONCLUSIONS

Gasification studies of Jharia coal were conducted using ASPEN Plus model. The effects of operating conditions ER, SCR, Gasifier temperature, Gasifier pressure are studied. The conclusions of the study are as follows;

ER, SCR, gasifier temperature are more significant than gasifier pressure. Syngas composition depends on the gasification atmosphere and effect of parameters are interdependent. To assess this synergetic effect LAFR is used. For each SCR and gasifier temperature there exists a LAFR.

H_2 formation is enhanced by increase in SCR whereas it is reduced by increase in ER and gasifier pressure. A lower ER and pressure are favorable for H_2 formation. From the results it can be concluded that an ER below 0.4 is suitable for maximum H_2 formation. Even though higher SCR promotes higher H_2 formation, beyond an SCR of 1.2 no significant increase H_2 quantity is observed. CO decreases with increase in ER and SCR whereas it is increased by increase in gasifier temperature. A lower ER and higher temperature is favorable for CO formation. Maximum amount of CO is formed in the temperature window of 600 to 900 °C.

The variation of CO_2 and H_2O is similar. As ER and SCR increases both CO_2 and H_2O increases. CO_2 increases with temperature till a maximum value and then starts decreasing. Whereas H_2O decreases with increase in temperature, reaches it minimum value and then starts increasing. CO_2 peak and H_2O trench occurs in the same temperature window. For maximum efficiency, a lower ER, SCR and higher temperature are favorable.

At lower temperatures carbon conversion is incomplete. A threshold temperature is to be maintained for char free gas which ranges between 600 and 900 °C depending on SCR. The HHV and hence the efficiency depends on the amount of H_2 and CO produced. The results indicate that the efficiency increases with temperature, reaches a maximum value then decreases gradually. Maximum efficiency occurs in the temperature range of 600 to 900 °C.

Optimization studies were conducted to obtain ideal combination of operating conditions with objective of maximizing HHV and minimizing CO_2 emissions. It is observed that the optimum value of equivalence ratio is 0.1 in both cases of with and without constraint on CO_2 emissions. The optimum value of steam-to-coal ratio is 0.417 and 0.438 with and without constraints respectively. The optimum value of temperatures are 1085.23 °C and 918.006 °C with and without constraint respectively.

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