CHAPTER X.

VISIT TO LEUNA ON NOVEMBER 7th 1938

The following gentlemen were present:

Ringer Michael Duftschmidt Scheuermann Peters Meisenheimer	I.G., Ludwigshafen
Sabel Baehr Becker Kratz Wirth Herold	I.G., Leuna
Mansfeld Ribblet Roberts Johnson	Kellogg Company
Scharmann Spicer Roethelli	Standard Oil of New Jersey
Langen van der Valk	Bataafsche, Shell Development.

We were received by Dr Sabel, who explained briefly by means of a model the lay-out and growth of I.G.'s Plant at Leuna.

A visit was paid to some of Leuna's installations, of which the following are of direct interest to U.S.A.C.:

1. Dr Winkler's Pilot Plant for the production of liquid products from water gas according to his method with oil circulation;

2. a Cowper oven for the production of synthesis gas;

3. Dr Sabel's Pilot Plants for the production of liquid products from a gas consisting of 1 CO + 2 H_2 with catalysts of the same type as used by Ruhrchemie;

4. a Linde-Fränkl Plant for the production of oxygen.

1. The Winkler Pilot Plant was explained by Dr Duftschmidt.

Capacity: 60 Litres catalyst volume, producing 250 kg product per day.
Pressure: 100 atm.
The Plant can also run at 200 atm., giving, in principle, the same yield and same kind of products as at 100 atm. The flow of oil and gas is as follows: a circulating pump, taking suction from the bottom of a separator, pumps the heavier parts of the liquid produced via a steam preheater and a heat-exchanger into the top of the reactor. Gas enters the reactor directly at the top and can be added either cold or hot (in the latter case by preheating it separately). After leaving the reactor the gas + oil passes the heat-exchanger, is then cooled and, thereafter, separated in a separator, from which the circulating pump takes suction.

As gas water gas was used, having a CO-H_2 ratio of 1 : 1.25.

According to Dr Duftschmidt the same kind of products and the same yield is produced as in the installation at Oppau. They hope to be able still to increase the "Leistung".

2. The Cowper installation consists of two towers about 10 m high with a free cross section of 0.5 m2 at the bottom and about 0.8 m2 at the top connected by an empty chamber lined with brickwork. The bottom and hottest part of each tower is filled with high-quality brickwork (Sillimanit), the top part which has a lower temperature with ordinary brickwork (Chamotte). This installation is used for the production of hydrogen or Fischer synthesis gas by first heating up one tower and then introducing a mixture of the gas to be converted and steam or CO_2 into this tower, which, after being converted, gives off part of its heat while passing through the second tower. Air and gas for heating are introduced into the chamber and can be directed through either tower.

The cycle of operation is as follows: The heating gas and air enter the tower to be heated at the bottom, the other tower being shut off during this time. This heating takes 30 minutes. Then steam and/or CO_2 and gas to be converted are introduced in the just heated tower at the top and the converted gas mixture after leaving this tower at the bottom passes through the other tower from bottom to top. Duration: 30 minutes. Then the first tower is shut off and the other tower is heated up for 30 minutes, after which gas and steam or CO_2 enter this second tower at the top and pass through the Cowper in reverse direction, leaving the first tower at the top. Purging with steam is done for only 5 - 10 seconds.

As gas so-called Hy-gas was used, i.e. residual gas from the Hydrogenation Plants, consisting of about 40 % hydrogen, 40 - 45 % methane up to and including butane the balance being CO_2, CO and N_2. One m3 of this Hy-gas produces about 3.5 m3 synthesis gas.

When operating in this way with steam about 4 - 10 g carbon is formed per m3 gas produced. In order to reduce this amount, oxygen is introduced into the converted gas through orifices in the chamber, which lowers the carbon content to 300-400 mg/m3 gas produced. The amount of oxygen is about 10-20 % on Hy-gas.

With steam a gas could be produced from Hy-gas having about the following analysis:

88-89 % $CO + H_2$, with 11-14 % CO
and less than 1 % CH_4.

~~With the addition of CO_2 a gas was obtained~~ containing:

56 % H_2
29 % CO
and again less than 1 % CH_4.

However, with CO_2 the converted gas contained still more carbon.

It proved to be possible to reduce the carbon formation by impregnating the brickwork with certain metaloxides, which could give off oxygen during H_2 making periods and be oxidized again during the heating periods.

The temperatures during the heat period are about 1300-1400°C. I.G. would like to go still higher, but are afraid that the present equipment cannot stand such higher temperatures.

Experiments are being made at the moment to purify such gases by passing them over hot brickwork at 1100°C. Loss of heat is covered by burning about 0.5 - 1.0 % of the gas with O_2.

3. Pilot Plant experiments at Leuna for the production of liquids according to the Fischer process are being carried out with various types of converters.

a. a low pressure converter of about the same construction as Ruhrchemie's low pressure converters, i.e. removing the reaction heat with water flowing through horizontal tubes which are connected in vertical direction by metal sheets. This oven has a volume of 500 Litres catalyst, producing 150 kg liquid product per day. Originally it was thought necessary to preheat the entering gas, which was done with 45 atm. steam. This preheating was discontinued later on, as it was found that the top part of the converter acts as preheater.

When experimenting with recirculation of tail gas, the paraffin produced had a lower melting point, which gave rise to troubles owing to the fact that the cooler was not of sufficient size to separate this paraffin, which then entered the active carbon (A carbon).

This converter has now been in operation since January 1938 without renewal of catalyst. Production is on an average 70 g liquid products per m3. However, the synthesis gas is not always of uniform composition, as this gas is produced by a special experimental water gas plant, having a capacity of about 600 m3 per day and working on both browncoal and bituminous coal. With browncoal gas about 8-10 % lower yield is obtained. It was furthermore found that sulphur is very poisonous and that so-called resin-forming products are also very detrimental to the catalyst.

It is worth noting that it is necessary with this converter to supply heat in order to keep the reaction going.

b. The same type of oven, but of somewhat smaller size, was built in order to investigate the Fischer reaction under pressure. However, it has not been possible to operate this converter owing to the fact that the temperature could not be controlled; even at a pressure of 4 atm. the temperature could not be kept in hand.

The low pressure sheet converter could also have been run under a somewhat increased pressure, but this converter was considered to be too large and not flexible enough for pressure experiments (large consumption of catalyst).

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c. Tubular converters for pressure experiments. First start was made with three such ovens, having 19 tubes per converter, 1.5 m high with a diameter of 15 mm. Cooling of the products is done in 3 stages, viz.:

1. with steam;
2. with hot water;
3. with cold water.

Once every 8 hours a complete analysis is being made.

There are furthermore 11 small tube converters, all having tubes with a height of 1.5 m and diameters of 6, 10 and 15 mm, each converter being equipped with two active carbon absorbers.

Two more ovens having tubes of 4.5 m height and a catalyst volume of 12 Litres were in operation, one running at 8 atm., the other, although being a pressure oven, operating at atmospheric pressure.

A converter with tubes of 10 m height was in course of construction.

Composition of products and yields depend entirely, according to Dr Sabel, on catalyst composition, space velocity, pressure, etc. The usual space velocity was the same as applied by Ruhrchemie, viz. comparable to 1000 m3 ideal gas per Ruhrchemie converter per hour. Yields were in general:

110 - 115 g/m3 ideal gas for atmospheric pressure operation;

140 g/m3 for pressure operation.

One converter having 159 tubes of 6 mm diameter, 1.5 m long, was filled with 5 mm pellets, which were carefully arranged one on top of the other. This converter was operating at 25 atm., producing 130-135 g primary product per m3 ideal gas, 4 % of which consisted of ethyl alcohol. The greater part of the products produced in this case consisted of wax.

We were shown one converter of about 1.5 m height, not operating at that time, which had a peculiar design. This converter had a central tube with water cooling and about 100 small tubes of 6 - 8 mm diameter, subdivided into 5 sections. After each section the products can be analyzed and it was found that even on the first day of operation the total catalyst bed was contributing to the conversion, e.g. the gas entering contained about 3 % of nitrogen; after the first section this percentage proved to be 3.4, after the second section 3.8, after the third section 4.2, etc.

Another converter was still in course of construction. It will have a catalyst volume of 150 Litres, a height of about 3 m, 15 mm tubes and will be able to operate at up to 25 atm.

As an example of Leuna's method of investigating we were informed by Dr Sabel that at present one 1.5 m oven was operating without pressure, one 1.5 m oven with pressure, one 4.5 m oven without pressure, one 4.5 m oven with pressure, all with the same catalyst.

Formerly, regeneration was carried out in situ. However, as the temperature had to be much higher, it was found of advantage to put the whole tube bundle into another larger tube with special arrangement for obtaining higher temperatures.

It takes about 3 months to collect a complete set of data in order to obtain a sufficient idea of the activity and lifetime of a catalyst.

No catalyst is being investigated on pilot-plant scale which has not been found to produce at least 110 g primary product per m3 synthesis gas over a period of 2 - 3 weeks in the laboratory.

600 - 800 different catalysts are at present under investigation in the laboratories at Leuna. About 200 of such catalysts are discarded after one or two weeks' operation on account of not showing sufficient activity and, therefore, replaced by newly made catalysts.

It was found in the laboratory that tubes in horizontal position gave better yields than when installed vertically. In the Pilot Plant the yields were the same whether horizontal or vertical tubes were used. However, it was found that the heat dissipation was better in the case of horizontal tubes.

Gas entering the converters is measured by the usual dry gas meters enclosed in pressure vessels, each being equipped with a small high pressure looking glass.

A special meter was developed at Ludwigshafen, registering the specific gravity of the tail gas. With the aid of an empirical graph it is then possible to determine immediately the methane content of the tail gas.

4. Linde-Fränkl Plant.

This plant was built by the Linde Co. according to their usual design. By far the larger part of the air to be separated into oxygen and nitrogen is passed under a pressure of 4.8 atm. through so-called regenerators, which work in cycle and are cooled down to -160°C by the cold oxygen and nitrogen produced in the fractionating tower.

There are 4 such regenerators, two for oxygen and two larger ones for nitrogen. These gases after leaving the fractionating tower separately, pass each through one of their regenerators, whereas air to the fractionating tower goes through the other two regenerators. After 3 minutes oxygen and nitrogen are passed through the latter two regenerators, air entering through the regenerators just being cooled down. By this way of operation the air entering is freed entirely from water and CO_2 and enters the fractionating apparatus entirely clean, making it possible to run this apparatus for months at a time without any interruption. The regenerators are continuously cleaned from CO_2 and water by the oxygen and nitrogen. These regenerators are completely filled with strips of corrugated aluminium, tightly wound, of about 4 cm height, and put on top of each other in such a way that the grooves pressed in the strips of each layer have an opposite direction to the grooves of the next layer. It is said that the efficiency of these regenerators is 98 - 99 %.

A small part of the air is compressed to 200 atm. and after being freed from CO_2 and water (ammonia cooling), enters the top part of the fractionating apparatus where it is released to 0.5 atm.