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A Statistical Analysis on Tar Reduction in Producer Gas for IC Engine Application

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Abstract

The automobile sector attained a boom in the last decade; this made the necessity for more fuels consumption. To meet the demand, many alternate fuel systems are introduced. Among that, producer gas blend carbonated fuels are highly recommended. Since, it is adaptive to the current Internal Combustion (IC) engines with modified carburetor. This paper aims to investigate the production of producer gas from wood and the methods to filter the tar content in it. The chemical-catalytic method is the cost and eco effective way to reduce the tar in producer gas. Therefore, a new Ni based TiO_2 supported catalysts are synthesized to improve the efficiency of the tar cracking. The catalytic system is affected by other factors such as Bed Temperature (BT), Catalyst Weight (CW), Gas Feed Rate (GFR) and Gas Residence Time (GRT). Design of Experiments (DOE) is framed with L_{27} design table and the response is optimized using Taguchi methodology. The derived regression equation attained 98.4% of adequacy in response prediction and the identified optimaltar cracking efficiency is 99.46%forNi-Pr/TiO_2catalyst.

1. Introduction

In recent years, the research on Internal Combustion (IC) engine has beenfocused on renewablefuels instead of conventional fossil fuels to control the increasing environmental concerns like air pollution (Kan et al. 2018). The commonly preferred renewable fuel for automobiles with IC engine is gaseous fuel, which is produced through biomass gasification process. Since, its physical impedimentis insignificant. Apart from this kind of fuel, alcohol mixed fossil fuel (Wulff et al. 2000), dual fuel (Brusca et al. 2014), tri fuel (Kumaran et al. 2013), natural organic oil mixed fossil fuel (Johnet al. 2017) and so on are examined but the

better commercial and productive alternate for fossil fuels are not identified. Since, these alternate fuels are supplied through intakeair manifold;it displaces the equivalent amount of air and results invery lessengine volumetric efficiency (Okoronkwo et al. 2017).

While considering the gaseous fuel through biomass gasification process, the tar generation is the terrible factor creating many consequences to the engine. Therefore, tar cracking is a vital process to be carried out before fuel usage. El-Rub et al. (2008) stated that char behaves as a good catalyst for tar cracking as effective as Nickel (Ni) and dolomite with bulk density of 260Kg/m^3.Akiaet al. (2014) stated that the bulk catalyst suffers from disadvantages such as mass transfer resistance, larger time requirement for tar cracking, faster deactivation and creates disposal problems. Hence, nano catalysts are being employed in tar cracking process to overcome the above stated disadvantages of bulk catalysts. Chan andTanksale(2014) stated that the nanocatalyst was expected to perform better than bulk catalysts as they have higher number of active sites per gram. In addition they possess higher surface area and associated higher catalytic activity.

Many researchers worked on nanocatalytic tar cracking systembut still, the tar cracking efficiency is not achieved entirely. Since, the tar generation is not a predictable through selective factors. Therefore, a high performance optimal tar cracking procedure is requirement. In this research article, the Ni based transition metal supportcatalysts are reinforced with Cerium (Ce) and Praseodymium (Pr) to improve the efficiency of the catalytic activity in tar cracking. In further, the factors influencing tar formation in downdraft gasifier and catalytic system are optimized using Taguchi methodology.

2. Methodology

2.1. Synthesis of Nano Ni/TiO_2 Based Catalyst

The silica gel is synthesized from condensation of Titanium Tetra IsoPropoxide$Ti\{OCH(CH_3)_2\}_4$(TTIP), hydrochloric (HCl) acid and ethanol in the ratio 1:0.25:6 respectively at 60°C for one hour. The hydrated nickel nitrate (Ni $(NO_3)_2.6H_2O$) and silica are dissolved in deionized water in the mole fraction of 0.15:0.85. Further, CetylTrimethyl Ammonium Bromide (CTAB) (2.1×10^{-4} mol/l) is also dissolved and allowed to constant stir. After complete mixing, remove the possible absorbed ions and chemicals. Then it is dried in hot air oven at 120°C for two hours at heating rate of 10°C/min. Dried samples are calcined in muffle furnace at 600°C for six hours at heating rate of 20°C/min. This process resulted in formation of Ni/TiO_2 nano structured catalyst. The Ni $(NO_3)_2.6H_2O$

is replaced by Cerium nitrate ($Ce(NO_3)_3.6H_2O$) and Praseodymium nitrate ($Pr(NO_3)_3.6H_2O$) for obtaining Ni-Ce/TiO_2, and Ni-Pr/TiO_2nano catalyst. The obtained powders are pulverized and pelletized. The photographs of the Ni/TiO_2, Ni-Ce/TiO_2, and Ni-Pr/TiO_2nano catalyst pellets are shown in Figure 1 (a-c)respectively.

Figure 1 Photographs of Synthesized Nano Structured Catalysts (a) Ni/TiO_2 (b) Ni–Ce/TiO_2 and(c) Ni-Pr/TiO_2

2.2. Characterization of Nano Ni/TiO_2 Based Catalysts

2.2.1. Surface Morphology

Further, the pellets of synthesized catalyst are analyzed using Scanning Electron Microscopy (SEM-JOEL, JSM-5600 model). The formation of NiO, CeO_2, Pr_2O_3onTiO_2supports are evidenced from Figure 2.The Scanning Electron Microscopy (SEM) images reveal a high yield of spherical Ni-Ce/$TiO_2$and Ni-Pr/TiO_2nano particleswith diameters of 25 nm, which are self-assembled without agglomeration.

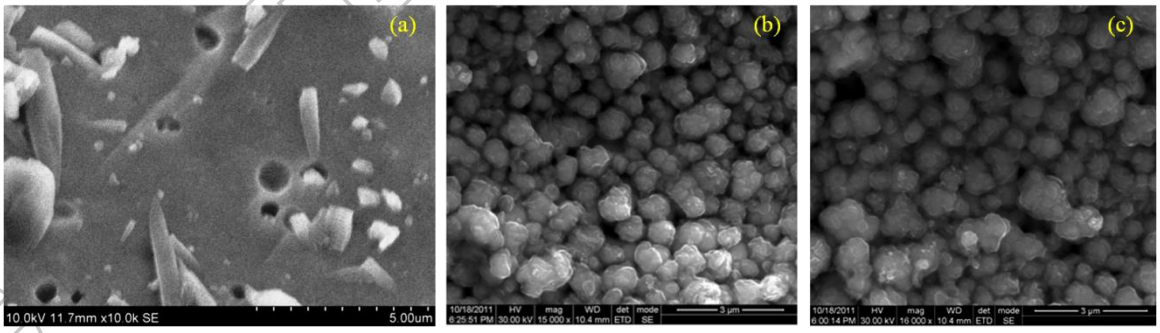

Figure 2 Surface Morphology of Synthesized Nano Structured Catalysts (a) Ni/TiO_2 (b) Ni–Ce/TiO_2 and(c) Ni-Pr/TiO_2

2.2.2. Surface Area Analysis

The surface area and pore volume are calculated by Brunauer- Emmett- Teller (BET) and Barrett-Joyner-Halenda(BJH) methods respectively.The surface area of the catalyst is

evaluated using the Micrometrics ASAP 2020 model and the sample is outgassed at 200°C for 12 hours. The surface area of Ni/TiO_2 and pore volume of nano catalysts is found to be $81m^2/g$ and $0.089990cm^3/g$ respectively. Similarly, the surface area and pore volume of the Ni-Pr/TiO_2nano catalyst is found to be $110m^2/g$ and $0.197400cm^3/g$ respectively. The surface area of Ni-Pr/TiO_2nano catalyst is 26.3% higher than Ni/TiO_2nano catalyst. Hence, it is presumed that the tar cracking ability of the Ni-Pr/TiO_2nano catalyst would be higher than Ni/TiO_2nano catalyst.

2.3. Design of Experimentation in Bio Gasifier

The downdraft, dry bottom fixed bed gasifier and the catalytic tar cracking system are fabricated as shown in Figure 3.

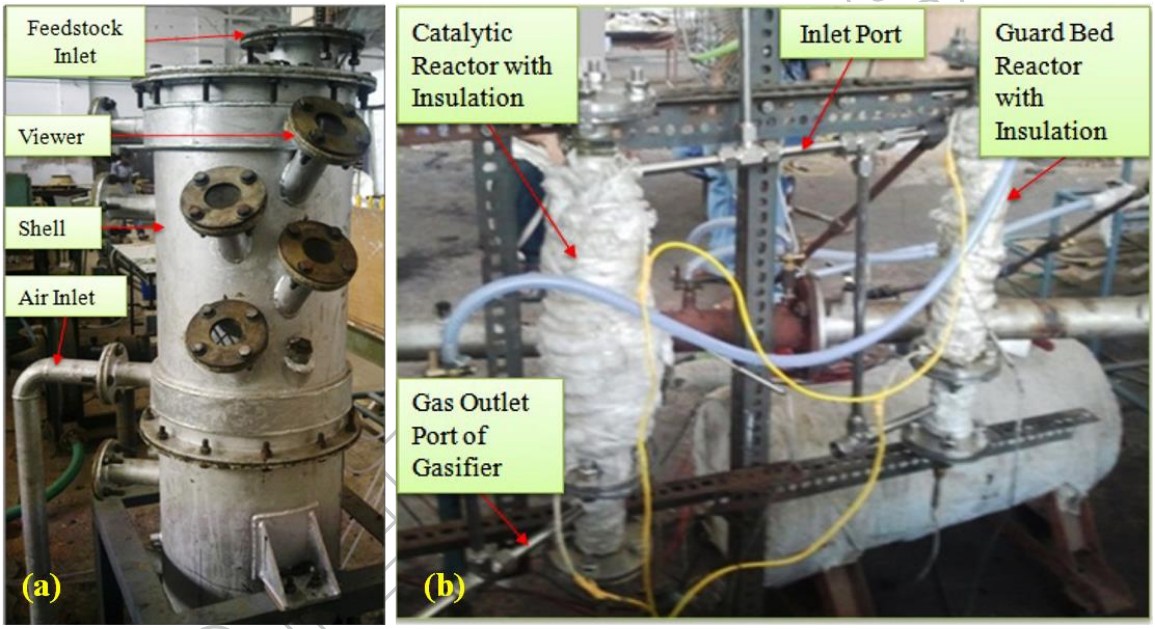

Figure 3 Photographs of (a) Downdraft Gasifierand (b) Catalytic Tar Cracking System

Casuarina wood is used as the feedstock at the rate of 6 kg/h for 24 kg full load capacity in gasifier. The catalytic tar cracking unit is placed downstream of the gasifier. It consists of a guard bed containing 100 g of crushed Dolomite ($CaMgCO_3$) stones and the main catalytic reactor containing synthesized nano catalysts. Both the guard bed and main catalytic reactor are wound with electrical resistance heating coils to maintain the desired bed temperature (Ramasubramanian and Chandrasekaran 2018). The tar content of producer gas is measured by sampling unit as per the guidelines of international protocol for measurement of organic contaminants in producer gas (CEN BT/TF 143, 2005).

The catalytic system is influenced by Catalyst, Bed Temperature (BT),Catalyst Weight (CW), Gas Feed Rate (GFR) and Gas Residence Time (GRT) in optimizing the tar concentration. Design of Experimentation (DOE) is framed and optimized using Taguchi methodology. The factors and their levels are tabulated in the Table 1. Based on Table 1, the orthogonal (L_{27}) design table is selected and it is shown in Table 2.

Table 1 Factors Influencing Catalytic System

Variable	Factor	Unit	Level		
			Low (-1)	Middle (0)	High (+1)
A	Catalyst (Ni/TiO_2)		Neat	Ce	Pr
B	Bed Temperature (BT)	°C	700	775	850
C	Catalyst Weight (CW)	g	3	6	9
D	Gas Feed Rate (GFR)	l/s	0.01	0.02	0.03
E	Gas Residence Time (GRT)	s	1	2	3

3. Results and Discussion

3.1. Statistical Analysis on Tar Concentration

The statistical analysis of catalytic system on tar concentration is done and tabulated in Table 2 as per the L_{27} design table. The residuals of the run are plotted in Figure 4 to estimate the normality of the selected runs/trails. The normality plot falls on a linear trend, which reveals that the selected data are uniformly distributed with respect to the catalyst.

Table 2 Statistical Analysis of Influencing Factors as per L_{27} Design Table

S. No.	Catalyst	Bed Temperature (°C)	Catalyst Weight (g)	Gas Feed Rate (l/s)	Gas Residence Time (GRT) (s)	Tar Concentration (mg/Nm^3)
1	Neat	700	3	0.01	1	0.857
2	Neat	700	3	0.01	2	0.832
3	Neat	700	3	0.01	3	0.81
4	Neat	775	6	0.02	1	0.822
5	Neat	775	6	0.02	2	0.794

6	Neat	775	6	0.02	3	0.762
7	Neat	850	9	0.03	1	0.711
8	Neat	850	9	0.03	2	0.687
9	Neat	850	9	0.03	3	0.621
10	Ce	700	6	0.03	1	0.397
11	Ce	700	6	0.03	2	0.381
12	Ce	700	6	0.03	3	0.36
13	Ce	775	9	0.01	1	0.251
14	Ce	775	9	0.01	2	0.238
15	Ce	775	9	0.01	3	0.218
16	Ce	850	3	0.02	1	0.345
17	Ce	850	3	0.02	2	0.324
18	Ce	850	3	0.02	3	0.29
19	Pr	700	9	0.02	1	0.228
20	Pr	700	9	0.02	2	0.21
21	Pr	700	9	0.02	3	0.198
22	Pr	775	3	0.03	1	0.212
23	Pr	775	3	0.03	2	0.19
24	Pr	775	3	0.03	3	0.175
25	Pr	850	6	0.01	1	0.182
26	Pr	850	6	0.01	2	0.151
27	Pr	850	6	0.01	3	0.12

Based on the data plot, the mathematical models are developed for the Ni/TiO_2, $Ni–Ce/TiO_2$ and$Ni-Pr/TiO_2$catalysts (Equation 1-3). From the first order equations, the regression

models are derived with adequacy of 99.12% i.e., 98.86% of R^2_{adj} and 98.40% of R^2_{pred} values.

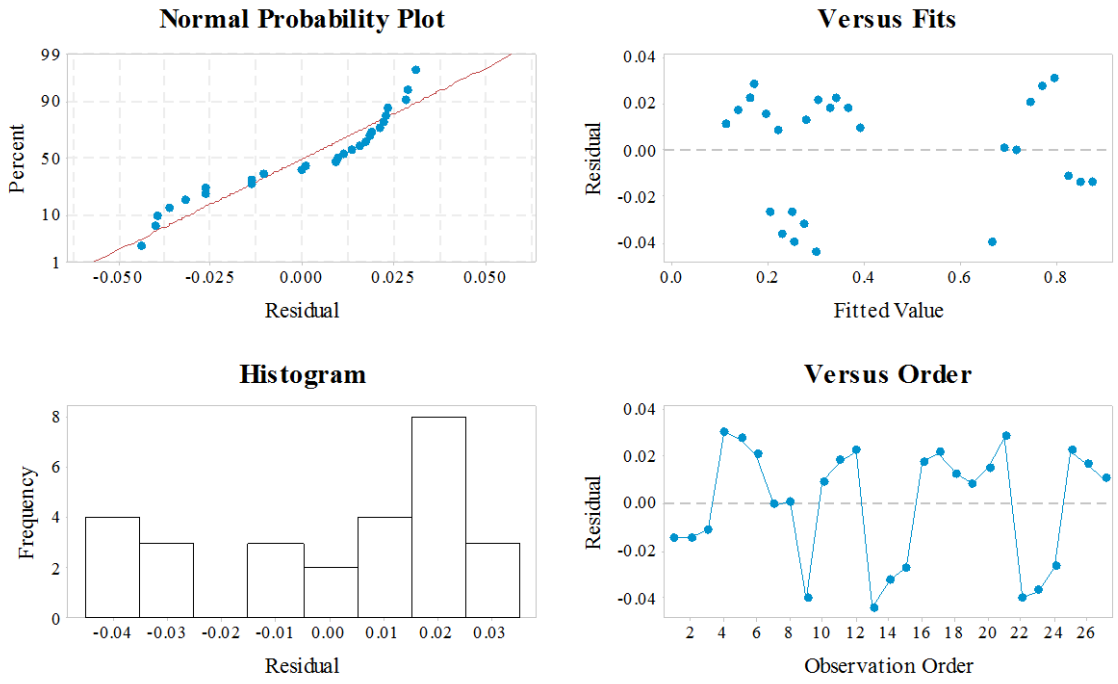

Figure 4 Residual Plots on Tar Concentration

$$Neat_{Tar\ Concentration}=1.3661 - 0.000624\ BT - 0.01246\ CW + 0.417\ GFR - 0.02506\ GRT \quad (1)$$

$$Ce_{Tar\ Concentration}=0.9115 - 0.000624\ BT - 0.01246\ CW + 0.417\ GFR - 0.02506\ GRT \quad (2)$$

$$Pr_{Tar\ Concentration}=0.7850 - 0.000624\ BT - 0.01246\ CW + 0.417\ GFR - 0.02506\ GRT \quad (3)$$

Table 3 Analysis of Variance (ANOVA)

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Regression	6	1.75736	0.292894	376.41	0.000
BT	1	0.03939	0.039387	50.62	0.000
CW	1	0.02516	0.025163	32.34	0.000
GFR	1	0.00031	0.000313	0.40	0.033
GRT	1	0.01130	0.011300	14.52	0.001
Catalyst	2	1.68120	0.840600	1080.29	0.000
Error	20	0.01556	0.000778		

Total	26	1.77292			

Analysis of Variance (ANOVA) is carried out to evidences the level of contribution and confidence of factors (Saravanan et al 2015)and it is tabulated in Table 3 for the data. The student trail error (P-Value) is less than the F-value and also it is lesser than 0.05 (5%), which states that the level of confidence is 95%. Since the level of confidence is above 95%, the selected factors are significant to the system. From the regression form, the most contributing factors on response are identified as catalyst and bed temperature compared others.

3.2. Taguchi Methodology for Optimization of Catalytic System

Taguchi methodology working on the principle of ranking the influencing factors based on its levels and contribution towards the response (Baradeswaran et al 2013). The contour plot is drawn in Figure 5 to study the major influencing factors i.e. significant and insignificant factors. The Figure 5 reveals that the increase in level for all factor decreases the tar concentration i.e. increases in the tar cracking percentage.

The major approaches to identify the contribution rate of the factors on response are mean and Signal to Noise Ratio (SN ratio). The response table for Signal to Noise Ratios is tabulated in Table 4 and graphically represented in Figure 6 with considering “smaller is better” ideology. Similarly, the response table for means is tabulated in Table 5 and graphically represented in Figure 7. Both the approaches state that the catalyst and bed temperature are the most influencing factors on response tar concentration. This result agrees with the ANOVA result. The other two factors catalyst weight and gas feed rate are mutually contributing to an extent of significance.

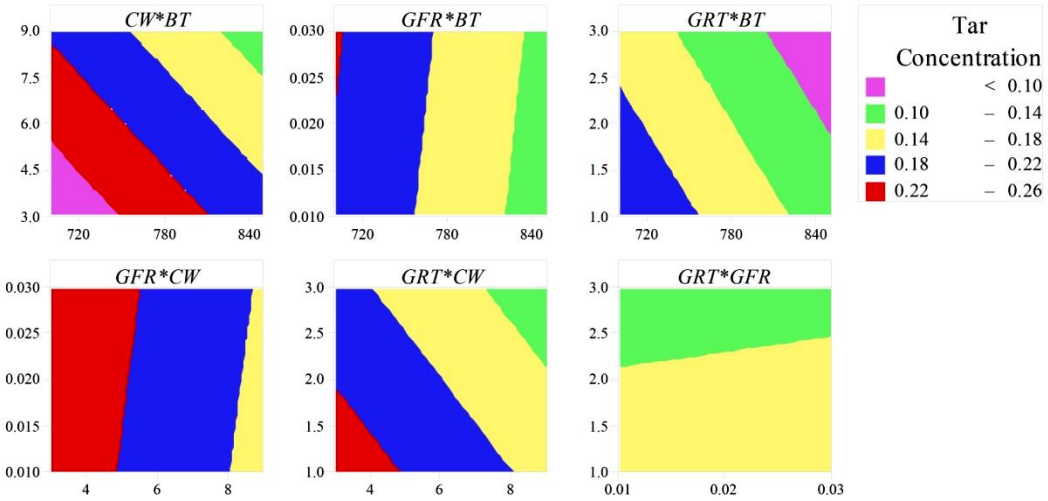

Figure 5Contour Plots of Tar Concentration

Table 4 Response for SN Ratio

Level	Catalyst	BT	CW	GFR	GRT
1	2.355	7.835	8.621	10.234	8.488
2	10.308	9.646	8.998	8.479	9.100
3	14.793	9.976	9.837	8.742	9.868
Delta	12.438	2.141	1.216	1.755	1.379
Rank	1	2	5	3	4

Table 5 Response for Mean

Level	Catalyst	BT	CW	GFR	GRT
1	0.7662	0.4748	0.4483	0.4066	0.4450
2	0.3116	0.4069	0.4410	0.4414	0.4230
3	0.1851	0.3812	0.3736	0.4149	0.3949
Delta	0.5811	0.0936	0.0748	0.0349	0.0501
Rank	1	2	3	5	4

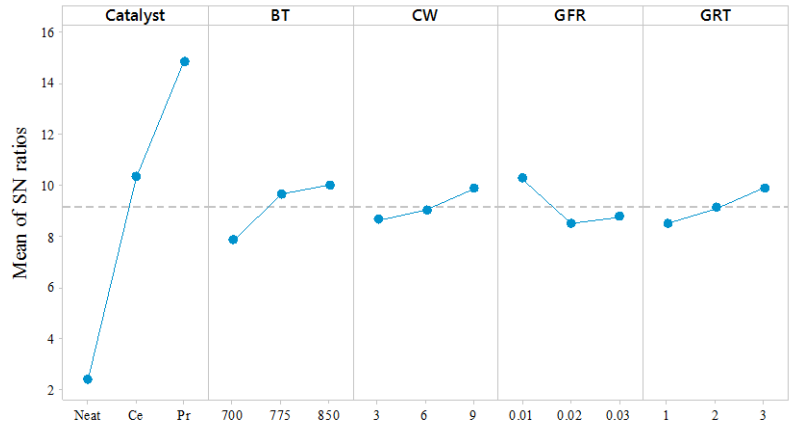

Figure 6 Main Effects Plot for SN ratios

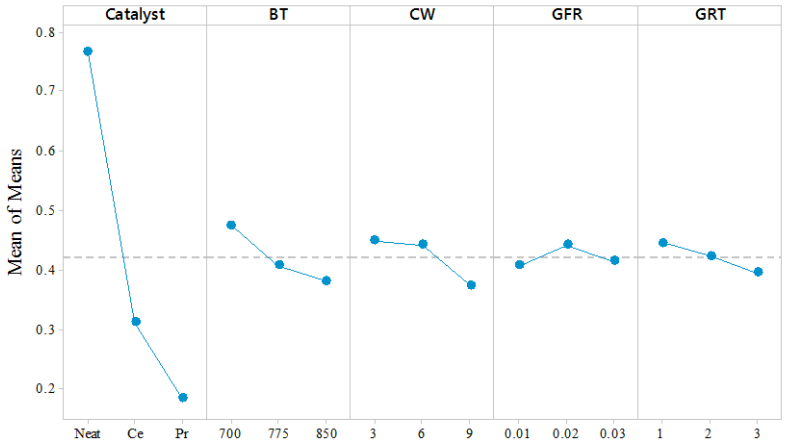

Figure 7 Main Effects Plot for Means

The optimal configuration of catalyst system for the reducing tar concentration is shown in Figure 8. The optimal configuration for tar reduction using Ni-Pr/$TiO_2$of 9g at 775°C of bed temperature with 0.01l/s gas feed rate and 2s of gas residence time. This configuration is experimented and the obtained optimal tar concentration is 0.14mg/Nm^3, which is deviated about 2.7 % from the computational result. Similarly, for other Ni/TiO_2 catalysts are tabulated in Table 6.

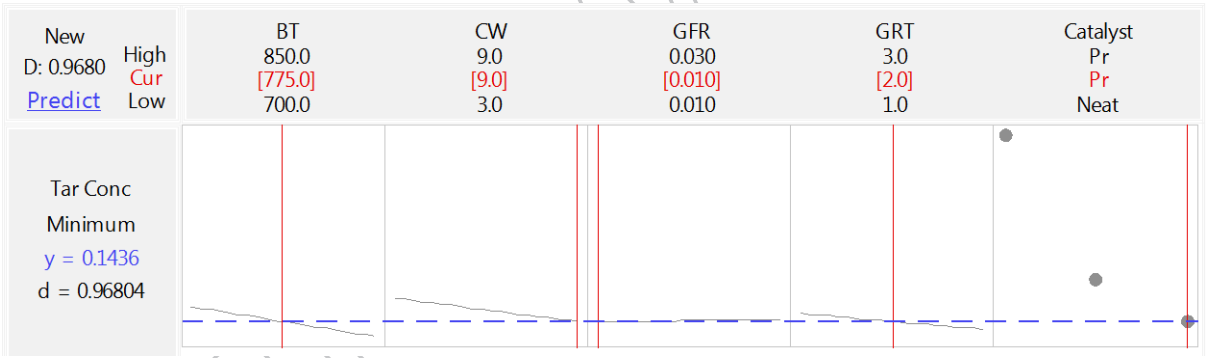

Figure 8 Optimal Configuration of Catalyst System

Table 6 Optimal Configuration for Catalyst

Catalyst	Tar Concentration (mg/Nm^3)	Tar Mitigation Efficiency (%)	Bed Temperature (°C)	Catalyst Amount (g)	Gas Feed Rate (l/s)	Gas Residence Time (s)
Ni/TiO_2	0.72	97.3	850	9	0.01	2
Ni-Ce/TiO_2	0.27	99	775	9	0.01	2
Ni-Pr/TiO_2	0.148	99.45	775	9	0.01	2

3.3. Emission and Performance of Producer Gas

The gas composition and the calorific value of the producer gas produced under thermal method (baseline) and optimal configuration are compared in Figure 9 and 10 respectively. The producer gas obtained using Ni-Pr/TiO_2catalyst has the calorific value of 5.44 MJ/m^3, which is high thanthe calorific value of producer gas produced from other catalyst.

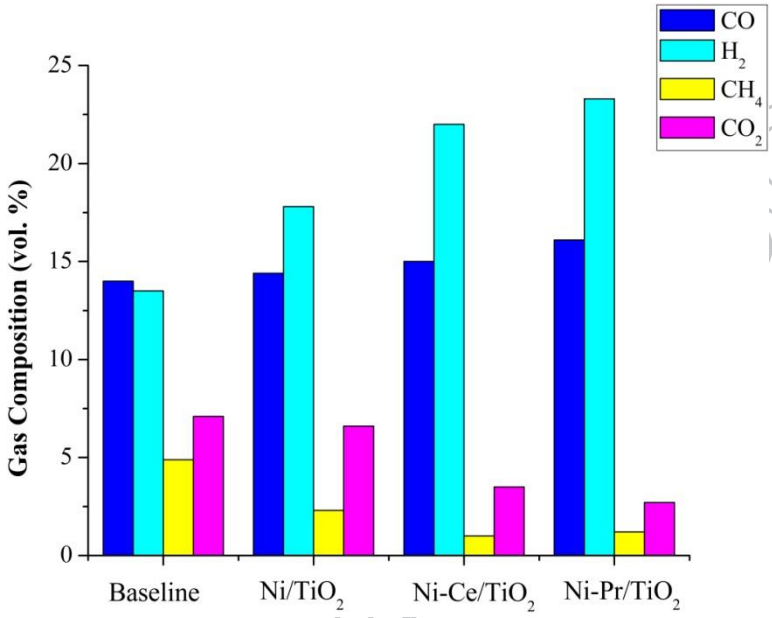

Figure 9 Gas Compositions of Producer Gases

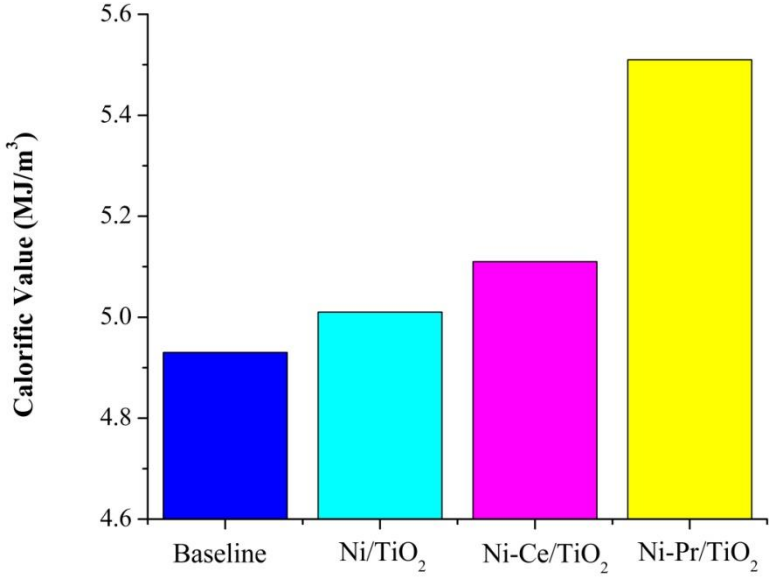

Figure 10 Calorific Values of Producer Gases

4. Conclusion

The new Ni/TiO_2, $Ni-Ce/TiO_2$, and $Ni-Pr/TiO_2$ catalysts are synthesized using CTAB process and its spherical support particle size is determined in range of less than 25nm in diausing SEM. The surface area and pore volume of $Ni-Pr/TiO_2$nano catalyst is found to be $110m^2/g$ and $0.197400cm^3/g$ respectively. Hence, the tar cracking ability of$Ni-Pr/TiO_2$nano catalyst would be higher than NiO/TiO_2nano catalyst. Since, the surface area of $Ni-Pr/TiO_2$nano catalyst is 26.3% higher than Ni/TiO_2nano catalyst.The baseline study for catalyst tar cracking is done by thermal method and the insignificant factors are fixed. Based on this analysis, the levels of significant influencing factors, design table and the optimization methodology are constructed.The regression models of tar concentration are derived with adequacy of 98.4%, and from the ANOVA, the most predominant factorsi.e. catalyst and bed temperature are inferred. The optimal configuration for tar reduction using $Ni-Pr/TiO_2$ of 9g in 775°C of bed temperature with 0.01l/s gas feed rate and 2s of gas residence timeis identified by Taguchi methodology. This configuration is experimented and the obtained optimal tar concentration is 0.14 mg/Nm^3, which is deviated about 2.7 % from the computational result. The producer gas obtained using $Ni-Pr/TiO_2$ for the optimal configuration is better in emission and calorific values also compared others. This work can be further extended with MetaporousMobile Crystalline (MCM) catalysts may also be endeavored for catalytic tar cracking.

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