Use Of Natural Gas Become a Petrochemical with Zeolite Catalyst

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Use Of Natural Gas Become a Petrochemical with Zeolite Catalyst

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Abstract. Methane is a hydrocarbon compound that can be used for various purposes. Currently, methane is widely used as fuel, and continues to be developed as a synthetic gas material and petrochemical products that have added value, one of which is formaldehyde. This study aimed to study the partial catalytic oxidation reaction of methane to formaldehyde using MoO₃/zeolite as a catalyst. Reactant gas stream consisting of pure O₂ and CH₄were contacted with a rate ratio of 1:20, put into Fixed Bed Reactor therein contained 30 grams of catalyst MoO₃/zeolite, and the pressure was maintained at 1 atm. The temperature and flow rate of the incoming reactant gas were used as variables. The main product produced was formaldehyde, besides that there were also CH₃OH, CO, CO₂, H₂O. The results were analyzed using Gas Chromatography, so it was known that the converted methane was 5.18% and the volume fraction of formaldehyde formed was 1.46%.

Keywords: Natural Gas, Zeolite Catalyst, Methane

1 Introduction

Zeolite is an acid catalyst that is interesting enough to be investigated and studied further for its performance in catalyzing hydrocarbon reactions. This is due to the nature of zeolite which is generally suitable as a catalyst or as a catalyst support. Therefore, zeolites can be used as filters, ion exchangers, absorbent materials and catalysts [1]. This suitability is due to the structure of the zeolite which generally provides effective site cavities for several chemical reactions, at high temperatures the catalyst activity is more effective, Hzeolite has good activity stability [2].

In Indonesia, the availability of zeolites is in large enough deposits. However, its benefits are still not so widespread. Methane is a hydrocarbon that can be utilized for various needs. The main use of methane at this time is as a fuel, recently it has been continuously developed as a synthetic gas material and also other petrochemical products. In connection with very excessive and cheapness of methane encourage people to convert it into other products that have added value. Which now has done are converting vapor methane into syngas, converting the syngas into methanol and then converting methanol into chemicals which are more useful for example, methanol can be oxidized to formaldehyde through commercial catalytic processes. Formaldehyde is known as a raw material for

The partial oxidation reaction of methane to formaldehyde was carried out by Spencer [4]. Through the experiments, Spencer reacted methane and oxygen gas with a molar ratio of 19: 1 to 6: 1 at a pressure of 1 atm. The gas was flowed through the MoO_3/SiO_2 catalyst at a temperature variable of $848\,^{\circ}$ K to $923\,^{\circ}$ K and a gas velocity (GHSV) of 2500-10000 per hour. Results of Spencer's experimet showed that methane was converted around 0-5%, while the selectivity of formaldehyde between 30-89%. As well as known that mechanism of oxidation of methane into formaldehyde of the order of 1 to methane, the reaction equation as follows:

CH₄ + O₂
$$\xrightarrow{k_1}$$
 HCHO + H2O (1)
HCHO + 1/2 O₂ $\xrightarrow{k_2}$ CO + H2O (2)
CH₄ + 2O₂ $\xrightarrow{k_3}$ CO₂ + H2O (3)

manufacturers of insulation and adhesive materials. Formaldehyde industries generally use metal catalysts of Fe and Mo, as the metal catalyst is more tolerant of contaminants so that widely applied in the industries [3].

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2 Research Method

2.1 Research procedure

This research uses a flow reactor in the form of a pipe fixed filling. The reactor filled with MoO₃/zeolite catalyst grains as a catalyst (Fixed Bed Reactor). The flow rate of the reactants into the reactor is expressed with GHSV. The reactants consist of fuel gas as a source of CH₄ and pure O₂, with a rate ratio of 20: 1. Inlet gas velocity (GHSV) is set between 2500 to 10000 per hour and the reactor operates at atmospheric pressure. Overall research is done in several stages as follows: Preparation of catalyst/buffer, Impregnation of catalyst. catalyst, and continued Characterization of with catalytic oxidation.

2.2 Experimental Variables

- a. Operating temperature : 300 $^{\rm o}$ C ,400 $^{\rm o}$ C, 500 $^{\rm o}$ C, 600 $^{\rm o}$ C, 6 50 $^{\rm o}$ C
- Methane gas flow rate: 1 lt/mt; 1.5 lt/mt; 2 lt/mt; 2.5 lt/mt; 3 lt/mt

2.3 Drawing Tools

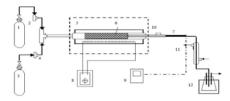


Fig. 1. Equipment Scheme.

Description of Figure 1:

- Methane gas cylinder
- 2. Flow meter of methane flow
- 3. O₂ gas
- 4. Flow meter of flow of O2 gas
- 5. Furnace reactor
- 6. Catalyst chamber
- 7. Thermocouple
- 8. Thermoset
- 9. Temperature display
- 10.Gas sampling
- 11.Condenser
- 12.Liquid sample container.

3 Results and Discussions

The results of these studies were analyzed using Gas Chromatography. The detector used was a TCD (Thermal Conductivity Detector), while the column type was of a packing column (filling column) PEG, in this column were silica coated with Poly etylen glycol as a liquid. Due to the limitation of existing tools, the products could be analyzed were only formaldehyde, methanol, H₂O and residual of methane gas. While CO

and CO_2 gasses that were supposed to come out were undetected in the chromatogram data. This was probably due to the low resolution of the column packing so that gas CO and CO_2 mixed together with methane gas. The separation resolution in the packing column was not as good as the capillary column.

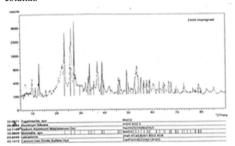


Fig. 2. Impregnation Zeolite

In Figure 2, the diffractogram pattern resulting from the impregnation above can be seen at an angle of 2 theta, namely 23. 34°; 25.70°; 25.88° and 27.34° These four angles were high peaks and indicate the presence of Mo metal. But the highest at an angle of 27.34°. After the AAS test, the Mo content was found to be 11.43%. According to Wu H.C [5], in the catalytic partial oxidation of methanol to formaldehyde using MoO3 and Fe2 (MoO4)3 catalysts, these two catalysts have high selectivity. According to Chun J.W [6] on the catalytic oxidation of methane, if the operating pressure is high, the reaction leads to methanol products, whereas if the operating pressure is low, the reaction leads to the formation of formaldehyde. Natural Zeolite from Lampung which has been in BET has surface area of 44.21 m²/g, while H-zeolite's surface area was 90. 17 m^2/g .

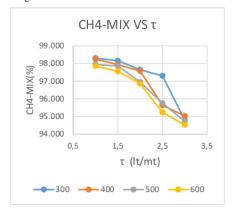


Fig. 3. Relationship between CH4 mixed and $\boldsymbol{\tau}$ at various temperature

Mixed methane (CH₄,CO₂CO₂). This Mixture is caused by the low resolution of the packing column. So CO and CO₂ Combine with methane gas. The decrease in the concentration of mixed methane is due to the

length of residence time in the reactor, so the catalytic oxidation rection will run well.

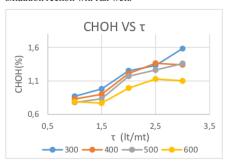


Fig.4. Relationship between CHOH and $\boldsymbol{\tau}$ at various temperature

As seen in the figure, there is a gradual increase in formaldehyde, then tends to stabilize. this is because the maximum volume of the reactor is no longer capable, so that formaldehyde becomes stable.

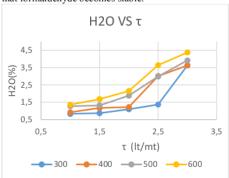


Fig.5. Relationship between H_2O and τ at various temperature

It can be seen in the figure that H_2O increased with increasing residence time. Due to the activity of catalyst and also by the formation of H_2O

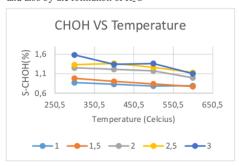


Fig.6. CHOH selectivity relationship with temperature at various τ

The decrease in selectivity of formaldehyde was due to the formtion of other products during the oxidation

reaction, namely CO, CO₂, H₂O and metanol. The highest selectivity of formaldehyde occurred at 300 °C about 97.8 %

4 Conclusion

- From the results of oxidation reaction methane to formaldehyde obtained the highest conversion CH₄ of 5.18% with a volume fraction of formaldehyde out of the reactor 1.46%
- The catalytic oxidation reaction studied is an order reaction 1 to methane.

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