CHAPTER IV

EXPERIMENTAL SYSTEMS

AND EXPERIMENTAL PROCEDURES

This chapter describes the experimental systems and the experimental procedures used in this works. A description of catalyst preparation method is presented in section 4.1-4.4

4.1 Preparation of catalysts

Materials

The chemicals used in this experimental are normally analytical grade. Alumina (Al_2O_3) support (type NKH-3) was obtained from Sumitomo Aluminium Smelting Co., Ltd. Japan.

Apparatus

1) Unit for grinding and screening support

This unit which consists of a mortar and sieves, is used for reducing the size of the catalyst support to 40/60 mesh.

2) Unit for impregnation

This unit which consists of pipettes, flasks, droppers and volumetric flasks is used for preparing aqueous solution and impregnating the solution onto the support.

3) Unit for calcination

This unit which consists of an electrical furnace, an automatic temperature controller, a variable voltage transformer, is used for calcining the impregnated catalysts at high temperature.

4.2 Preparation of support

Alumina support was grounded to the required mesh size of 40/60 followed by washing with distilled water 3-4 times to remove the very fine particles and other impurities, then dried at 110°C for overnight. Subsequently, the support was calcined in air at 300°C for 3 hours.

4.3 Preparation of stock solution

Copper stock solution

The copper solution was prepared by dissolving copper(II) nitrate in de-ionized water to total volume of 25 ml.

Other types of stock solution were prepared from following metal compounds:

Type of stock solution	**prepared by**
Cerium	Cerium (III) nitrate
Zinc	Zinc nitrate
Zirconium	Zirconium nitrate
Samarium	Samarium (III) nitrate
Neodymium	Neodymium nitrate
Praseodymium	Praseodymium (III) nitrate

4.4 Preparation of Copper Catalyst

1) The concentration of impregnating solution, for 3 grams of catalyst support, was prepared by calculating the amount of the stock solution to yield the required metal loading (Appendix A). De-ionized water was finally added until 3 ml. of the solution was obtained.

2) 3 grams of support was placed into a 100 ml. Erlenmeyer flask and the impregnating solution from 1) was slowly dropped to the support using a dropper. Continuously stirring of mixture in the flask while impregnating was

required in order to achieve the homogeneously distributed metal component on the support.

3) Leave the mixture in the flask for 6 hours to obtained good distribution of metal complex.

4) The impregnated support was dried at 110°C in air overnight.

5) The calcination step was carried out by placing the dried material obtained from 4) in a quartz tube, 10% hydrogen/nitrogen was firstly introduced into the tube at a flow rate of 100 ml./min. Then the tube was heated up at an increasing rate of 10°C/min. until the temperature reached 500°C. The material was held in this condition for 3 hours.

6) After calcination in 10% hydrogen/nitrogen gas for 3 hours, the tube was purged by nitrogen to cool down until the temperature decreased to room temperature.

4.5 Reaction of methanol synthesis

Materials

Gas mixture of 33% carbon monoxide in hydrogen supplied by Thai Industrial Gas Co.,Ltd. was used as feed stream for methanol synthesis in this study. The hydrogen was used for reducing the catalyst, while the nitrogen was used for purging the system.

Apparatus

Flow diagram of the methanol system is shown in figure 4.1. The system consists of a reactor, an automatic temperature controller, an electric furnace, and gas controlling system.

1) Reactor

The methanol synthesis reactor is made from 316 stainless steel tube, it can be operated from room temperature up to 600°C under atmospheric pressure. Sampling points are provided. Catalyst is placed between silicon carbide layer.

2) Automatic temperature controller

Automatic temperature controller consists of a magnetic switch, a variable voltage transformer, a temperature controller (PF-4, RKC), and a thermocouple. Temperature control set point is adjustable within the range between 0 to 800°C.

3) Electrical furnace

An electrical furnace supplied heat to the reactor for methanol synthesis. Therefore, the reactor can be operated from room temperature up to 800 °C at the maximum voltage of 220 Volts.

4) Gas controlling system

The system consists of ;

4.1) A cylinder of 33% carbon monoxide in hydrogen, equipped with a pressure regulator (0-400 Psig), a Whitey on-off valve, a Nupro fine-metering valve, and a Nupro check valve used for adjusting the flows rate of these gases.

4.2) The cylinders of hydrogen and nitrogen, each equipped with a pressure regulator (0-200 Psig), a Whitey on-off valve, a Nupro fine-metering valve, and a Nupro check valve used for adjusting the flows rate of these gases.

5) Gas chromatograph

A gas chromatograph equipped with a thermal conductivity detector (TCD) was used to analyze the composition of hydrocarbon in the feed and product streams. The operating conditions are illustrated in table 4.1

Table 4.1 Operating conditions of gas chromatograph (GC 8 ATP)

Model	GC 8 ATP
Detector	TCD
Packed column	MS-5A/Porapak Q
Nitrogen flow rate	30 ml./min.
Column temperature	100 °C
Injector temperature	110 °C
Detector temperature	110 °C

4.6 Experimental System

The reaction system consists of a reactor installed in a tube furnace. The diagram of the system is exhibited schematically in figure 4.1. The furnace temperature is controlled by a temperature controller. The reactor is constructed from a 316 stainless steel tube. A syngas (33% CO/H_2) is used as reactant gas. During the experiment, the reaction temperature is monitored using a thermocouple and a digital temperature indicator. The effluent gas is analyzed by a gas chromatograph equipped with a thermal conductivity detector. The operating conditions of the GC are shown in table 4.1

Experimentation

The experimental procedures are described in detail as follows :

1) 1 gram of catalyst was packed in the middle of the stainless steel tube reactor, which placed in the furnace. The nitrogen gas was introduced into the reactor at a flow rate of 150 ml/min.

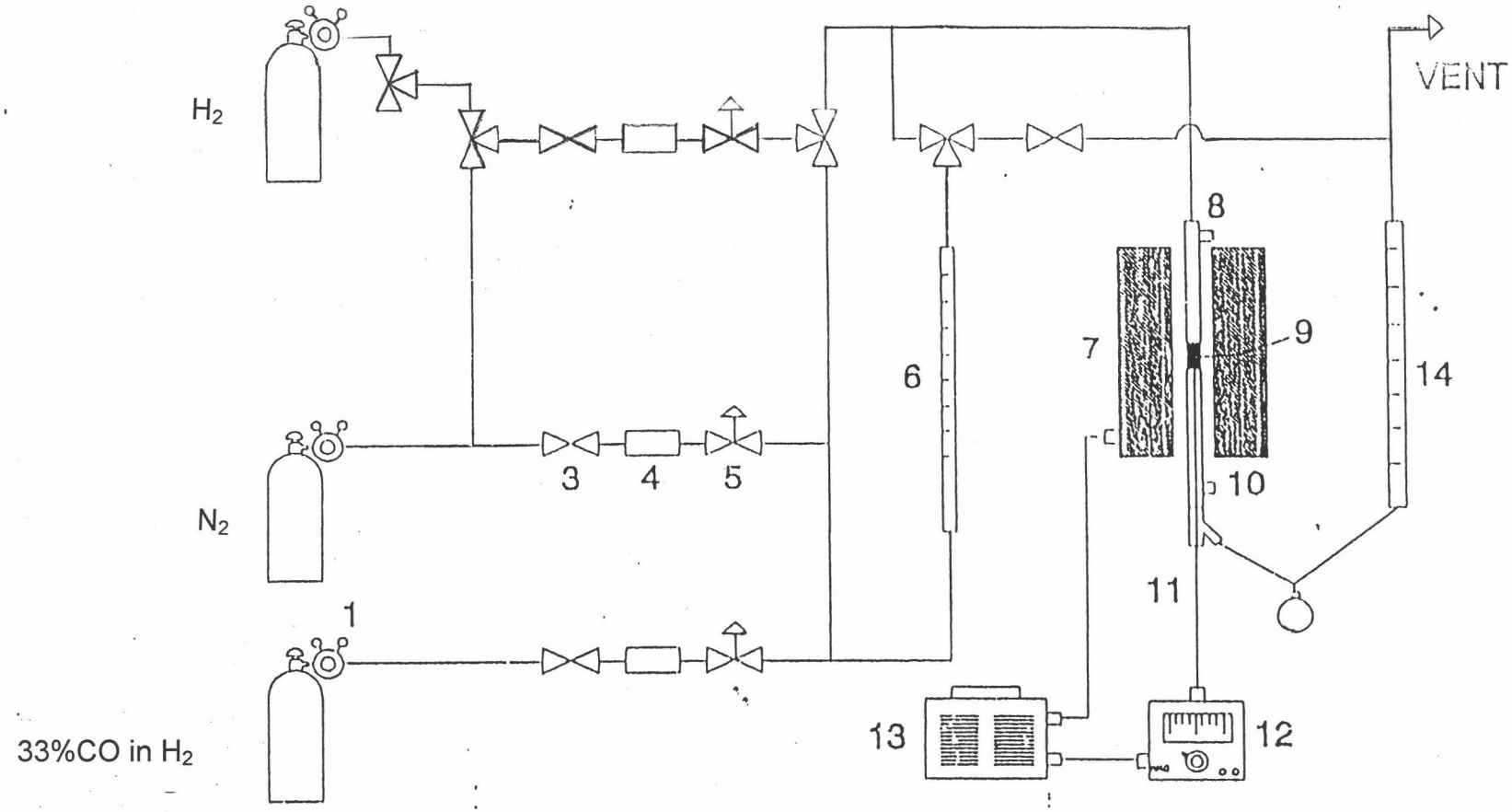
2) The reactor was heated up at a heating rate of 10°C/min.until the catalyst temperature reached 100°C. Then, the nitrogen gas was replaced by 10% hydrogen/nitrogen gas at allow rate of 170 ml/min. The catalyst was reached at this temperature for 1 hour.

3) When the reduction process was completed, the hydrogen/ nitrogen gas was replaced by the syngas and held up at this temperature for 10 min.

4) The gas sampling was taken at 10 min.

5) When, the reactor temperature was heated up until the temperature reached 250°C then, repeat step 3),4) and heated up to 300, 350 until 400°C

6) The reactor was cooled down by switching from the syngas to nitrogen gas.



1. Pressure Regulator
2. Three-way Valve
3. On-off Valve
4. Gas Filter
5. Needle Valve
6. Flow Meter
7. Furnace
8. Reactor
9. Catalyst Bed
10. Sampling Port
11. Thermocouple
12. Temperature Controller
13. Variable Voltage Transformer
14. Bubble Flow Meter

Figure 4.1 Flow diagram of the methanol synthesis system.