6. Hydrogenation Products Separation (b)(cont'd.)

All of these distillation stages were quite conventional in operation, and hence this phase of the coal hydrogenation process is not discussed in detail. Some of the important properties of the various motor fuels and gasolines are presented in tables in Section X of this report, but for more detailed information about aviation gasoline the reader is referred to U. S. Naval Technical Mission in Europe Report entitled "The Manufacture of Aviation Gasoline in Germany" (24) U. S. Naval Technical Mission in Europe Report entitled "German Diesel Fuels" gives more complete information on the characteristics of diesel oils produced by hydrogenation.

7. Sump Phase Solids Removal and Oil Recovery.

(a) Centrifugation of Slurry.

In the hydrogenation of the coal a small fraction was not converted during the passage through the converters, and this together with the coal ash and catalyst had to be removed from the cycle. The most common way was to centrifuge the slurry from the hot separator into a clarified recycle product plus a heavy sludge that contained most of the solids.

The hot slurry from the hot separator, after having been cooled to about 200°C and blown down to atmospheric pressure, as described in Section IV, was diluted with a lighter oil to reduce the viscosity. The solids content in the original slurry was usually about 20-22 percent, and approximately 25 percent of diluent oil was added, so that the resultant solids content was about 16 percent. The choice of thinning oil played an important part in the operation of the sump phase, since the characteristics of the pasting oil could be controlled at this point. The composition of the oil could be a blend of clarified oil from the centrifuging, the residue from the cold separator distillation after removing the middle oil, and middle oil itself. The use of the clarified centrifuge oil helped to reduce the asphalt content in the system, since on a second pass through the machine an additional quantity was removed with the solid residue. The use of some middle oil in the diluting feed helped to thin the slurry more, and at the same time it pre-

7. Sump Phase Solids Removal and Oil Recovery (a)(cont'd.)

cipitated some of the heavier asphalts so that they could be removed from the system. Combinations of these methods were used depending upon the existing conditions at the plant, but in any case it was always necessary to thin the viscous slurry prior to centrifuging.

After dilution the slurry passed to continuous centrifuges where it was separated into an oil containing 6 to 10 percent finely divided solids, and a heavy residue containing 35 to 40 percent solids. The ash content of the solids in the oil averaged about 35 percent, the balance of the solids being asphalt type constituents and fusain. The solids in the residue contained about 50 percent ash plus the major fraction of the unconverted coal. The centrifuges employed were commercial machines, many of them made by DeLaval, which operated on the automatic solid ejection principle. The inside diameter of the housing was about 480 millimeters and the vertical rotating shaft carried about 200 perforated conical disks spaced about one-half millimeter apart. The rotor turned at about 3200 r.p.m. so that a very high centrifugal force of about 10,000 times gravity was obtained. Tungsten carbide valves were employed at the residue discharge to resist the abrasion of the solids. The slurry capacity of these machines was approximately 2.5 tons per hour so that a coal hydrogenation plant had a large battery of them handling the hot separator product.

The lack of complete removal of the solids in the centrifuges was caused by the relatively viscous medium, the low density difference between some of the solids and the oil, and the extremely fine size of many of the particles. The operation of the centrifuges was a compromise between through-put and completeness of purification, since longer contact times resulted in more complete elimination of the solids. It was necessary to remove the residue as a thick slurry in these machines, since too thick a sludge caused the automatic discharge valves to become fouled. This method of centrifuging divided the feed into approximately two-thirds recycle oil and one-third residue, and the large amount of oil in the latter material constituted

7. Sump Phase Solids Removal and Oil Recovery (a)(cont'd.)

a heavy load on the sludge coking plant. The Germans realized the liabilities in the process, had done some research work on the problem, but had apparently not found a better solution.

In 1938 the Leuna plant of the I. G. Farbenindustrie conducted tests on the filterability of the slurries using three different types of equipment, a Wolf cell filter, a cartridge filter, and a Dorr rotary filter.(25) The cartridge type unit gave the highest filtering rate of about 130 kilograms of slurry per square meter per hour, while the cell filter averaged 100, and the Dorr machine only 50. The solids content in the pasting oil, the washing of the filter cake, and the wash oil requirements were most favorable with the cartridge filter. The results of these tests, however, were not sufficiently attractive to warrant the installation of filter equipment in the new hydrogenation plants built in 1940-1943.

(b) Coking of Centrifuge Residue.

As previously mentioned the residue from the centrifuges contained 60 to 65 percent oil, which if it had been discarded, would have been a serious loss. It was therefore necessary to further treat the sludge in order to recover the oil content for recycle in the system. This operation was carried out in coking ovens where the material was heated and the oil distilled off leaving a dry, high ash residue which could be burned by mixing with fresh coal.

The sludges from brown coal, which were low in asphalt were frequently processed in a screw type oven. See drawing no. C-12 for construction and operation of this unit. The apparatus consisted of a gas-fired furnace which contained two steel drums each about one meter in diameter by 16 meters in length, so supported, one over the other, that the upper drum discharged into the lower drum. The feed inlet to the upper drum and the coke discharge from the lower drum were at one end of the unit and the inter-connection between the two drums at the other end. Inside of each drum was a screw that turned at about one revolution

7. Sump Phase Solids Removal and Oil Recovery (b)(cont'd.)

per minute. In the lower part of the furnace a set of steam superheater coils were located. Auxiliaries consisted of a feed pump, and a traveling rake to remove the solids as they were discharged from the lower section of the kiln. The gases and vapors evolved were freed from dust, and were then cooled. The condensed oil was recovered in a series of heat exchangers and coolers, and collected in settling tanks. The gaseous products were used for fuel.

In operation the sludge was pumped through a heat exchanger counter-current to the outlet oil, gas, and water vapors before it entered the oven. Ordinarily the warmed material entered the hollow shaft of the top screw, and passed through the length of the oven where it was discharged into the drum. It was possible, however, to inject the feed directly into the kiln, should the lines become stopped up with material. Superheated steam equivilant to 5-10 percent of the charge was injected into each tube to lower the partial pressure and reduce cracking of the oil. After the residue had been moved by the screw from the inlet end to the rear of the top drum, it dropped down into the lower drum and was carried forward until it finally was discharged into a sump where it was cooled by water. A traveling rake then carried it out and loaded it into cars for disposal.

The oil vapors, gas, and steam together with a small amount of dust left through a connection at the rear of the kilns. The dust was removed in a cyclone separater and was discharged by gravity into a water sump and was pumped or carried away for disposal. The gaseous products were next partially cooled in the heat exchanger by the feed material, and then in to a water cooler. The condensate from these two units was a heavy oil suitable for recycle as a pasting medium. A light oil as well as most of the water vapor was condensed in an after cooler using water and the noncondensable gases were used for heating purposes. The light oil, after separation from the water was sent to the distillation building and blended with the sump phase overhead product to recover an additional quantity of middle oil.

7. Sump Phase Solids Removal and Oil Recovery (b)(cont'd.)

The operating temperature of the oven was about 550°C, and its through-put was approximately two tons of sludge per hour. About 75 percent of the oil was recovered. The major difficulty in operation was coking up of the screws especially in the lower tube. Usually the on-stream time was about 100-120 days for the top section and only 17 to to days for the bottom between cleanings. These furnaces would not handle high asphalt content feeds on account of coking of the screws. Even with relatively good feed stocks the operating time was quite short between maintenance periods.

In order to handle the sludges from bituminous coal hydrogenation plants, a combination ball mill rotating oven was employed. This apparatus consisted of a large drum about two meters in diameter by 11 meters long inclined at about 6° from the horizontal, and mounted in a gas fired furnace. See drawing no. C-13 for flow details of this type of system. The mill oven was lined with high chrome abrasive-resistant steel and held about 9-10 tons of balls. The feed and part of the steam were injected through pipes in the hollow trunnion at one end, and the remaining steam was introduced at the opposite end in a similar manner. The vapors left through the trunnion at the feed end of the oven. The solid coke was removed at the end opposite the feed and collected in a water-sealed sump. In the upper section of the furnace setting were coils for superheating steam and also for preheating the feed. The auxiliaries were essentially the same as those employed for the screw type furnace, the dust collector and the heat exchanger being combined into one unit.

The operating temperature was usually a little higher, about 560-590°C, than that used for the screw coking oven, but the oil recovery was essentially the same. About 10-15 percent of steam was added to the charge to reduce cracking and gas loss. These ovens coked up in about five days of operation, but due to the grinding action of the steel balls they were self cleaning by merely shutting off the feed and continuing the rotation for 10-12 hours.

The sludge through-put was considerably greater than

7. Sump Phase Solids Removal and Oil Recovery (b)(cont'd.)

in the screw-type furnace, 3.2 tons as compared with 2 tons per hour. Another big difference was the much lower oil content in the residue, about 1-2 percent as compared with 20-25 percent. The higher temperature produced more gas so that the overall oil recovery was essentially the same. It was of interest, however, that the ratio of heavy oil to light oil was greater in the ball oven than in the screw type, despite the fact that the gas yield was almost double. The ratios of heavy to light oils were 4.8 and 3.4, respectively, from the two units operating on the same feed.

Table No. 8 in Appendix A contrasts the operation of these two types of furnaces.

8. Description of the T.T.H. Process.

The T.T.H. process or Tief Tempratur Hydrierung (low temperature hydrogenation) represented a markedly different approach to the hydrogenation of coal tar to obtain diesel fuel and lubricating oils. In 1935 experimental work was started on the problem of preparing lube oils from brown coal tar by means of a limited hydrogenation so as not to destroy the paraffinic nature of the tar. By 1938 the work had reached the stage for commercial application, and in 1939 the first and only plant in Germany was started at Zeitz.

The feed for the process consisted chiefly of brown coal tar from low temperature carbonization with about 10 percent of light oil added. The raw tar contained about 0.5 percent finely suspended dirt and 0.4 percent water which were largely removed by centrifuging. The clarified tar was next filtered and the dirt content reduced to a maximum of about 0.01 percent. An analysis of the tar feed showed that it contained 83.4 percent carbon, 10.5 percent hydrogen, 2.0 percent sulfur, 0.4 percent nitrogen, and 3.7 percent oxygen. The specific gravity at 50°C was about 0.95, and analytical distillation showed that 6 percent boiled up to 180°C, 33 percent up to 325°C, and 52 percent up to 350°C.