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Parallel Reaction Kinetic Modelling Of Biogas To Biomethanol With Zno/Sio$_2$ Nanoparticles

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Abstract:- Kinetic model proposed in this study is a single reaction kinetic model of parallel biogas into biomethanol (CO_2 + H_2) and CO_2 gas (CO + O_2) in the gas phase. The phenomenon of this reaction is a new breakthrough in the field of renewable energy and very exciting to be modeled kinetic. These parallel reactions biomethanol and biogas into CO_2 gas in this study utilizes content of methane (CH_4) which is converted to CO_2 by nanoprticles biomethanol and ZnO impregnated silica. Nanoparticles ZnO made with Zn (SO_4) 7H_2O and NaOH with co precipitation method with stirring pressure of 1.2 bar and an isothermal drying temperature 180^{o}C. Reaction biogas into biomethanol and CO_2 takes place at a temperature of 350^{o}C and a pressure of 1.2 bar produce biogas conversion into biomethanol 70% and 25% CO_2 gas. at a temperature of 350^{o}C and volumetric rate of 5 ml per minute. Effect of water formed in this reaction will inhibit the formation of biomethanol and hydrocarbons.

Keywords:- Biogas, biomethanol, nanoparticles, kinetic, parallel

I. INTRODUCTION

Currently, the production of methanol from methane can be done through the process of decomposition, partial oxidation and steam reforming. Methanol to gasoline (MTG) with the catalyst ZSM 5 has been investigated very effective in converting methanol into gasoline. Merging two single reaction is a new process innovations used to develop a model reaction biogas into biogasoline through pyrolysis in a fixed bed reactor, the main advantage of this reaction process model does not require oxygen compounds, minimizing unwanted product and get the process closer to the scale technical industrial applications and allow the process to work with reactants in the gas to form a more homogeneous.

In line with the above reasons need to develop and transform the role of catalyst into nanokatalis in order to improve product quality gasoline. The development of synthetic material that has a structure of nanometer dimension has attracted the world of science and stimulate researchers to explore its application in the industrial, medical, pharmaceutical, food, consumer goods. Nanoparticles are catalysts that have a size of 1-100 nm. The reduced dimensions of the catalyst into nano size would increase the surface area of the catalyst and consequently increase the specific activity of the catalyst in the reaction. Society tends to desire to obtain goods at the lowest possible price with satisfactory quality. This is possible if the product is used in the manufacturing process at a minimum cost. Methane (biogas) in addition to the abundant amount, also can be used as an alternative energy source instead of fuel oil, because it has a lot of similarities between fuel oil with gas. So its use does not require a drastic change of the infrastructure which is generally used in fossil fuel use. The process of converting methane into liquid hydrocarbons can be done in three ways, namely steam reforming, autothermal reforming and partial oxidation. Methane has a low potential and do not last long so that when the conversion is done by means of steam methane reforming or autothermal reforming, commercially would be less profitable because it requires expensive investment. Whereas when converting methane partial oxidation method has a low selectivity, because there is competition between the reaction of oxygen with methane gas into methanol and the methanol newly formed gas into CO2 and H2O. So as to obtain high selectivity, oxygen is added to be controlled by staying. Control of oxygen can be performed by using oxygen ion conductive membrane.

This membrane and beyond merely supplying sufficient oxygen to convert methane to methanol by using oxygen ions in the crystal lattice of the membrane material. Thus, the use of oxygen for the conversion of methanol to be minimized so that the chances of further oxidation reaction of methanol with oxygen more be lower. Previous research on the sole reaction of methane into biomethanol has been found that the metal oxide catalysts capable of converting 92% achievement of this conversion is able to eliminate the inert ingredient in the product. These series of reactions in this study using biogas utilizing the content of methane (CH4), which is converted into the metal oxide catalysts biomethanol then simultaneously biomethanol converted into biogasoline with synthetic zeolite catalyst ZSM 5. Using serial reaction method can drive the reaction intermediate reaction route as be biogasoline, while the others can be minimized so that it takes a technique that is capable of directing the reaction series and is able to give the concept of merging a single reaction. **(Chumaidi 2008)** found that the metal oxide catalysts can reduce the occurrence of side reactions during the formation of methane that can biomethanol controlling permanently speed. In a single reaction, the formation of biomethanol able to produce high conversion besides that do not seem significant physical changes to the metal oxide catalysts. In subsequent studies **(Chumaidi 2009)** has obtained the data that affect the reaction process biomethanol change into biogasoline with modified natural zeolite catalyst consists of three parameters: the amount of catalyst, reaction temperature and volumetric velocity, of the three most influential parameter is the volumetric rate. **Tang (2012)** has made several attempts to transform methane into methanol. believe this work is interesting for the energy efficiency of natural gas utilization. 'Low cost methanol feedstock is very flexible which can be used for the production of gasoline and diesel or used directly as fuel. In addition, the advantages of this technology is that it can be used as an alternative method for handling remote gas or associated gas in small quantities. The proposed technology could stop the burning of natural gas that can not be processed further and reduce emissions from oil production. Although it has long been used as fuel in motor vehicles, but wider use of methanol is hampered by the high cost of producing it from methane - despite relatively low methane from natural gas is widely available and as a waste product from garbage dumps and livestock region. Differences methanol with methane is very small, ie, there is an excess of oxygen atoms in methanol, but to get this oxygen atoms without producing carbon dioxide is quite difficult and usually requires high temperatures and pressures.

Hibino, (2008) have discovered a new method for converting methane to methanol, which can operate at moderate temperatures (80C) and air pressure. They use a new material, tin phosphate doped with small amounts of indium, as the conductor material (electrolyte) in a fuel cell hydrogen / air. In addition to have succeeded in making a fuel cell that can transform methane (the main component of natural gas) into methanol (fuel useful) at moderate temperatures. **Buren (2009)** explained that biogas can be made from materials include human and animal waste, agricultural waste, municipal waste, industrial waste agricultural and other materials that contain organic ingredients. Biogas is a mixture of methane, carbon dioxide, hydrogen gas slightly, hydrogen sulfide and or nitrogen.

Price and Paul (2011) describes the methane gas or biogas CH4 contained in the magnitude of 60 to 70%, while the rest is in the form of CO_2, H_2S, nitrogen and hydrogen gas. Biogas has flammable properties with blue flame color, non-toxic and has a caloric value of 2.24 x 104 J / m^3. Methane gas is a gas that is the most dominant component in biogas has properties odorless, colorless and tasteless, the presence of other gases causes a stink. 0.554 methane gas density, low solubility in water, at a temperature of 20^oC and a pressure of 1 atm only 3 parts methane dissolved in 100 parts of water. Methane gas including stable gas. Methane gas energy value is high enough so that it can be used for various purposes, such as lighting, drying, cooking and other purposes. Complete combustion of methane will produce large amounts of heat. Complete combustion of 1 cubic meter (0.716 kg) of methane gas can liberate 8562 to 9500 kcal of heat and raise the temperature to 1400oC.

Planck and Schuth (2009), in explaining his biogas as an unused resource previously and have developed metal oxide catalysts convert biogas (methane) into biomethanol in a simple and efficient process by using the concept of solid catalyst activity and selectivity have high, and also have a high degree of stability to a series of processes to recycle. This study provides hope for the possibility of the implementation level of commercial production. The catalyst is a refinement of the catalyst platinum complex compounds, which previously. Subsequently has been successfully synthesize methyl sulfate, methane oxidation at temperatures of 200^oC, in a solution of concentrated H2SO4. Methyl sulfate can then be converted into methanol, with a recovery rate (yield) and selectivity high. Fischer Tropsch synthesis producing kinetic models with irrational order.

Dube and Carlson (2011) conversion liquid methane to methanol using a new approach uses a laser beam to reduce methane reaction barriers to creating long-chain molecules or fuels this process works without the need for heat or catalytic. This results in absolute methanol without water content.

Kovac and Davorain (2010) to convert biogas into methanol with simulation and modeling using the model equations NPL (non-linear programming) with a pressurized heating system with the parameters temperature, and pressure produces optimum conditions 840^oC temperature and pressure of 8 bar. Conversion of

99.7% achieved by the enthalpy 208.65 kJ / kg. **Sohraby and Fattahi (2009)** describes a model of oxidation of methane to methanol in a fixed bed reactor apply V2O5 / SiO2 as a catalyst to produce a model of CDF conversion with 84.4% 91.9% selectivity. In the simulation of mathematical models CDF direct conversion of methane to methanol is influenced by pressure and temperature. Conversion is very sensitive to changes in pressure thus methanol selectivity decreases with decreasing pressure, reduced pressure resulted in gas akspansi.

Husni and Husin (2007) describes the partial oxidation of methane to methanol and formaldehyde using a copper molybdenum oxide catalyst supported silica (CuMoO3 / SiO2) produces methane conversion when using a catalyst with a W / F of 5.82 grcat hours / mol, the ratio of Mo: Cu 1, 5: 1 and a reaction temperature of 450oC was 31.33%. Methane conversion increased to 33.08% in the W / F 9.7 grcat hours / mol, will further increase to 35.95% on the W / F 14.55 grcat hours / mol. Increased conversion also occurs in the catalyst ratio Mo: Cu of 2.5: 1 and the reaction temperature 450^{o}C. At the price of W / F of 5.82 grcat hours / mol methane conversion obtained was 31.59%. Conversion of methane in the W / F 9.7 grcat hours / mol increased to 33.74% and the subsequent conversion of methane in the W / F 14.55 grcat hours / mol increased to 34.20%. From these results it can be concluded that increasing the price of W / F will produce a greater reaction conversion. The increase is due to the conversion of the reaction the greater the W / F, the amount of catalyst used is also growing. Thus it would be perfect reactants adsorbed on the catalyst surface, causing the collision orientation reactant molecules can be more precise. This phenomenon is also in accordance with those reported by Piccoli (1992) that the increase in W / F will lead to increased conversion reaction. increase in residence time and reaction temperature causes a decrease in the selectivity of methanol. Methanol selectivity is lower at the time of stay and high temperatures. Methanol selectivity of the catalyst with the ratio of Mo: Cu 1.5: 1 and the reaction temperature 450oC at W / F of 5.82 grcat hours / mol is 9.53% and the W / F 9.7 grcat hours / mol decreased to 8, 67%, and the W / F 14.55 g h / mol lower selectivity becomes 8.53%.

Zhang (2008) describes the direct oxidation of methane to methanol with solid catalyst is a phenomenon of the most interesting and challenging to develop general high selectivity of methanol yielding large, these results can be obtained when using the medium as a replacement O2 NO2 catalyst for this type of peroxide has more reactive and more effective in the formation of methanol.

Kralj and Davorin (2010) describes the natural gas can be replaced with non-food sources of renewable energy Natural gas can be replaced by biogas to synthesis gas production of methanol. Biogas to methanol reaction kinetics were modeled with nonlinear programming (NLP) with the simulator Aspen. Parameters that influence the formation of NLP model is the flow rate, temperature and pressure. The model equations obtained from this study convergent and non-convergent models which includes the correlation of concentration, pressure and temperature. This model is likely to boost conversions by modifying the more varied the parameters - parameters that influence.

II. EXPERIMENT SECTION

2.1 Catalysts

Material ZnSO4.7H2O mixing as much as 397.67 grams and 3 mmol ascorbic acid with 197.6 grams of NaOH. The mixture is then in an autoclave at a temperature of 190^{o}C for 10 hours with agitation. After going through the autoclave process the mixture is then cooled to room temperature of about 25^{o}C naturally. White precipitate formed was filtered and then washed with distilled water. After the white precipitate is dried at a temperature of 80 ° C

2.2. Equipment and Reaction Conditions

The reaction equipment used is provided with an plug flow reactor, which has been conditioned for operating up to 10 bar and 350 °C. The reactor is a vertical hollow cylinder of 10 mm internal diameter and 64,4 mm length with a volume of 4 cm^3 for the catalyst. It is placed withina stainless steel chamber heated by an electric resistance, andthe inside is clad with a 9 mm thick thermal insulator. Theequipment is provided with a temperature controller that actuates on the catalyst bed and on the wall of thereactor. The molar flowrate of the reactants (CH_4, CH_3OH, H_2, CO, and CO_2) is controlled by Brooks 5850 mass flow meters. The reaction mixture is passed through several purifiers to eliminate possible traces of oxygen, water, and iron carbonyl that may deactivatethe catalyst.In order to carry out kinetic modelling, runs have been carried out under the following reaction conditions: 250-350 °C; 10-20 bar; space time, 1.6-57.0 (g of catalyst) h (mol H_2)$^{-1}$; time on stream, 6 h, which is sufficiently short for deactivation to be insignificant; (H_2 + CO), (CH_4 + CH_3OH) and (H_2 + CO_2) feeds, both with a 4:1 molar ratio between the components biogas to oxygen. These ranges havebeen established subsequent to a detailed study of the effect of operating conditions.

2.3 Product Analysis

The reaction equipment is connected on-line to aMicro-GCVarianCP-4900 gas chromatograph provided with a thermal conductivity detector (TCD) and a Porapak Q (PPQ, 10 m) column, where the lighter reaction products (H_2, CO, CO_2, methane, methanol, and water) are separated. Both detector and column are placed in the oven and operate within the 0-150°C range. The identification of the reaction products has involved gas chromatography / mass spectrometry (Hewlett-Packard 5890 II/ MS-engine) and the injection of pure standards. These techniques have allowed most of the components of the gaseous product stream to be identified.

III. RESULT

The results quantified are the yields of methanol, Methane, and hydrocarbons. The yields of methanol (*Y*MeOH) and CH4 (*Y*CH_4) have been calculated as the percentage of carbon atoms that convert to methanol or CH_4 from CO or CO_2 in the feed. Reaction engineering models of biogas into bioethanol is an alternative design that can be applied in parallel reaction kinetic models. Reactions to convert methane into methanol, CO_2 and water. Levels of methane, methanol, CO_2 and water are calculated as mole percentage

Kinetic scheme

$$\begin{tikzcd}
 & CH_3OH \\
2\,CH_4 + 2{,}5\,O_2 \arrow[ur, "k_1"] \arrow[dr, "k_2"'] & & (1) \\
 & CO_2 + 2\,H_2O
\end{tikzcd}$$

Reaction mechanism :

Oxidation reaction

$$CO_2 + \tfrac{1}{2} O_2 \xrightarrow{k_2} CO_2 \qquad (2)$$

$$H_2 + \tfrac{1}{2} O_2 \xrightarrow{k_2} H^2O \qquad (3)$$

Hydrogenation reaction

$$CO + 2\,H_2 \rightarrow CH_3OH \qquad (4)$$

$$4\,CH_4 + 5O_2 \longrightarrow 2CH_3OH + 2CO_2 + 4H_2O$$

(5)

Speed changes for the third component

$$-r_{CH_4} = k_1\,C_{CH_4} + k_2\,C_{CH_4} = (k_1 + k_2)\,C_{CH_4} \qquad (6)$$

$$r_{CH_4} = k_1\,C_{CH_4} \qquad (6)$$

$$Y_{MeOH} = \frac{C_{MeOH}}{(C_{Meth} + C_{CO2} + C_{H2} + C_{MeOH})}100 \qquad (8)$$

$$Y_{CO2} = \frac{C_{CO2}}{(C_{Meth} + C_{CO2} + C_{H2} + C_{MeOH})}100 \qquad (9)$$

$$Y_{H2} = \frac{C_{H2}}{(C_{Meth} + C_{CO2} + C_{H2} + C_{MeOH})}100 \qquad (10)$$

$$Y_{Meth} = \frac{C_{Meth}}{(C_{Meth} + C_{CO2} + C_{H2} + C_{MeOH})}100 \qquad (11)$$

$$-\ln\left(\frac{C_{CH_2}}{C_{CH_{4(o)}}}\right) = (k_1 + k_2)C_{CH_4} \qquad (12)$$

$$\frac{C_R - C_{Bo}}{C_S - C_{S_o}} = \frac{k_1}{k_2} \qquad (13)$$

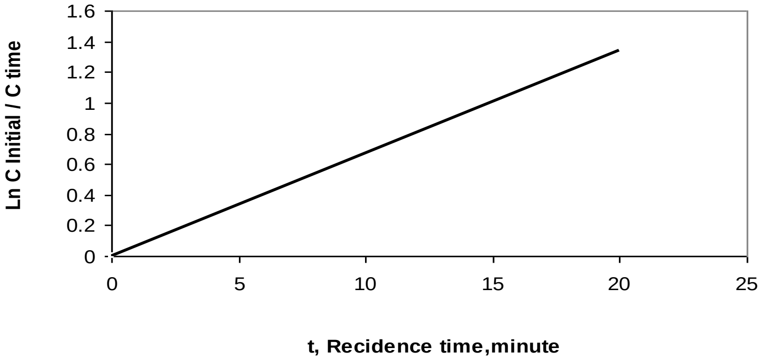

Figure 1 Model of the biogas order reaction becomes biomethanol (Eq - 1)

In Figure 1 Determination of reaction order nanoparticles ZnO-based biogas to be biomethanol reaction followed first order reaction with a reaction rate constant (k1) 0.1935 per minute, whereas the reaction order rreaksi for biogas into CO2 and H2O biometanol become impregnated ZnO based in SiO2 follow first order reaction the price of the reaction rate constants (k2) 0.1403 per min Second-order reaction indicates the direction of the reaction goes in one direction because the price is greater than the k1 k2 means the mechanism of this reaction to the relatively fast intermidiate compound is not dependent concentration residence time of the reactants in reactor, as a result of the mechanism linearly biogasoline products tend to follow the compound intermidiatenya According to **Price and Paul (2011)** content of methane in the biogas compounds with a range of 50-54% is quite stable if for reaki isothermal reaction and no impact on unwanted pollutants

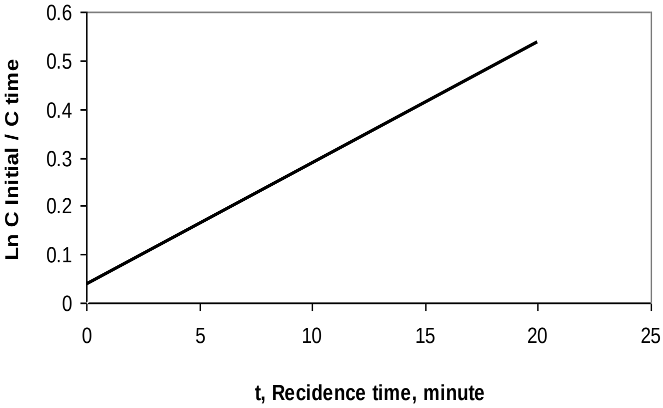

Figure 2 first order reaction model biogas into CO2 (Eq - 1)

In figure 2 Modelling reaction-based series of biogas into biogasoline CuO and CuO / SiO2 explain experimental model comparison experimentally in the laboratory in the simulation. This series of reactions take place in one direction of initiation (biogas) then intermidiate (biomethanol) until termination (biogasoline) in modelling the price difference will affect the concentration profile of all these compounds. For this model the price k1> k2 means the price of biogas into biomethanol reaction speed greater than the speed of reaction biomethanol into biogasoline, this case indicates that at t = 0 concentration of biogas would tend to diminish the extreme because these compounds will be controlled by the activity of the catalyst permanently the activation energy is very large so that the pattern of this reaction is not through a process of absorption but cracking at high temperatures. While the reaction biomethanol be biogasoline reaction patterns through absorption followed by cracking at high temperatures. At t> 0 trended uniform pattern of reaction reaction because intermidiate role has been controlled by the catalyst CuO / SiO 2 catalyst which acts as a steering termination. For comparison shown form a series reaction modelling biogas into biogasoline, modeling data on a more regular and follow the rules of the reaction kinetics series because there are some who assumed including inert product dispensed, the results of the analysis is complete, the temperature and pressure are relatively unchanged, there is no loss of compound

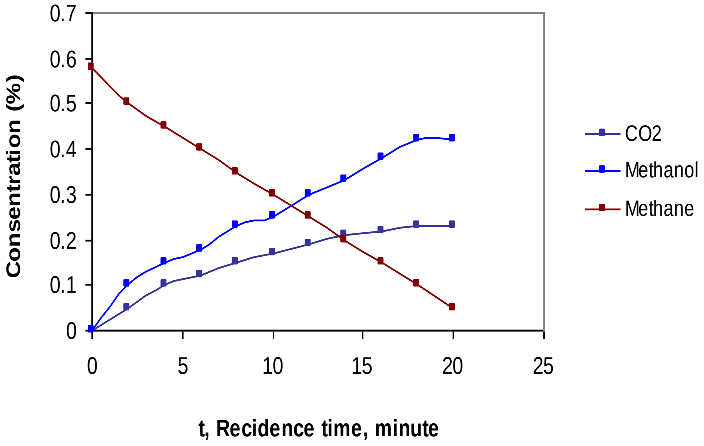

Figure 3 the conversion of methane into CO2 and H2O

In figure 3 the determination of the conversion of biogas into biogasoline follow the pattern of first order reaction, the same as the ZnO-based catalysts 0.1795 per minute price k1 and k2 = 0.1247 per minute in this reaction shows k1> k2 means the role of the reaction rate is dominated by the biogas transformation to biomethanol. Nevertheless conversion initiation to intermidiate greater than intermidiate reaction to this termination is due at the time t = 1 minute biogas converted linearly larger and lasted until constant at t = 10 min. Conversion difference in these two stages due to methane in the biogas compounds more volatile than biometanol so cracking and reforming processes faster on oxide catalysts.

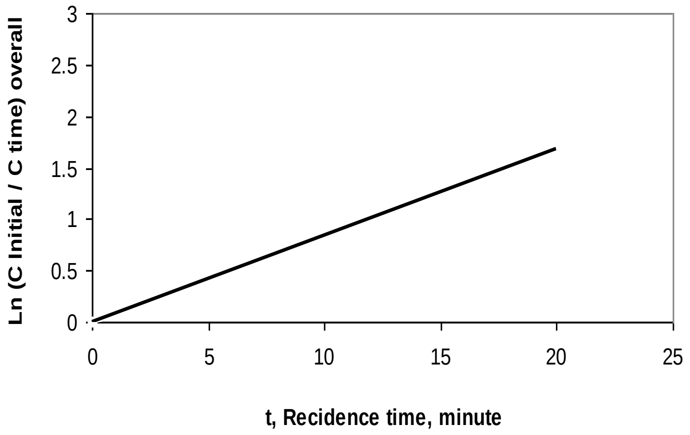

Figure 4 Modeling parallel reactions biogas into biogas overal

In Fig 4 the experimental biogas compounds in concentrations decreased linearly but are significantly different simulation at t = 2 min until at t = 6 min difference is due to the contact between the experimental compounds catalyzed methane turbulent flow occurs on the porosity catalyst that changes the concept of radical reactions often occur at the time during which the catalyst does not undergo deactivation. On the side that reached the maximum value intermidiate extreme enough for biogas concentrations approaching this encourages intermidiate compounds simultaneously turned into biogasoline. Condition is very interesting because of a decrease in the concentration of biogas and maximum price intermidiate occurred at almost the same time so that the reaction is relatively rapid absorption followed by reaction.

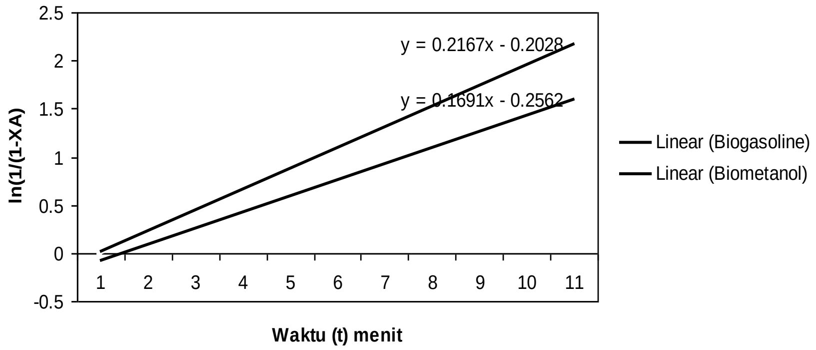

Figure 5 first-order reaction model based catalysts ZnO / SiO2

In figure 5 shows that quite contradictory with two types of catalysts previously, a significant difference occurs in the price k1 = 0.2167 per ment and k2 = 0.4691 per minute mean reaction speed will be controlled by the reaction intermidiate be biogasoline. These conditions led to the formation of intermidiate not quite as quantitative act as reactants for biogasoline. Other differences occurred in the pattern speed of the reactants, respectively, on the side of the biogas flow pattern tends laminar flow pattern on the side while intermediate turbulent.


Results

Z-Average (d.nm): 359.0
PdI: 0.264
Intercept: 0.934
Result quality Good

	Size (d.nm...	% Intensity:	St Dev (d.n...
Peak 1:	375.7	97.9	158.8
Peak 2:	5370	2.1	328.5
Peak 3:	0.000	0.0	0.000

Figure 6 Characteristics of ZnO nanoparticles

In figure 6 nanoparticles ZnO occurs at a wave length of 320 μm and does not occur amorphous particles.

IV. CONCLUSION

Model significant reaction biogas into biogasoline relevant series on ZnO catalyst / SiO2. value maximum intermidiate 53% on ZnO-based catalysts / SiO2. Reaction of biogas into biogasline series follows the pattern of first-order reaction value of the reaction rate constants for the two reactions are relatively similar

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