Coal carbonization. - Dr. Reerink is one of the outstanding authorities on coal carbonization, having for some years been associated with the Verein für Überwachung der Wärmewirtschaft zur Ruhrzechen and having collaborated with Drs. Kurt Baum, W. Litterscheidt, and Steinschlager. The work on this subject for the Bergbau-Verein, under Dr. Reerink, is headed by Dr. Scheer, who has associated with him Dr. Lameck and Obering.Sassonhoff. The experimental work has as objectives standardization of coke oven plants, thermal efficiency of operation, development and examination of new processes, broadening the classes of coal that can be coked, quality of blast-furnace and foundry coke, and recovery of byproducts.

There is still no agreement on what is good coke. Many laboratory tests were developed in the period when there was an excess in the supply of coke, but as the demand for coke increased, investigations of new methods of characterization of coke decreased, and a wide range of physical properties was accepted as between different districts. Specifications of coke no longer included special physical tests, and even standard tests such as the Micum test were not emphasized; requirements generally included only percentages of ash, sulfur, and phosphorous. A few laboratories used the Wolff pressure test and virtually none the ASTM shatter test for determining coke strength. An investigation of the qualities desirable in foundry coke was planned at the beginning of the war; but it was only started in 1944 and was soon discontinued because of lack of facilities. The main problem for foundry coke was to learn how to make coke in the more modern narrower ovens similar in quality to that made in the older wide ovens. The coke made in the narrow ovens required higher pressures in operation of the cupola than normally available. The Verein der Deutsche Eisenhüttenleute (Dr. Rummel) had in recent years discontinued their research on coke quality. Coking tests were considered to give only qualitative information on the quality of coke made from blends of coals, and quantitative correlation with strength of coke produced commercially was not possible; use of such tests declined during the war.

In Dr. Reerink's opinion, ovens 4 to 4.5 meters high and 450 millimeters wide were best for Ruhr coals. Ovens 6 meters high could be justified only where ground space is limited and additional capacity is needed. Such ovens were built by Still at Nordstern (Gelsenkirchener Bergwerks A. G.) and could be uniformly heated and operated without mechanical difficulties, contrary to other reports. In the past 10 years, however, emphasis had been on byproduct recovery rather than oven construction. The Still process was regarded to have value only with the right combination of coal, oven temperature, and other operating conditions; it had been tried in three or five plants in the Ruhr, but was discarded; only one plant in the Saar, Röchling, was known to be using the Still process, and thereby obtained an increase of 20 to 30 percent in light oil. The light oil is high in paraffins and not suitable for nitration but only for motor fuel.

The Gesellschaft für Kohlentechnik has continued its studies of byproduct recovery and utilization under Drs. Grosskinsky, Klempt, Umbach, Keller, and Adelsberger. A method of recovery of thiocyanate developed by them has been installed at Zeche Graf Moltke (Gelsenkirchener Bergwerks A.G.)

in Gladbeck. Other studies include methods of recovery of byproduct ammonia and its tranformation to fertilizer without the use of sulfuric acid, recovery and utilization of hydrogen sulfide from coke-oven gas and ammonia liquor, and recovery and utilization of hydrocyanic acid. Production of synthetic resins from both phenol and urea is also being studied. The Berichte der Gesellschaft für Kohlentechnik was published irregularly during the war, the last volume, V, being issued in three parts in September 1939, May 1940, and January 1942. Dr. Reerink recommended the following list of publications as worthy of study for learning about recent developments in coal carbonization in Germany:

Die Ölausbeute bie der Verkokung, by W. Reerink. Glückauf, 1942, vol. 78, pp. 597-602.

Erfahrungen bie der Verkokung und Schwelung von Saar und Lothringischer Kohle, by W. Gollmer. Stahl und Eisen, 1942, vol. 62, Heft 38, pp. 789-795.

Die Wärmetechnische Beurteilung Neuzeitlicher Koksofen, by W. Litterscheidt. Archiv für bergbauliche Forschung, November 1941, pp. 101-124.

Die Bedeutung der Trockenen Kokskühlung für den Kokereibetrieb, by W. Scheer. Stahl und Eisen, 1944, vol. 64, Heft 4, pp. 53-62.

Das Treiben der Steinkohlen bie der Verkokung, by K. Gieseler. Glückauf, 1941, vol. 77, pp. 309-319, 328-332.

Die Methoden zur Bestimmung der Kohlenwertstoffausbeuten bie der Verschwelung und Verkokung der Steinkohle, by P. Hoffman, Feuerungstechnik, 1941, Heft 9, pp. 205-209.

Die Methoden zur Bestimmung der Bildsamkeit von Steinkohlen, by P. Hoffman. Feuerungstechnik, 1942, Heft 11, pp. 249-255.

Fortschritte und Erkenntnisse in der Verarbeitung des Destillationsgases der Kokereien, by W. Gras. Glückauf, 1942, vol. 78, pp. 57-61, 73-76.

Gasschwefelgewinnung nach dem Ammoniakverfahren auf der Kokerei Reden, by H. Lohrmann and P. Stöller. Archiv für bergbauliche Forschung, July 1942, pp. 43-48.

Oxidation of Coal. - Since 1942, considerable work has been done on production of materials of interest to the chemical industry by controlled oxidation of coal. This work is directly under Dr. Grosskinsky's supervision with assistance of Dr. Klempt, Dr. Lange, and Dipl. Chem. Jüttner (B. Jeuttner, Coal Research Laboratory, Carnegie Institute of Technology 1930-1939.) Coal is first oxidized with air or oxygen until the caking power is destroyed, the reaction being carried out slowly at the start to prevent ignition. Steam in amounts of 10 to 50 percent is added to the gas stream for controlling the reaction rate. With oxygen, the maximum temperature used is 220° C., and with air, 300° C. The time of reaction depends on the coal used, a typical result being 30 hours at 250° in air for coal sized between 10 and 20 millimeters, or 15 hours at 250° in air for 1 to 2 millimeter coal. The product can be activated to give an active carbon, which may be reacted with sulfur to produce carbon disulfide, or it may be carbonized at low temperatures to yield an active raw material for gas manufacture, or it can be treated with chemical reagents to yield organic acids. The latter studies were carried out at the R.W.K.S. laboratories in Essen by Dr. Lange, whereas studies of utilization of the acids produced were carried out in Dortmund-Eving by Mr. Jüttner. Patent applications have been made since 1942 on these processes.

Several different chemical oxidizing agents were studied, but most work was done on a preferred process with oxygen and nitric acid. With chlorine and oxidized coal at 100° C., the reaction required 1 week and yielded 30 percent organic acids - work discontinued. About the same yield was obtained with alkaline permanganate, but separation of the acids was difficult and too expensive. It was reported that oxygen and aqueous alkali at 420° to 450° C. and 300 atmospheres total pressure gave a yield of 60 grams of acids per 100 grams of oxidized coal in 3 hours; the aqueous alkali was of 10 to 30 percent concentration of sodium or potassium hydroxide, lime being unsatisfactory; work was discontinued because of cost of equipment and difficulty with filtering, recovery of free acids, and regeneration of alkali. Other reagents were tried but showed no promise.

The nitric-acid oxidation was made with nitric acid of 1.2 specific gravity, with recirculation of the oxides of nitrogen evolved plus added oxygen to regenerate the nitric acid. At normal atmospheric pressure and a reaction temperature of 110°, the time of reaction was 2 days, and at 5 atmospheres and 140° this time could be cut in half. The reaction is carried out in glass, enamel-lined, or V-2A steel (18-8 alloy) equipment. It was claimed that 1 part of coal to 10 parts of acid could be used in a continuous process, regenerating the acid with a loss of only 1 percent and adding fresh coal with removal of crystals of the acids from the reaction vessel continuously. Equipment was built to produce 5 kilograms of acid per day but only operated 8 days before bombing forced its discontinuance in March 1944.

From 100 grams of oxidized coal, 100 grams of acids and 100 grams of other products were obtained. The acids, containing about 50 percent by weight of the carbon of the original coal, were reported to consist principally (80 percent) of mellitic and pyromellitic acids containing a trace of picric acid. The acids were considered to be a new raw material of promise for the resin and lacquer industries; when esterified, as plasticizers for synthetic (Buna) rubber; and when esterified and transformed to ethers, as lubricants or additives to lubricants. The cost of production of the crude acids was estimated at 0.10 M per kilogram (sic!) which was stated to be equivalent in cost to phthalic acid. Amylesters had been prepared and tested by Continental Gummi in Hannover, who requested an additional 10 kilograms for further tests. Higher esters were said to be difficult to prepare because of cracking. The acid number of the esters was said to range from 1 to 5, purification being achieved by filtering through lime. Work is to be resumed as soon as possible.

Coal-Oil Mixtures. - This work was under direct supervision of Dr. Grosskinsky and had two objectives. The Navy was interested in the "colloidal fuel," but difficulties were experienced in transportation, owing to sedimentation. Fine grinding to give true colloidal suspensions yielded a product too viscous to handle at 30 to 40 percent concentration of coal. It was reported that the work was done on a 10 kilogram per day scale and that a plant of 30 tons per day capacity had been built but not operated, as the facilities were bombed out.

The coal-oil suspensions had also been investigated as a means of preparing low-ash coal for electrode carbon and possibly hydrogenation by using them in a process similar to that proposed by Trent of the United States in about 1917. The work had been carried out on laboratory scale only, but the results were regarded as sufficiently promising so that large-scale work had been planned but not carried out.

Furnace Technique and Coal Gasification. - Several problems on these subjects were reported to be under investigation, supervised by Dr. Gumz of Berlin, although the work was not mentioned in discussions either at the Bergbau-Verein or the Gesellschaft für Kohlentechnik. The problems included development of high-capacity furnaces of smaller and greater sizes, development of steam-driven road vehicles, examination and further development of known gasification processes, development of new gasification processes, e.g. dust gasification as being tested at the Nitrogene Works at Wanne-Eickel, and examination and development of small gas producers.

Technical Mining Department. - The head of the department is Dr. Ing. W. Vogel, who is assisted in planning the work of the department by Dipl. Ing. Rauer and the Committees on Mining Technique, Mining Technical Economy, Mining Supply, and Power Economy. Little of value was gained on interrogating the personnel familiar with and active in underground mining research, as apparently much of the research pertains to improved designs of existing equipment, except for the "oscillating conveyor." This is a face conveyor built in 6-meter sections, each one with separate drive motor 1-1/2 kw. It was stated that vibrations are 800 per minute, that material flows 80 centimeters per second, capacity is 200 tons per hour, there is no loss in capacity on pitches up to 5 degrees and a 30 percent loss on a 12-degree pitch uphill, and that 20 such units were planned for experimental work. A drawing of a section of the oscillating conveyor and a 25-page mimeographed copy (unpublished) of German Mining Developments during the war are contained in Technical Oil Mission microfilm reel No. 63 (original designation No. 34A).

A group of mining problems was reported to be under supervision of Dr. Glebe and Dipl. Ing. Rauer, including ascertainment of the most favorable division of mine fields and of size and location of surface plants, improvements in drilling, shot firing, loading, and driving of headings; standardization of steel supports for main roads; improvements of steel props; and testing and development of new support elements.

Under Obering. Cuyler and Bergassessor Dr. Ing. Schluter are investigating coal-mining machinery, including development of existing machines and testing of new machines.

Dr. Ing. Passmann is in charge of work in connection with electrical power economy underground. He also, assisted by Dipl. Ing. Schmitt, and working in collaboration with the wire rope-testing institute in Bochum, the coal and iron research institute in Dortmund, and the machine technical institute of the Mining Academy in Clausthal, and various manufacturers, has charge of standardization of machines by type and by kind, of parts of machines, haulage arrangements, electrical fittings, and other appliances and lubricants.

Advice to the collieries, one of which is shown in figure 8, on power economy is provided by Obering. Presser and Prof. Dr. Koch, who are concerned also with improvements in the use of underground locomotives.

<u>The Laboratory of the Gesellschaft für Kohlentechnik at Dortmund-Eving.</u>- The facilities of the laboratory prior to damage by bombing, as judged from inspection, were excellent for experimental investigation of a wide range of problems. A workshop was available for construction of special equipment, including high-pressure autoclaves for operating up to 3,000 atmospheres. An engineering department was available for designing large-scale equipment from data obtained in the laboratory, such large-scale work being done largely in plants of interested concerns. The library, which had been evacuated to a mine in Minister Stein colliery across the street, was said to have been one of the most complete on fuel technology in Germany. The staff that could normally be accommodated was stated to be 80, of whom about 50 were still available. Work was seen in progress in only two laboratories and was on chlorination of hydrocarbons for production of "invert" soaps. The most serious shortage at present, aside from usable space, was stated to be trained chemists, many of whom had left for the Army and not yet returned.

Dr. Grosskinsky told of work done between 1938 and 1942 on indirect fuel cells using gas. The cells operated at 800 degrees to 1,000 degrees and higher and gave about 1.1 volts on open circuit, which dropped to about 0.8 volt on load. The work was done by Dr. M. Blanke and Dr. J. Karweil, Guttingen, in collaboration with Prof. Emil Bauer of Zurich. Patent applications were filed but allowed to lapse because of impracticability, unavailability of materials of construction, and the expressed feeling that such methods of power generation are not economically feasible for at least 20 years. No technical reports on this work were issued, and all records were still in a mine at Minister Stein colliery.

COAL AND COKE RESEARCH, H. KOPPERS G.m.b.H., ESSEN[19/]

Conclusions

The laboratories are largely inactive at present, and according to Dr. Koppers, there was neither personnel nor time to do much research in recent years. An exception was work done both on laboratory and larger scale on gasification of pulverized coal with steam and oxygen. The method was said to offer promise, but all records and data were said to have been removed previously by an officer of the United States Navy.

Report of visit. - A tour through the laboratories, which normally accommodated 25 persons, showed almost no activity nor equipment of interest. Tests to determine whether a coal could be safely carbonized in a coke oven without damage to oven walls from coal expansion were made regularly in the Koppers small-scale test oven as described in Koppers Mitteilungen, Heft 1, 1930. Interpretation of the data obtained in this test was aided by development of a medium and a large-scale test to determine scale factors, but use of these larger pieces of apparatus has been discontinued. It was stated that the pressure gage developed by Dr. F. Ulrich of the Rheinisch-Westfälisches Kohlen Syndikat at Essen was not used nor highly regarded by Koppers, though it may have been investigated by Dr. A. Jenkner, a former associate of Dr. H. Koppers some years previously. The advantage of the Koppers test over those developed by Damm, Nedelmann, Macura, and others was the experience behind the test that gave assurance that the test data were interpreted according to commercial practice.

Although Koppers is not further interested in coking preheated and dried coal and has not done work similar to that of Didier Werke, as reported by Dr. W. Litterschedit in Glückauf, 1935, p. 173, they do have experience in this field. Koppers built the plants for production of Carbolux in Bruay and Carmeaux in France, which still charge hot dry coal to ovens of 350-millimeter width. They also built a plant at Barsinghausen, west of Hannover, with ovens 3.7 m. high, 10.9 m. long (chamber length), and 200 to 230 millimeters wide, in which dried coal was originally used and flue temperatures were maintained at 820 to 850 degrees. The time for carbonization under these conditions was 17 to 18 hours. More recently, the flue temperatures have been raised to 1,100 to 1,150 degrees and, using undried coal, the time of carbonization was reduced to about 12 hours. This latter procedure is advantageous, as the coke produced is as good and commands the same price as that produced when using lower temperatures and preheated dry coal. It was admitted that the capacity of the coke ovens could be increased by preheating and drying, but their experience would not permit estimation of the percentage increase owing to the different flue temperatures used in the operations described above.

19/ Prepared from Combined Intelligence Objectives Subcommittee report "Coal and Coke Research, H. Koppers G.M.B.H., Essen," by H. H. Lowry and H. J. Rose, 1945. (C.I.O.S. No. XXXI-31, Item No. 30; Solid Fuels No. 28).

The highest coke oven which Koppers has built is 4.5 meters, and they do not care to build them higher because (a) the greater packing of coal would increase the danger from expansion during coking and (b) it is hard enough to keep 4.5 meter-high doors tight. He felt that they could heat higher flues evenly with the proper circulating system.

In reply to questions on pulverization of coal for carbonization, sizing of the coal was stated to be of special importance with poor coking coals where stamped charges are used. While the sizing specification requires only that 80 percent pass a 2-millimeter screen, this should give about 50 percent through 1 millimeter, and it is desired that 40 percent be below 0.5 millimeter. If these figures are not obtained, although the 80 percent through 2 millimeter is realized, adjustments are made to the crushing machinery to give the desired size distribution.

Koppers have no new developments in low-temperature carbonization, although they have a plant in Upper Silesia that produces in continuous vertical ovens a coke of 8 percent volatile-matter content from nut coal or briquets of noncoking coal. The oven walls are of ceramic construction, and the flue temperatures range from 900 to 1,000 degrees. Dr. Koppers did not regard highly the Krupp-Lurgi LTC system because of the metal walls. He stated that construction had been started by Brennstoff-Technik at Kleinrosseln near Vorbach in the Saar but that a Püning movable-wall coke-oven installation with metal walls had not been completed. The Berlin test plant of the Püning oven was not regarded as promising by H. Koppers. Other test ovens were built at Altenessen and in Hindenburg, Upper Silesia. Semi-coke was not considered to be economically attractive as a domestic fuel because of the prices allowed by the syndicates: Semi-coke was priced the same as high-temperature coke (22 marks per ton) rather than the same price as anthracite (28 marks per ton).

Desulfurization other than by mechanical cleaning of coal for carbonization had been carried out only on a laboratory scale.

It was stated that physical testing of coke was standardized and had not changed for many years. In general, coke was not bought by specification but from coke plants associated with mines that produced coke of a quality that experience had shown was satisfactory from the standpoint of the end use.

Control of the temperature of the free space above the coal charge in a coke oven was said to be important in determining the yield of naphthalene and the paraffin content of the light oil. H. Koppers have done some work on differential heating for this purpose, but the work was discontinued during the war.

Recently, work that appeared to hold promise was carried out, both on a laboratory scale and on larger scale, on gasification of coal with steam and oxygen. Records of this work and the data were reported to have been taken by an officer of the United States Navy who was thought to have formerly been connected with the Chicago By-Products Co.

With regard to coal dryers, Koppers referred to a new-type dryer manufactured by the Büttner Co. at Urdingen, near Krefeld, as well as a direct-fired dryer of the Haas Co. in Lennep, south of Wuppertal-Barmen.

FUEL-RESEARCH ACTIVITIES, THE A. G. DER KOHLENWERTSOFF-VERBANDE BOCHUM[20]

Conclusions

The management, as represented at the interview, appeared most cooperative and eager to supply any information desired. Dr. Haver speaks English fluently and should be an excellent source of detailed information regarding specific questions with respect to production, distribution, and sale of all byproducts from the carbonization of coal.

Structure. - The A. G. der Kohlenwertsoff-Verbande is made up of seven companies that controlled the sale of byproducts of coal carbonization in Germany and in the export trade. It is also the largest distributor of petroleum products in Germany, operates two plants of its own for the manufacture of sulfuric acid from pyrites and two plants for making pitch coke, and maintains a central laboratory (now bombed out), which had a staff of about 100, of whom 10 are graduate chemists. Most of the laboratory work is on quality control in connection with sales and is not on coal itself, but on byproducts. The normal staff of A.G.K.V. was about 4,000 persons, of whom 1,200 were in the administration at the Bochum headquarters, the rest comprising the sales force. It was stated that 500 to 600 of the staff were still available, including a large proportion of the laboratory technicians.

Synthesis of toluene. - It was reported that work had been done on the synthesis of toluene from benzene and methyl alcohol over phosphoric acid. Xylene was obtained as a byproduct.

Pitch coke and electrode carbon. - In addition to owning and operating two plants for production of pitch coke, one using ovens designed by Paul Hilgenstock and the other using the American Koppers system, which was said to be the preferred process, the A.G.K.V. sold almost all the electrode carbon produced in Germany from coal. They sold 139,745 tons of pitch coke, including fines, in 1943, and in 1944 it was planned to increase capacity to 175,000 tons per year but air raids prevented. The ash content ranged from 0.4 to 0.6 percent, and the selling price was 90 marks per ton. About 12,000 tons of Pott-Broche coke with an ash content of 0.1 percent was sold in 1944 and the price was 180 marks per ton. The question was asked, why does Pott-Broche coke with 0.1 percent ash bring twice the price that pitch coke having 0.4 to 0.6 percent ash bring? The reply was that in wartime it was necessary to make a special high-quality aluminum as a substitute for copper, and coke made from the Pott-Broche coal extract permitted this. They doubted that the aluminum industry would pay as high a premium for extremely low-ash coke in peacetime. They also sold about 12,000 tons of coke prepared from coal

20/ Prepared from Combined Intelligence Objectives Subcommittee report "Fuel Research Activities, The A. G. der Kohlenwertsoff-Verbande Bochum," by H. H. Lowry and H. J. Rose, 1945 (?). (C.I.O.S. No. XXXI-29, P. B. 4324.)

cleaned by flotation at the Koenigin Elisabeth mine in 1944 and which had an ash content of 0.7 to 1.0 percent and sold at 125 marks per ton; the price wanted by the producer was 140 marks per ton, but this was not allowed. An unstated amount of coke prepared by acid-cleaning of coal at the Karl Alexander mine and which had an ash content of 0.8 to 1.2 percent ash, was also sold, though not by A.G.K.V., for electrode manufacture at a price of 80 to 90 marks per ton.

Solvent refining of coal tar. - They had studied, but only on a laboratory scale, the solvent refining of coal tar with benzene, aniline, furfural, acetone, methanol, etc.

Weather-resisting road tar (VFT Wetterteer). - It was reported that road tar in Germany was 50 percent derived from coal tar and 50 percent from asphalt, total sales amounted to about 300,000 tons per year, of which about 9,000 to 12,000 tons was a special weather-resisting tar having a melting point of 32° to 35° C., described in the following two paragraphs, which are a translation of a special report on this subject prepared by Dr. Giesselbrecht, the original of which is filed in Technical Oil Mission (T.O.M.) microfilm reel No. 70 (original designation No. 25B) near the end of the reel as 000895, item 1 (2 pages).

> "The need which led to the development of Verkaufsvereinigung fur Teererzeugnisse (VFT), "weather-resisting" tar, was that of a practically unchangeable street-paving tar, which would be resistant to weather conditions prevailing in Germany. The changes which the tar naturally undergoes after incorporating into the road surface were to be effected in advance. This was to be achieved by subjecting the oil from which the tar was to be produced to a special preliminary oxidizing treatment. In practice the process is carried out by blowing air through normal anthracene oil at 160 degrees until the viscosity has reached about three Engler degrees at 50 degrees. The oil so obtained is then mixed with normal pitch of melting point 60 to 75 degrees (Krämer-Sarnow). A product is obtained that is less sensitive to moisture and exhibits smaller changes in use.
>
> The weather-resisting tar contains at most 1 percent middle oils, 5 percent heavy oils, and absolutely no phenols or naphthalene. The free-carbon content is under 18 percent. The drip point is about 30 degrees Centigrade, the freezing point below 25 degrees Centigrade. Thus, the zone of plasticity extends over 55 degrees. The security against melting in heat and becoming brittle in the frost is greater than with a smaller span. The viscosity curve shows that the weather-resisting tar is still extremely viscous at 50 degrees Centigrade but has reached the thin liquid state of normal surface tar at 100 degrees Centigrade, and at 105 degrees Centigrade has the same degree of viscosity as the latter. The pitch content does not exceed 60 percent. At temperatures prevailing during the day, the weather-resisting tar is a semisolid mass. For this reason it is possible to