

## CHAPTER III

### EXPERIMENTAL

#### Materials

- Used industrial lubricating oil was obtained from BP Oil Co., Ltd.
- Sulfuric acid was commercial available from Fluka.
- Sodium carbonate was commercial available from Fluka.
- Fuller's earth was commercial available from Fluka.
- Isopropyl alcohol was commercial available from Fluka.
- Sodium sulfate (anhydrous) was commercial available from Fluka.
- NiO/MoO<sub>3</sub>/Al<sub>2</sub>O<sub>3</sub> was commercial available from United Catalyst Inc.
- NiO/WO<sub>3</sub>/Al<sub>2</sub>O<sub>3</sub> was commercial available from United Catalyst Inc.
- Raney nickel was commercial available from Merck.
- Universal indicator paper was commercial available from Merck.

#### Apparatus

##### 1. Hydrogenator

The apparatus consisted of four parts as follow:

##### a. Reactor

The reactor was a high pressure batch stirred autoclave model 4551 from the Parr Instrument company with 3750 cubic centimeters stainless steel 316 cylindrical bomb, split ring closures and bomb heater. The reactor could work in a pressure range of 0-2000 psig and in temperature range of 0-450 °C.

The reactor was shown as Figure 3.1.

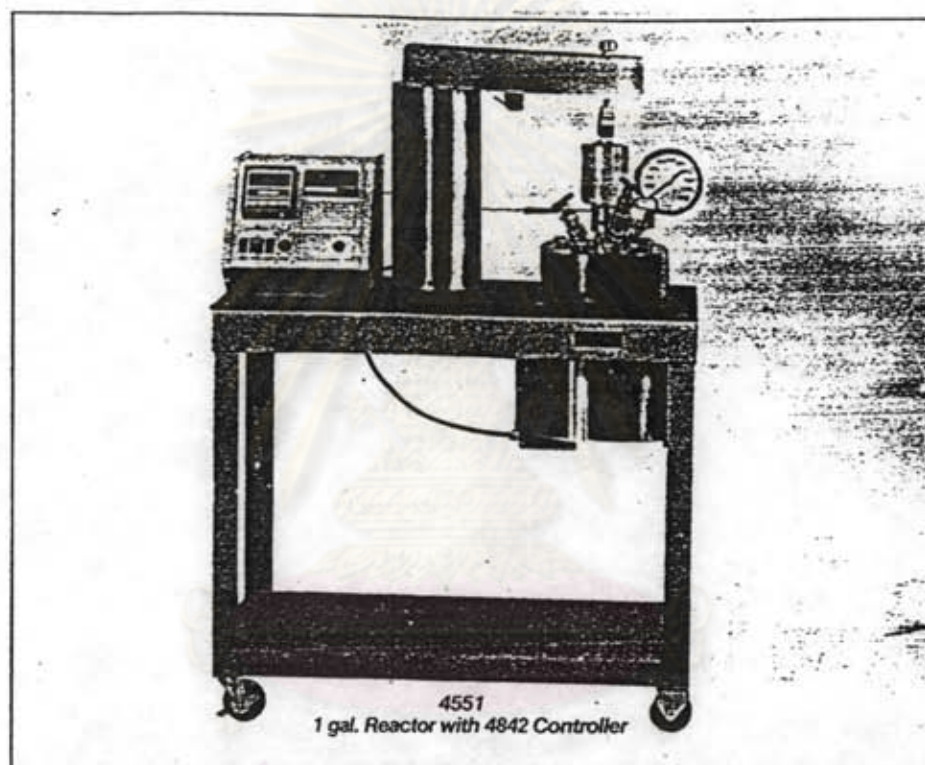


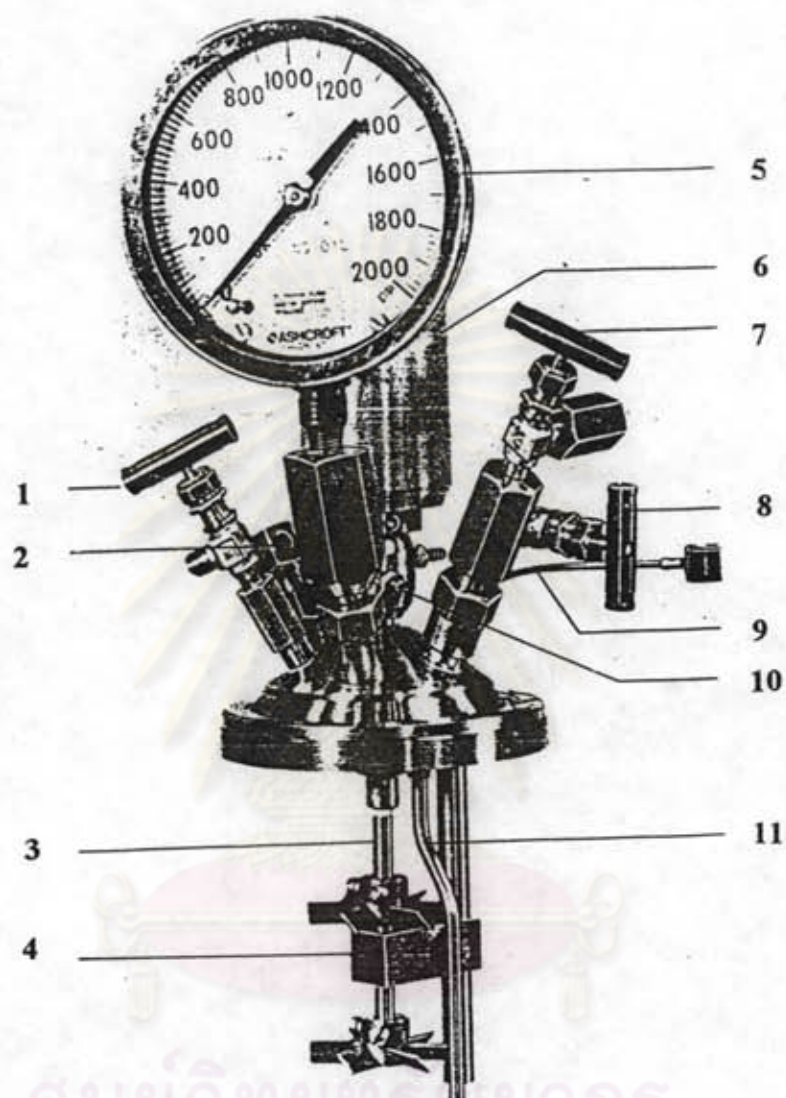
Figure 3.1 Floor stand reactor

b. Reactor fittings

The stirred reactor was equipped with convenient valves and fitting for handling the various functions as shown in Figure 3.2.

c. Automatic temperature controller

The controller used was model 4842 PID controller from Parr



- |                           |                             |
|---------------------------|-----------------------------|
| 1. a gas release valve    | 7. a gas inlet valve        |
| 2. a safety rupture disc  | 8. a liquid sampling valve  |
| 3. a stirring shaft       | 9. a thermocouple           |
| 4. a lower guide bearing  | 10. a water cooling channel |
| 5. a pressure gauge       | 11. a dip tube              |
| 6. a stirrer drive system |                             |

Figure 3.2 Reactor fittings

Instrument company. It was operated with a dual thermocouple. There were various enhancement modules to assist in monitoring and controlling the temperature, pressure, and stirring speed. The stirring speed could be adjusted in the range of 0-1000 rpm.

d. Gas controller system

The system consisted of a hydrogen tank with a pressure regulator (0-200 psig).

2. Viscometer

The apparatus model K-234A from Hochler Instrument Co., Inc. was used.

3. Colorimeter

The fisher ASTM colorimeter was used.

4. Sulfur analyzer

The apparatus model SLFA-800 from HORIBA was used.

5.  $^{13}\text{C}$ -NMR spectrometer

The NMR spectrometer model AC-F 200 from Bruker was used.

6. Thermogravimetric analyzer

The TGA model NETZSCH STA 409 C was used.

## Procedure [35,44]

### A. Physical and Chemical Treatment

- Used lubricating oil was treated by the following steps:
- raw material storage
- water removal
- acid treatment
- sludge settling and separation
- neutralization and washing
- clay treatment
- sludge settling and separation

Five hundred volume of a sample of used lubricating oil obtained from the first (raw material storage) and the second step (water and sediment removal) was heated at 50°C to reduce its viscosity, and to assist mixing with an acid. Then concentrated sulfuric acid (10% by volume of oil) was added to the oil with vigorously agitated to divide the acid into minute particles which was necessary to increase its active surface.

After the reaction had occurred about 2 hours, allowed the mixture to settle down overnight. The acid treated oil was separated from sludge and neutralized with the mixture solution of sodium carbonate (10% by weight) and isopropyl alcohol, this purpose is to neutralize the residual acidity in resulting oil and to extract the sodium sulfonates byproduct.

The oil mixture was then allowed to settle down overnight. The top layer was the neutralized oil diluted with some isopropyl alcohol and water. After the oil was separated, water was mixed to leach some acidity in the oil again. When the pH of water layer was reached a value of about 7, the oil was dried with anhydrous sodium sulfate. Further improvement in color was performed by treating the oil with fuller's earth (10% by weight of oil) at about 80-100 °C and stirred for 10 minutes. Finally, the treated oil was separated from sludge by decantation.

#### **B. Determination of the Physical and Chemical Properties of Treated Oil**

##### **Physical Properties :**

Color, visual (ASTM D 1500)

API gravity (ASTM D 1298)

Pour Point (ASTM D 97)

Sulfur Content (ASTM D 2622)

Kinematic Viscosity @ 40 °C, @ 100 °C (ASTM D 445)

Viscosity Index (ASTM D 2270)

##### **Chemical Properties :**

The percentages of oxidative compounds and oxidation point were determined by Thermogravimetric Analyzer.

The composition (%Cp, %Cn, %Ca) of oils were determined by <sup>13</sup>C-NMR method.

### C. Catalytic Hydrogenation

#### C:1 Effect of Various Catalysts on Hydrogenation of Acid-Clay

##### Treated Oil

The acid-clay treated oil (500 g) was charged into the reactor and the required quantity of catalysts were added. The reactor was then closed. After a thermocouple was inserted into a sturdy thermowell, the stirring motor was connected, and water was passed into the cooling channel. Before running the process, all oxygen gas in the reactor was removed by purging the system with some partial pressure of hydrogen gas (about 10-20 psig) for 2 minutes.

To achieve the catalytic hydrogenation conditions, temperature and time were set by the step controller, whereas the pressure of hydrogen gas was adjusted by the gas regulator. All tests were run under the following conditions.

weight of acid-clay treated oil = 500 g

reaction temperature = 350 °C

reaction time = 4 hours

stirring speed = 550 rpm

concentration of catalyst = 4 %

catalysts used = R-Ni, NiO/MoO<sub>3</sub>/Al<sub>2</sub>O<sub>3</sub>

### **C:2 Determination of the Physical and Chemical Properties of Hydrogenated Oils**

The procedures were the same as in the previous section.

### **C:3 Effect of Various Catalysts on Hydrogenation of Used Oil**

In this step, was the same procedure as in section C:1, used oil was treated under conditions below:

weight of used oil = 500 g

reaction temperature = 350 °C

reaction time = 4 hours

stirring speed = 550 rpm

concentration of catalyst = 4 %

catalysts used = R-Ni, NiO/MoO<sub>3</sub>/Al<sub>2</sub>O<sub>3</sub>

### **C:4 Determination of Physical and Chemical Properties of Hydrogenated Oils**

The procedure was the same as in the previous section.



### **C:5 Effect of Concentrations of Raney Nickel Catalyst on Hydrogenation of Used Oil**

The experimental procedure was done under the following conditions.

weight of used oil = 500 g

reaction temperature = 350 °C

reaction time = 4 hours

stirring speed = 550 rpm

concentration of catalyst = 4, 6, 8 % by weight

catalysts used = R-Ni

### **C:6 Determination of the Physical and Chemical Properties of Hydrogenated Oils**

The procedure was the same as in the previous section.

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### **C:7 Effect of Reaction Times on Hydrogenation of Used Oil**

The experimental procedure was done under the following conditions.

weight of used oil = 500 g

reaction temperature = 350 °C

reaction time = 4, 6, 8 hours

stirring speed = 550 rpm

concentration of catalyst = 4 % by weight

catalysts used = R-Ni

### **C:8 Determination of the Physical and Chemical Properties**

**of Hydrogenated Oils**

The procedure was the same as in the previous section.

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