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Novel Simulation and Optimization for the Production of Green Products I.E., Green Gasoline, Green Diesel and Green Waxes.

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Abstract: During the last decagon, there has been expanding international worry over the rise of anthropogenic CO_2 discharge into the Earth's atmosphere. The application of CO_2 into a valuable asset is a major concern. The generation of syn gas and then usage of the syn gas into liquid fuel, seems to be one of the promising options in terms of industrial employment, as it offers several advantages: (a) reduction of CO_2, (b) conversion of gases, specially bio-gas, natural gas, LPG, and etc. and CO_2 into syngas (c) producing syngas with H_2/CO_2 ratio 1:1.9 to 1:2.1 which may further be used for the generation of valuable petrochemicals. The present research focusses on the gas to liquid conversion using the simulating software, Aspen Plus ®. The outcomes are then subjected to Design Expert ® for calculation of the optimal generation rate. The feedstocks used for the proposed present examination are bio-gas or pyro-gas, natural gas and LPG. The research scheme, gas to liquid conversion is carried out using three steps: (a) gas (feedstock) to syngas from the combination of dry reforming and steam reforming of methane, (b) Fischer Tropsch process to produce long chains of hydrocarbons and (c) usage of unconverted CO and H_2 and other alcohol derivates in the CHP unit for the production of electricity. Amongst all the feedstocks natural gas production or generation is maximum followed by bio-gas or pyro-gas and then LPG. Due to non-available resources of natural gas and generation of GHG emission, for countries like India, bio-gas or pyro-gas can be used as a promising sustainable feedstock for reducing GHG emission and global warming. The outcomes of Aspen Plus ® of biogas or pyro-gas are then subjected to Design Expert ® for the prediction of the maximal production. It can be confirmed that with 6997.54 kg/h of biogas flowrate and 99.39% recycling of CO_2, the production of green gasoline, green diesel and green waxes are 565.24 (kg/h), 545.45(kg/h) and 642.68 (kg/h) respectively. The outcomes are in good agreement with the theories, thus proving the process to be a realistic one in nature. Therefore, bringing its viability for India in terms of reduction in CO_2 emission and development of gas to liquid conversion process.

Keywords : Dry reforming of methane, Aspen plus ®, green products i.e., green gasoline, green diesel and green waxes, CO_2 valorization, Fischer Tropsch

I. INTRODUCTION

Major socio-environmental concerns for recent time is greenhouse gas (GHG) emission and their impacts on climate change.

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According to the evidences (1), GHG emission has increases in the last decade in order to mollify the growing energy demand.

Without new developments GHG emission reduction is not possible and results in increase in the coming years. With this situation, alternatives based on the use of renewable energy resources are desired (2).

Amongst all the known renewable energy systems, those resources derived from biology are usually seen as convenient to avoid the usage of fossil fuels.

In generalized way, biogas or sometimes pyro-gas which is almost similar to biogas can provide a continual pathway for the production of valuable goods especially energy products and/or chemicals. Biogas is a mixture of gases regenerated by the breakdown of organic matter in anaerobic environment or fermentation processes(3). It primarily consists of methane (50 – 75%) and carbon dioxide (25 – 45%) and other minor components like hydrogen sulphide, nitrogen or water, varying the composition according to the substrate .(2),(3),(4) Biogas has the capability to reduce the need of fossil fuels and subsidize to satisfy with natural sustainability objectives. Moreover, the generation of biogas from organic waste can pave the way towards a bio-based and circular economy.(4) On the other hand, natural gas(5),(6) and pyro-gas(7) can also use for the generation of circular economy.

Natural gas, also called as fossil gas is a naturally occurring gas mixture primarily consisting of methane, but also includes varying amounts of other higher alkanes and traces of carbon dioxide, nitrogen, hydrogen sulphide or helium(6) whereas pyro-gas consisting of carbon dioxide, carbon monoxide, and methane (8). Pyro-gas sometimes are similar to biogas. So generally these two above mentioned gases (natural gas and pyro gas) also leads towards circular economy (1).

Now in recent time other gas namely, liquified petroleum gas or LPG (mixture of propane, butane and other gases) is used as an aerosol propellant (9) and a refrigerant(10), substituting chlorofluorocarbons in an attempt to reduce damage to the ozone layer, almost useful for generation of circular economy.

Apart from direct generation of heat and electricity (11), liquid biofuels can be produced through gas conditioning and cleaning, reorganizing to generate syngas, and the subsequent usage of Fischer Tropsch (FT) synthesis to produce long chain hydrocarbons. (12)

However, gas to liquid (GTL) process is challenging since the purification is costlier. (13) So, dry reforming of methane (DRM) is a suitable process for the production of syngas and then liquid biofuels (14),(12),(13) since CO_2 can act as a reactant which in return reduces the cost of purification therefore removal is not required. Hence, DRM succeeded by FT synthesis could be a suitable gas to liquid roadway to produce synthetic biofuels.(1)

Now in terms of GHG emission and non-renewable energy demand, methane and carbon dioxide are the major reactants of the DRM process with environmental advantages. (15) DRM process mainly composed of carbon monoxide and hydrogen and shows a molar ratio around 1.9:1 or 2.1:1, which is convenient for the generation or production of liquid fuels via FT synthesis.(16),(17) Additionally, this process (GTL) can be coupled with combined heat and power scheme for coproduction of electricity and turn into energy sufficient process. (18),(19),(20), (21), (22).

This research article focuses on the production of syngas from biogas or pyro-gas, natural gas and LPG (15), (23), (24), (25), (26), (27) and biofuel from syngas via the FT synthesis (4),(28), (29) and syngas, through an analysis of the combined DRM and FT system. The products generated are green gasoline, green diesel and green waxes and unconverted CO and H_2 and derivatives of alcohols. The derivates of alcohols and unconverted CO and H_2 are then subjected to combined heat and power system (30) for the production or generation of electricity. The aim is to calculate the maximum production rate of green products, mainly green gasoline, green diesel and green waxes using Aspen Plus® (31), (32) and Design Expert® (33) and generation of energy from the combined heat and power scheme (30). Though the design calculation does not give the real-life production environment but it can provide relief from making wide range of experiments without making the small-scale reactor plant.

II. METHODOLOGY

For simulation purpose, GTL (gas to liquid) are good methods to convert gaseous fuel to the synthetic liquid fuels such as diesel, gasoline and waxes. Nowadays, most of the process usages natural gas as feedstock but these research article tried to use biogas or pyro-gas and LPG as feedstocks, apart from natural gas and then made a correlation amongst themselves. The process proposed comprises of three units.

- The generation or production of syngas from the combination of dry reforming and steam reforming of methane
- The Fischer Tropsch process to produce long chains of hydrocarbons
- The usage of unconverted CO and H_2 and other alcohol derivates in the CHP unit for the production of electricity.

Originally the feedstock (pyro-gas or biogas, Natural Gas and LPG) is preheated and then subjected to the reforming reactor in which it combines with steam and CO_2. At the exit of the reformer the temperature of the syngas is greater than needed at the inlet of the Fischer Tropsch reactor. Thus, the synthesis gas is cooled and the remaining water is ejected. CO_2 gas present at the exit of the FT reactor is recycled back in order to make the process CO_2 emission free. The remaining gas is then subjected to the separation unit (distillation column) in order to recapture the different fractions of diesel, gasoline, waxes and some amount of unconverted CO and H_2 and other derivates of alcohol. The overall process flowsheet is represented in Figure 1.

A. Aspen Plus Modelling

Aspen Plus ® was utilized for the production or generation of green products i.e., green gasoline, green diesel and green waxes and some amount of unconverted CO and H_2 and alcohol derivates. It generally generates accurate results compared to the real life (34) and comprehensive thermodynamics for accurate determination of physical properties (35). The present simulation was investigated using ideal and Peng -Robinson models with Boston-Mathias alpha function (PR-BM) (32) which better suits for biogas or pyro-gas, natural gas and LPG. The components (gases) used for the simulation were pyro-gas, natural gas and LPG. Figure 1 represents the schematic diagram for the production of green products i.e., green gasoline, green diesel and green waxes and generation of electricity from CHP unit. The whole process simulation was investigated using the following assumptions:

- Process will be substantial state and isothermal.
- Biogas or pyro gas, Natural Gas and LPG comprises of CO, CO_2, H_2, CH_4, H_2O, propane, butane and etc.
- All the components are gases were used from the Aspen Plus ® database itself.
- All the stream lines that were used based on SI units.
- Peng -Robinson model and Ideal model fits the equation of state [EOS] (32)
- All the unit processes were based on SI units.

The consecutive compounds had been selected from the Aspen Plus ® database (31): oxygen (O_2), nitrogen (N_2), carbon monoxide (CO), carbon dioxide (CO_2), hydrogen (H_2), water (H_2O), methane (CH_4), ethylene (C_2H_4), ethane (C_2H_6), propylene (C_3H_6), propane (C_3H_8), butene (C_4H_8), butane (C_4H_{10}) and all linear and saturated hydrocarbons C_5H_{11} to $C_{30}H_{62}$, $C_{32}H_{66}$ and $C_{36}H_{74}$. And oxygenated compounds such as methanol (CH_4O), ethanol (C_2H_6O), 1-propanol (C_3H_8O), 1-butanol ($C_4H_{10}O$), 1-pentanol ($C_5H_{12}O$), 1-hexanol ($C_6H_{14}O$) and acetic acid ($C_2H_4O_2$). Saturated and linear hydrocarbons were selected to describe the gasoline, diesel and waxes: C_5 to C_{11}, C_{12} to C_{18} and C_{20} to C_{60}, respectively. For the thermodynamic model, the equation of Ideal state, Peng Robinson with Boston-Mathias alpha function (PR-BM)(32) was applied in the main units (the two reactors, the distillation columns). For water segregation, the NRTL (Non-Random Two Liquid) model is applied. All the binary interaction parameter values needed for these models were available in Aspen Plus ®. The detailed simulation comprises of syngas production unit, Fischer Tropsch (FT) unit, and finally Combined Heat and Power (CHP) scheme.

➢ **Process Description**

- **Syngas production Unit**

The reformer unit consists of two parts: a pre-reformer and a reformer. In the pre-reformer, hydrocarbons with higher molecular weight contained in the biogas or pyro-gas, natural gas and LPG are comprehensively converted into a mixture of methane, carbon oxides, and hydrogen over a nickel catalyst.

The temperature of the reaction is 823K and the outlet pressure is 5 bar (36). In the reformer the reaction temperature varies in a temperature range : [973K, 1273K] and range of the pressure: [1-5] bar (37). The reaction involved are:

$$CO_2 + CH_4 \rightarrow 2CO + 2H_2 \quad \Delta H^O_{298} = 247\, kJ\, mol^{-1} \quad (1)$$

$$CH_4 + H_2O \rightarrow CO + 3H_2 \quad \Delta H^O_{298} = 206\, kJ\, mol^{-1} \quad (2)$$

$$CO_2 + H_2 \rightarrow CO + H_2O \quad \Delta H^O_{298} = 41\, kJ\, mol^{-1} \quad (3)$$

For the simulation purpose, the assumed reforming unit is fed by 128.9 t/h of feedstock with 94.9% of methane and 330 t/h of CO_2 and 263.5 t/h of H_2O (37).

- **The Fischer Tropsch (FT) UNIT**

The synthesis gas generated by the reformer is cooled and water is removed through a flash column. The resultant stream is then subjected to the FT reactor for the production of long chain hydrocarbons. The catalyst used in the FT reactor is Cobalt (37). Generally, Fischer Tropsch synthesis requires a H_2/CO ratio between 1.9 to 2.1 (36) during the examination the H_2/CO ratio as 2 is maintained. For the better production FT reactor can work at a relatively low temperature, so this type of FT reactor is known as low temperature Fischer–Tropsch (LTFT)(37). The FT synthesis was carried by using RStoic (suitable for all purpose) reactor model at 513K and 20 bars. The conversion of synthesis gas was predicted at 87% (36). The major FT reactions are summarized as:

$$(2n+1)H_2 + nCO \rightarrow C_nH_{2n+2} + nH_2O \quad (4)$$

$$2nH_2 + nCO \rightarrow C_nH_{2n} + nH_2O \quad (5)$$

$$CO + H_2O \rightarrow CO_2 + H_2 \quad (6)$$

$$2nH_2 + nCO \rightarrow C_nH_{2n+2}O + (n-1)H_2O \quad (7)$$

Using the above reactions, a series of reactions was introduced taking into account the selectivity for each product at low temperature. In generalized format 42 reactions are generated which are efficiencies and selectivities are available from the literature, were considered to represent the process generated paraffins, olefins and some oxygenated compounds and WGS (Water Gas Shift) reactions (36).

- **Combined Heat and Power Schemes**

The unconverted CO and H_2 and other derivatives of alcohol obtained from the FT reactor is the subjected for power (electricity) generation. The thermal power plant is assumed to run at 30% (38) efficiency for fuel to electrical energy conversion. Waste heat recovery from exhaust flue gas is assumed to be at an efficiency of 70% (39).

B. Parametric Sensitivity AND OPTIMIZATION

The effect of parameters namely, biogas flowrate (A) and % of CO_2 recycled (B) are two major response variables, were correlated mathematically in this research work, The model equations were developed with the assistance of response surface methodology (40),(41),(42) varying the values of A and B simultaneously. The values of A and B were fixed using User Defined method (33), (43), (44).

The mathematical relationship between the response (Y_i) and factors, biogas flowrate (A) and % of CO_2 recycled (B) are given by

$$Y_i = f(A, B) \quad (8)$$

where i = 1,2

It was assumed that the independent factors A and B were continuous and controllable by experiments with negligible errors. The generalized second order polynomial, correlating the responses with the independent factors, is in the following form,

$$y_i = \alpha_i + \sum_{j=1}^{2} \beta_{ij} X_i + \sum_{j=1}^{2}\sum_{u=1}^{2} \beta_{iuj} X_u X_j + \sum_{j=1}^{2} \beta_{ijj} X_j^2 \quad u \neq j \quad (9)$$

The significance of the coefficients and the adequacy of the fit were determined using Student -t test and Fischer F-test respectively (44), (40). The values of flowrates of Biogas or pyro-gas were maximized respectively. The development of model equation and optimization has been done using Design -Expert Software 7.0 ® (33), (40), (42).

III. RESULTS AND DISCUSSION

After investigating the simulation, it was observed that the production or generation of green products namely, green gasoline, green diesel and green waxes is best for Natural gas, followed by bio-gas or pyro-gas and then LPG.

a. Production Profile

Figure 2 and Table A. represents the production or generation of green products namely, green gasoline, green diesel and green waxes from Biogas or pyro-gas, LPG and Natural gas. It is clearly evident from the figure and table, that the production of green products is maximum from Natural gas followed by Biogas or pyro-gas and LPG. The disadvantage of natural gas is that for countries like India we don't have ample reserves of natural gas which means that most of the natural gas that is consumed by us has to be brought from other nations. Such constant purchases can turn into a rather expensive proposition over time. Another disadvantage of natural gas is that it emits carbon dioxide which is turn pollutes the environment. Constant establishment of CO_2 into the environment will lead to climate change and global warming. Therefore, usage of pyro-gas or biogas is a promising for country like India.

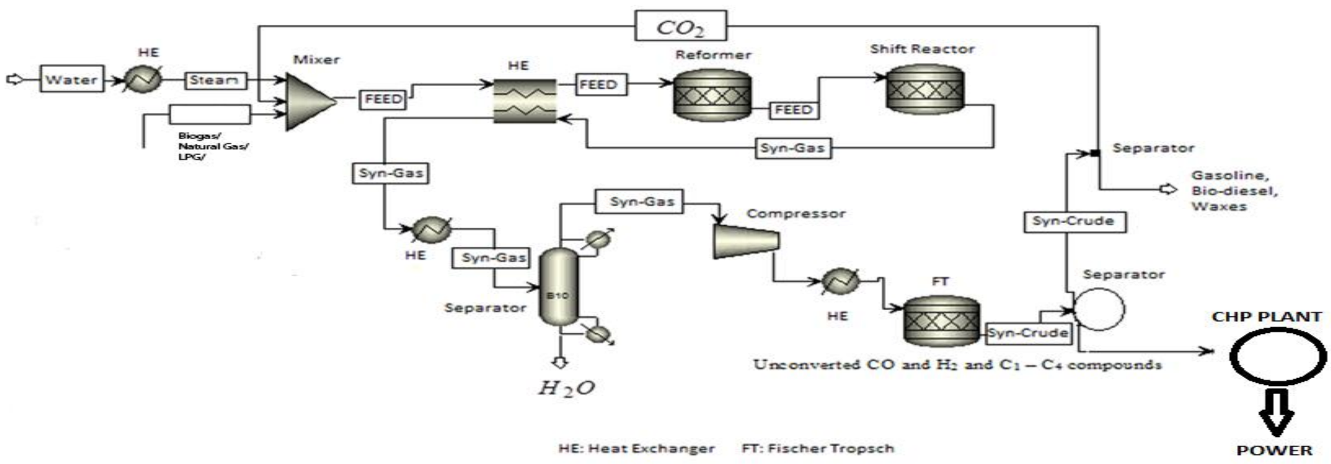

Figure – 1 Schematic diagram for the production of green products i.e., green gasoline, green diesel and green waxes and generation of electricity from CHP unit

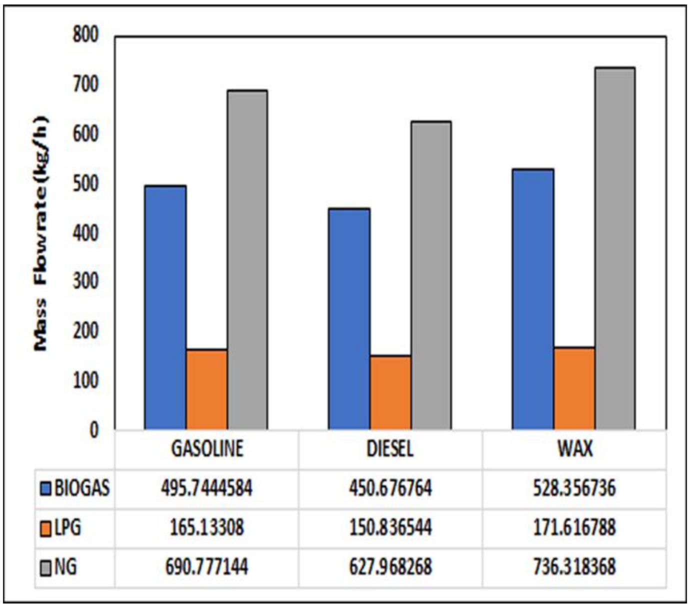

Figure – 2 "Variation of mass flowrate for green - gasoline, diesel and waxes

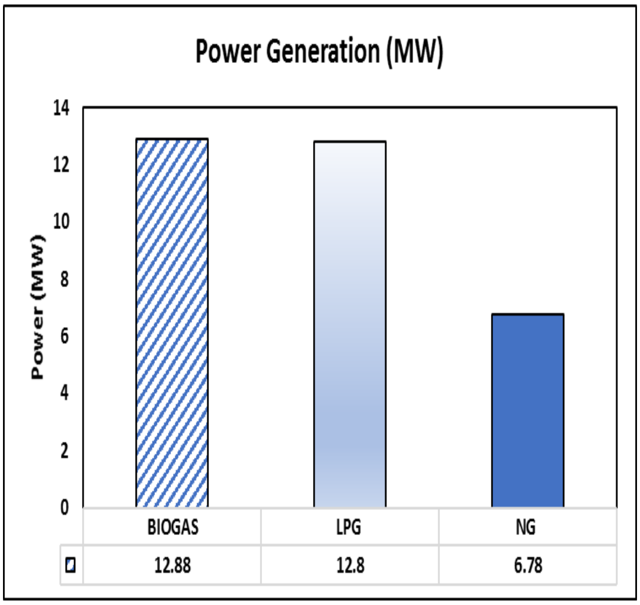

Figure -3 "Power generation from CHP plant"

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Table A. Variation of mass flowrate for green -gasoline, diesel and waxes (kg/h)

Gaseous feedstock	Green gasoline production (kg/h)	Green diesel production (kg/h)	Green waxes production (kg/h)
Bio-gas or pyro-gas	495.74446	450.67676	528.35674
Natural Gas	690.77714	627.96827	736.31837
LPG, Liquified petroleum gas	165.13308	150.83654	171.61679

Table B. Variation in production of power generation from CHP plant

Gaseous feedstock	Power Generation (MW)
Bio-gas or pyro-gas	12.88
Natural Gas	12.8
LPG, Liquified petroleum gas	6.78

Figure 3 and Table B, represents the production of power generation from CHP plant. From the figure and table, it is clearly evident that the production of electricity is maximum for biogas or pyro-gas followed by LPG (Liquified Petroleum Gas) and Natural Gas (NG). It also confirms that production of electricity in case of Natural gas is minimum and due to this reason also, the selection of bio-gas or pyro-gas proves to be the suitable gas for the production of green products namely, green gasoline, green diesel and green waxes for countries like India.

B. Sensitivity analysis

After selecting the bio-gas or pyro-gas as a promising feedstock for GTL process, the flowrates were varied from 5000 kg/h to 7000 kg/h with % of CO_2 recycled varied from 0 to 100.

Figure 4, 5, and 6 represents the variation of gasoline flowrate as a function of biogas flowrate and % of CO_2 recycled, variation of green -diesel flowrate as a function of biogas flowrate and % of CO_2 recycled and Variation of waxes flowrate as a function of biogas flowrate and % of CO_2 recycled respectively. From the ANOVA tables, Table 1, 2 and 3 the probability values were less than 0.0001, which suggests that the model fit for the maximum production of green products namely, green gasoline (Figure 4 and Table 1), green diesel (Figure 5 and Table 2) and green waxes (Figure 6 and Table 3). Equations 10, 11 and 12 suggests that the process is a
surface quadratic type, since the significant terms of the equation ends at square terms.

From the Anova Table, User Defined Method suggests that the R^2 values for gasoline, diesel and waxes are 0.9972, 0.9972, and 0.9941 respectively. It also suggests that the Adj. R-squared values is greater than the Pred. R- squared values, which makes the model feasible. The Adeq Precision is also in the suitable range; thus, the model is in reasonable agreement for the development of process in country like India.

Hence from figure 4, Table 1 and equation 10, it can be confirmed that with 6997.54 kg/h biogas flowrate and 99.39% recycling of CO_2 the maximum production of gasoline is 565.24 (kg/h). Subsequently, from figure 5, Table 2 and equation 11 and figure 6, Table 3 and equation 12 it can be proved that with the same biogas flowrate (6997.54 kg/h) and same recycling of CO_2 (99.39%), the maximal production of diesel and waxes are 545.45(kg/h) and 642.68 (kg/h) respectively.

IV. CONCLUSION AND RECOMMENDATION FOR FUTURE SCOPE

In recent occasion, need for conversion of gas to liquid is increasing for minimizing the GHG emission and reducing global warming. Various process have been tried by researchers, namely, thermal conversion (45), (46), (47) and biological conversion (48), (49) and etc. for treatment of biomass but few process have been tried (50),(51),(52) for gas to liquid. So, the demand for gas to liquid conversion in industrial scale is demanding. The present research is carried out for Gas to liquid conversion by DRM (Dry reforming of methane) followed by FT (Fischer Tropsch) synthesis for the production or generation of green products, i.e., green gasoline, green diesel, green waxes, some alcohol derivatives and some unconverted CO and H_2. During the simulation various feedstocks namely, bio-gas or pyro-gas, natural gas and LPG are considered in order to make a contrast amongst themselves and also to find the best suitable gas for feedstock in DRM method for countries like India. The process parameters were selected according to the previous researchers (37) following three steps: (a) gas (feedstock) to syngas from the combination of dry reforming and steam reforming of methane, (b) Fischer Tropsch process to produce long chains of hydrocarbons and (c) usage of unconverted CO and H_2 and other alcohol derivates in the CHP unit for the production of electricity.

The process was simulated using Aspen Plus ®. Amongst the selected feedstocks, the production or generation of Natural gas is maximum, followed by bio-gas or pyro-gas and then LPG, liquified petroleum gas. But for countries like, India, bio-gas or pyro-gas proves to be promising because of its wide availability. The simulated outcomes of Aspen Plus ® of biogas or pyro-gas are then subjected to Design Expert ® for the prediction of the maximal production. It can be confirmed that with 6997.54 kg/h of biogas flowrate and 99.39% recycling of CO_2 the production of gasoline, biodiesel and waxes are 565.24 (kg/h), 545.45(kg/h) and 642.68 (kg/h) respectively. The outcomes are in good agreement with the theories, thus proving the process to be a realistic one in nature. Therefore, bringing its growth for countries, like, India. Further advancement lies in the LCA development, exergy analysis and exergoeconomic, technical development and techno-economic feasibility of the process.

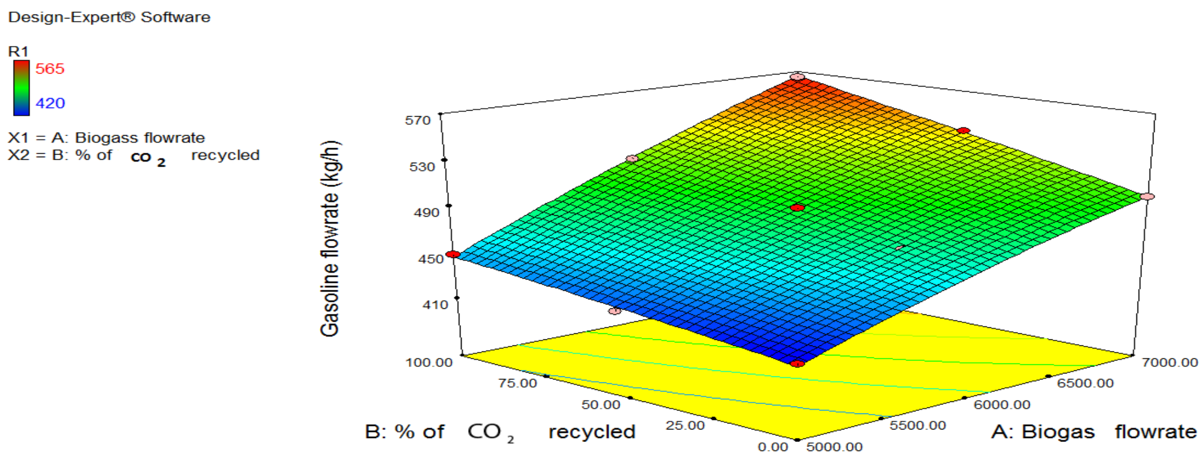

Figure 4 "Variation of gasoline flowrate as a function of biogas flowrate and % of CO_2 recycled"

$$\begin{aligned} Gasoline\ flowrate(kg/h) = 42.19888 + 0.099371 * Biogasflowrate \\ - 0.55558 * \%\, of\ CO_2\ recycled \\ + 1.80000E - 004 * Biogas\ flowrate * \%\, of\ CO_2\ recycled \\ - 4.85040E - 006 * Biogassflowrate^2 \\ - 4.84070E - 004 * \%of\ CO_2\ recycled^2 \end{aligned} \quad (10)$$

Table -1 ANOVA for response surface quadratic model for the production of gasoline as a function of biogas flowrate and % of CO_2 recycled

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F	
Model	17549.62	5	3609.92	478.70	< 0.0001	significant
A-Bio-gas flowrate	15100.17	1	15100.17	2029.80	< 0.0001	
B-% of CO_2 recycled	2679.13	1	2879.13	387.02	< 0.0001	
AB	324.00	1	324.00	43.55	0.0003	
A^2	55.83	1	55.83	7.50	0.0289	
B^2	3.07	1	3.07	0.41	0.5211	
Residual	52.07	7	7.44	498.70	< 0.0001	

R-Squared: 0.9972, Adj R-Squared: 0.9952, Pred R-Squared: 0.9723, Adeq Precision: 79.806

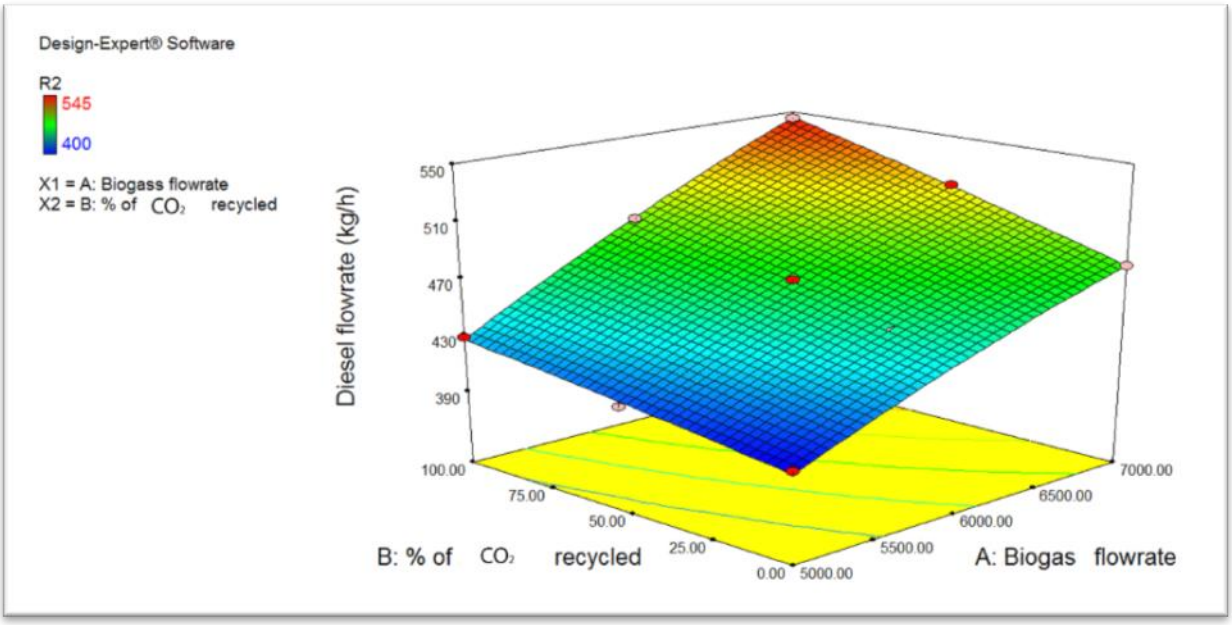

Figure – 5 "Variation of green -diesel flowrate as a function of biogas flowrate and % of CO_2 recycled"

$$\begin{aligned} Diesel\ flowrate(kg/h) = 2.31671 + 0.10638 * \text{biogas flowrate} \\ - 0.60726 * \%\,\text{of}\ CO_2\ recycled \\ + 1.8E - 4 * Biogas\ flowate * \%\ of\ CO_2\ recycled \\ - 5.43466E - 6 * Biogas\ flowrate^2 \end{aligned} \quad (11)$$

Table -2 ANOVA for response surface quadratic model for the production of green diesel as a function of biogas flowrate and % of CO_2 recycled

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F	
Model	18549.62	5	3709.92	498.70	< 0.0001	significant
A-Bio-gas flowrate	15100.17	1	15100.17	2029.80	< 0.0001	
B-% of CO_2 recycled	2879.13	1	2879.13	387.02	< 0.0001	
AB	324.00	1	324.00	43.55	0.0003	
A^2	55.83	1	55.83	7.50	0.0289	
B^2	3.07	1	3.07	0.41	0.5411	
Residual	52.07	7	7.44	498.70	< 0.0001	

R-Squared: 0.9972, Adj R-Squared: 0.9952, Pred R-Squared: 0.9723, Adeq Precision: 79.836

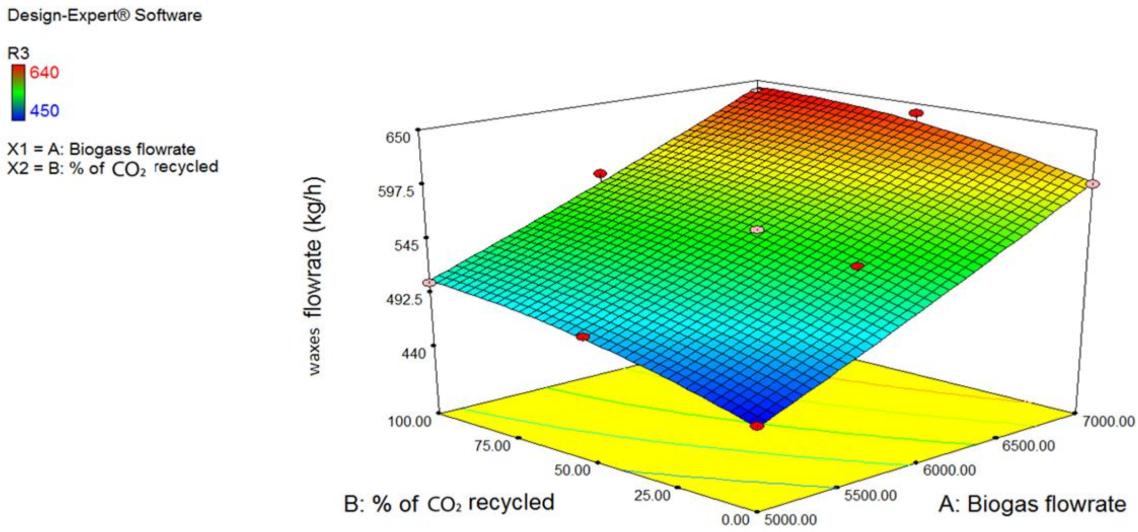

Figure -6 "Variation of waxes flowrate as a function of biogas flowrate and % of CO_2 recycled"

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\begin{aligned}
waxe\ flowrate(kg/h) = &+172.91123 \quad +0.040203 * Biogas\ flowrate \\
&+1.36255 * \%\ of\ CO_2\ recycled \\
&-7.50000E-5 * Biogas\ flowrate * \%\ of\ CO_2\ recycled \\
&+2.99003E-6 * Biogass\ flowrate^2 \\
&-4.17308E-3 * \%\ of\ CO_2\ recycled^2
\end{aligned}
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(12)

Table -3 ANOVA for response surface quadratic model for the production of waxes as a function of biogas flowrate and % of CO_2 recycled

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F	
Model	34615.10	5	6923.02	236.87	< 0.0001	significant
A-Bio-gas flowrate	31392.67	1	31392.67	1074.07	< 0.0001	
B-% of CO_2 recycled	3116.50	1	3116.50	106.63	< 0.0001	

AB	56.25	1	56.25	1.92	0.2079	
A^2	21.22	1	21.22	0.73	0.4224	
B^2	228.13	1	228.13	7.81	0.0268	
Residual	204.59	7	29.23			

R-Squared: 0.9941, Adj R-Squared: 0.9899, Pred R-Squared: 0.9439, Adeq Precision: 52.872

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CONFLICTS OF INTEREST

There are no conflicts to declare

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