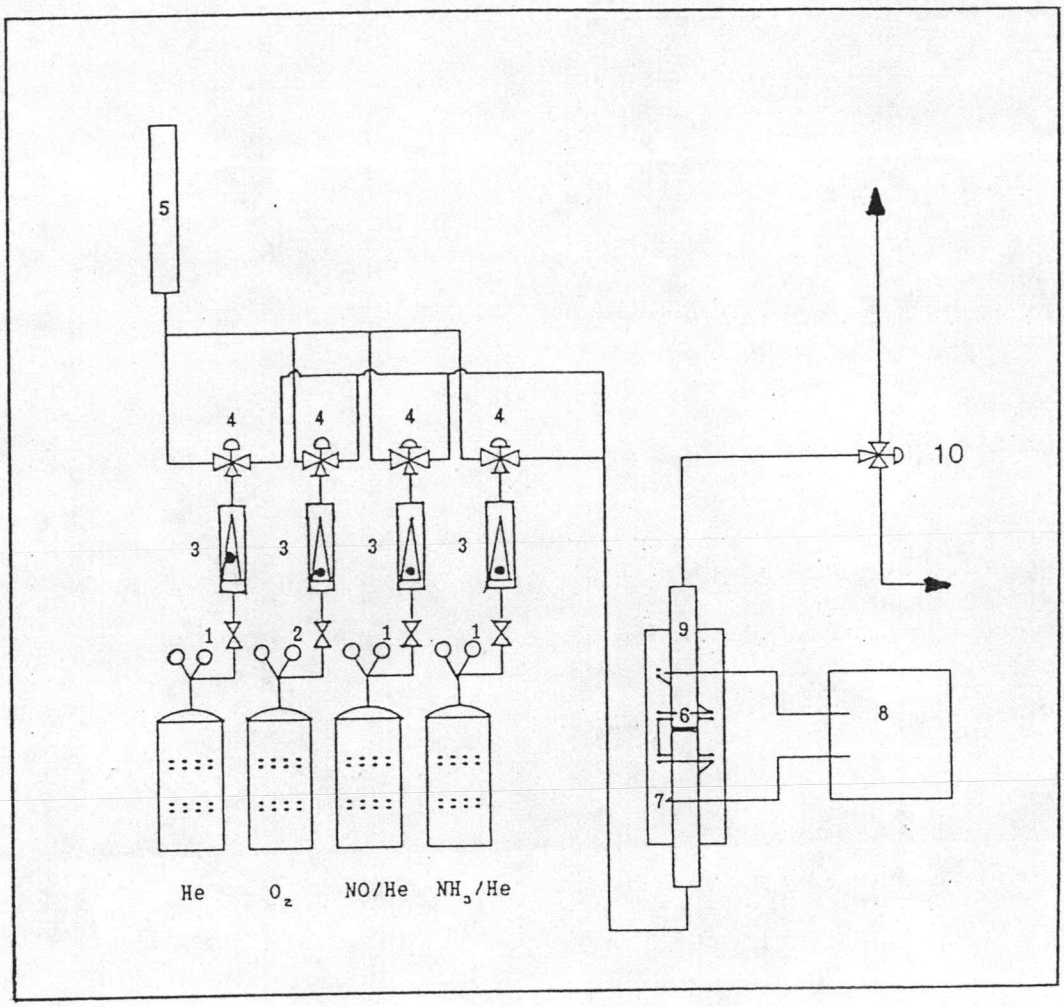
CHAPTER III

EXPERIMENTAL APPARATUS AND PROCEDURE

3.1 Construction of apparatus

The apparatus, used in this work, is designed to operate at atmospheric pressure, 100-400 ^{o}C in temperature ranges for reactor section. Swagelok valves and fittings are used to connect to the various size of stainless steel tubes. A complete flow diagram of the experimental apparatus is illustrated in Figure 3.1. It consists of three main sections. The first is a fed section which is four reactant gas cylinders containing He, O_2, 5000 ppm of NO balanced with He and 5000 ppm of NH_3 balanced with He respectively. The reactant gases are mixed before being transferred to the reactor by adjusting their outlet pressure with pressure control valves and volumetric flow rate is measured by a flow meter. The second is reactor section which consists of fixed-bed tube reactorand heating unit. The body of reactor is cylindrical stainless steel tube, 1.2 cm. inside diameter and 1.4 cm. outside diameter. Catalyst is packed in the middle of reactor tube. The heating unit consists of two parts, furnace and temperature control. The last one is sampling section, collecting gas products at the three ways sampling valve.



1. Metering Valve
2. Needle Valve
3. Flow Meter
4. Three Ways Valve
5. Bubble Flow Meter
6. Catalyst
7. Heater
8. Electrical Heating Temperature Control Unit
9. Reactor
10. Three ways sampling valve

Figure 3.1 A complete flow diagram of the experimental apparatus.

3.2 Catalyst preparation

There are three catalysts used in this work. The first is Pt-HY zeolite obtained form the laboratory of Prof. Hiroo Niiyama , Department of Chemical Engineering , Tokyo Institute of Technology, in Japan. The second is Pt-Cu-HY zeolite, prepared by loading copper to Pt-HY zeolite. The last one is V_2O_5, supported on TiO_2. Percentage of V_2O_5 is varied in series.

3.2.1 Copper loading method to Pt-HY Zeolite

To load Cu to Pt-HY Zeolites, Cu ions were exchanged from a solution of about 5 % of $Cu(NO_3)_2$ at room temperature for 24 hours. The copper solution was added dropwisely to the slurry containing Pt-HY zeolite in deionized water. Before being used in experimental reaction, the obtained catalyst zeolite is calcined at 300 ^oC in the presence of O_2 for 5 hours.

3.2.2 Impregnation of V_2O_5 to TiO_2

Five grams of the TiO_2, as the support, were previously washed with deionized water by soxhlet extraction and then dried overnight at 120 ^oC; this significantly decreased the concentration of the P and K. After drying, the desired amount of NH_4VO_3 was added to 20 ml. of 1M. oxalic acid, $H_2C_2O_4.2H_2O$, which upon heating yielded the deep blue $(NH_4)_2[VO(C_2O_4)_2]$ complex; for high V contents (more than 14 wt.% V_2O_5); stoichiometric amount of NH_4VO_3 and oxalic acid were dissolved

in 20 ml of water. The solution was added to the support, and the water was removed by evaporation with the continuous stirring. The resulting solid was dried overnight at 120 ^{o}C and a proportion was calcined in flowing air at 450 ^{o}C for 5 hours before used in the experimental reaction.

The composition of catalysts was analyzed by The Scientific and Technological Research Equipment Center Chulalongkorn University, (STREC). The ingredients of catalysts were shown in Table 3.1 and 3.2.

Table 3.1 Catalyst composition; Modified Zeolite

Weight (%)	Type of catalysts	
	Pt-Zeolites	Pt-Cu-Zeolites
SiO_2	71.580	68.565
Al_2O_3	23.392	22.407
Pt	5.000	5.000
Cu	0.000	4.950
Fe	0.028	0.028
SiO_2/Al_2O_3	5.206	5.206

Remark : Type of zeolites is HY

Table 3.2 Catalysts composition; V_2O_5-TiO_2

Catalyst Number	contents , wt%	
	V_2O_5	TiO_2
1	8.54	91.46
2	15.00	85.00
3	25.61	74.39
4	100.00	0.00

Besides, the fine powder form catalyst of desired catalyst was weighed and then compressed to obtain a pellet. After that, the catalyst pellets were cut and screened to select fragment size between 60/80 mesh. Then fragments were packed in to the middle section of stainless steel reactor (ID. 1.2 cm) for the reaction test.

3.3 Experimental procedure of reaction test

3.3.1 Experimental procedure

The experimental reacion is carried out in the reactor as shown in Figure 3.1. The procedure used to operate this reactor can be described as follows :

1) Helium was first used to purge the system before the start of each experiment for 15 minutes.

2) Temperature of the reactor was raised to 100 ^{o}C and kept constant by electrical heating control unit.

3) After 15 minutes of helium purging, each type of gas reactants was fed to the reactor and the flow rate was measured by adjusting needle valve at the outlet of their cylinder storage, and the total flow rate of feed mixture was measured by bubble-flow meter.

4) Wait until steady state condition was reached. After that, a sample of gas mixture products was taken each at three-ways valve sampling unit.

5) The reactor temperature was raised to another desired reaction temperature, namely, 200, 300 and 400 ^{o}C, respectively.

6) Gas sample was taken to analyze the composition of product at each reaction temperature whenever a new steady state condition had been achieved.

7) Repeat the same procedure (step 1-6) for the other catalysts.

3.3.2 Analysis of gas products from experimental reaction

Analytical determination of the reaction gas composition was carried out using gas chromatography which has a detector as Thermal Conductivity type, TCD, and two types of packing material were used. The first is 50/80 mesh of PORAPAK type Q which was packed in 1 meter long column, and the column temperaure was set around 25^{o}C to analyze NO, NH_3 and H_2O in gas products. The second is 80/100 mesh of MOLECULAR SIEVE 5A packed 2 meters long column, and the column temperature was set at 50 ^{o}C to analyze O_2 and N_2.