

CHAPTER III

EXPERIMENTAL

Catalyst Preparation and Characterization

1. Chemicals

Catalysts and other chemicals used in the experiments are listed with grade of purity and manufacturer as shown in Table 3.1.

All catalysts were dried in the oven at the temperature 120 °C for dehydrating and kept in the desiccator.

2. Catalyst Characterization

The catalysts were characterized by the following techniques:

1) Thermogravimetry (TG) and Differential Thermal Analysis (DTA)

TG/DTA (38) was used to determine the thermal stability of the catalysts. The experiments were carried out on a Dupont Thermoanalyzer under a flow of nitrogen in the temperature range of 40-900°C with a heating rate of 20 °C/min.

Table 3.1 Source of chemical

Materials	Grade of Purity	Manufacturer
Carbon monoxide (CO)	99.3%	TIG Co. Ltd., Samutprakarn, Thailand
Oxygen (O ₂)	99.8	TIG Co. Ltd., Samutprakarn, Thailand
Nitrous oxide (N ₂ O)	99.5	TIG Co. Ltd., Samutprakarn, Thailand
Nitrogen (N ₂)	99.5	TIG Co. Ltd., Samutprakarn, Thailand
Tricobalt tetraoxide (Co ₃ O ₄)	AR	BDH Chemical Ltd., Poole,England
Ferric oxide (Fe ₂ O ₃)	99.998	Aldrich Chemical Co. Inc., WI,U.S.A
Trimanganese tetraoxide (Mn ₃ O ₄)	97	Aldrich Chemical Co. Inc., WI,U.S.A
Chromic oxide (Cr ₂ O ₃)	AR	BDH Chemical Ltd., Poole,England

2) Brunauer - Emmett - Teller (BET) Method

The surface area of all catalysts were measured by BET method (11) using BET model ASAP 2000.

3) X-ray Diffraction (XRD)

XRD was used to identify catalysts during before and after the catalytic reaction. The XRD measurements (38) (scanned in the range $5^\circ < 2\theta < 90^\circ$) were performed on a Jeol type JDX 8030 X - Ray diffractometer with a Cu X - ray tube ($\lambda = 0.15 \text{ nm}$).

Experiments in Flow Apparatus

The catalytic activity and selectivity of the catalysts have been determined in an open continuous flow apparatus equipped with a tubular fixed bed reactor (Figure 3.1) (39).

The gas flows of the gases e.g. N_2 , O_2 , CO , and N_2O were purified by passing over a column containing a reduced copper deoxo (BTS) catalyst and a column containing a 0.5 nm molecular sieve. The gases were regulated by conventional flow controllers. The gas mixture (gas reactants and nitrogen) was led through a tubular fixed bed reactor loaded with the catalyst. In use, the reaction reactor was heated from 150 to 400 °C. Gas chromatography (GC) was used to analyze composition of the product.

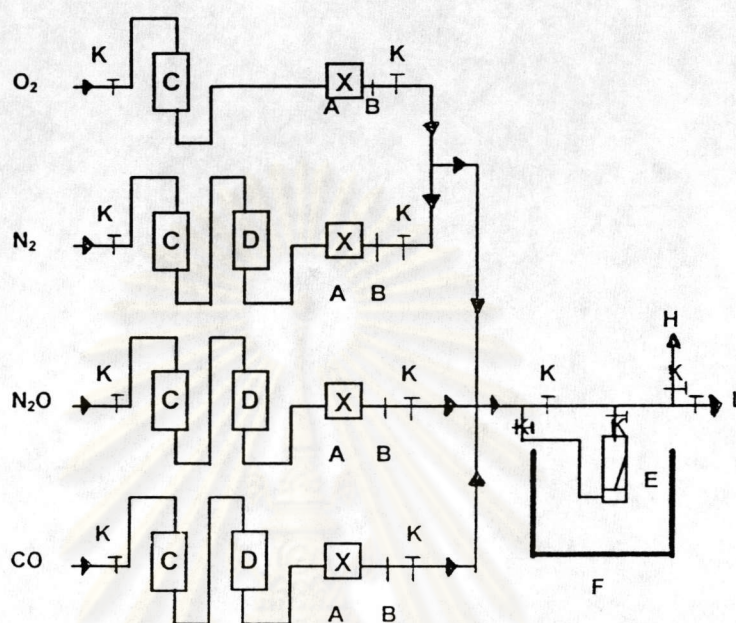


Figure 3.1 Schematic drawing of the flow apparatus

A = flow regulator

B = flow meter

C = purification columns

D = purification columns

E = reactor with catalyst

F = oven

H = gas chromatograph for gas analysis

L = outlet

K = valves

1. Experimental Conditions

After loading 300 mg. of catalyst in the reactor, oxygen was passed over the catalyst. The oven was heated to 400 °C and cooled down to 150 °C. Then, nitrogen was passed in the reactor until no oxygen remained (checking by GC). The gas mixture, the flow rates of CO, O₂ or N₂O, and N₂ were 2.5, 3.0, and 2.5 ml/min, respectively, were passed in the reactor without going through the catalyst for estimating 100 percent moles of CO. After the catalytic reaction was started by passing the gaseous mixture through the catalyst, the temperature of oven was increased at the rate of 20 °C/10 minutes. The product mixture was analyzed by GC and the samples were taken every 10 minutes. The reaction was stopped when CO conversion was 100 percent or when temperature reached 400 °C.

2. Gas Chromatography (GC)

The product analyses were performed on a Shimadzu model 9A gas chromatograph equipped with Unibeads C 80/100 (4.0 m. long, 3 mm. diameter) column. Thermal conductivity detector (TCD) (38) was used in the experiment. The GC experimental conditions were shown in Table 3.2.

Table 3.2 GC experimental conditions in the reaction of CO with O₂ and CO with N₂O

Conditions	CO + O ₂ Reaction	CO + N ₂ O Reaction
Sample size	1.0 ml	1.0 ml
He flow rate	60 ml/min	60 ml/min
Column temperature	110 °C (isothermal)	130 °C (isothermal)
Injection temperature	120 °C	140 °C
Detector temperature	130 °C	150 °C
Current	120 mA	120 mA

Data Evaluation

Percent conversion of CO according to the catalytic reaction has been calculated by following method:

Gas chromatogram of CO, the peak area of bypass CO (Experimental Conditions) is estimated as 100 percents moles of CO, thus the percent conversion of CO can be calculated by equation 3.1.

$$\% \text{ Conversion of CO} = 100 - [(A_{\text{cat.}}/A_o) * 100] \quad (3.1)$$

when, $A_{\text{cat.}}$ is the peak area of CO in the reaction.

A_o is the peak area of CO from by pass.