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RECENT PROGRESS IN FUEL TECHNOLOGY

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SUMMARY.

The great volume of work now in progress in the United States, for the production of oil from sources other than crude petroleum, is the result of a shortage of oil which persisted after the war until only a few months ago. Normal increases in the consumption of motor spirit, and the extensive substitution of oil for coal previously burned in locomotives and household furnaces, have combined to create a demand with which production has not been able to keep pace. Crude oil has been in short supply because, during the war, steel was needed more urgently for other purposes than for the drilling of wells and laying of pipe lines. However, the intensive drilling campaign instituted soon after the war has now borne fruit. From a country which was, on balance, an importer of oil to the extent of 500,000 barrels per day, the United States has become an exporter. A large volume of potential production has been closed down in Texas, Venezuela and elsewhere.

Because of the high cost of new production, future drilling activity is likely to be sensitive to market demands for oil. Prices of crude and petrol are likely to remain, therefore, substantially at their present levels. There are indications that the consumers' market will result in a rise in octane ratings in America, after a long period of stability. No early or substantial rise is expected in England, where appreciation in octane number is being resisted for economic reasons.

There has been a marked change, also, in the defence position of U.S.A. in respect of oil supplies. The discovery of a large field in Alberta has made the United States no longer dependent upon sea-borne supplies of petroleum to meet a possible wartime demand for large quantities of oil.

These considerations have caused a slackening of interest in the production of oil from shale, coal and natural gas. Research and development are proceeding, nevertheless, but not with any prospect of early application. It is desired, rather, to complete the work undertaken, and then merely to file the information for reference at some much later date.

The Bureau of Mines is engaged on an extensive study of oil shale and shale oil. Technique is being developed for mining a 70 ft. seam at Rifle, Colo. by underground quarrying. A new retort using the "Gas Flow" principle, is being developed and a small refinery has been installed for processing the crude oil by standard methods. In the laboratories at Laramie, Wyo, the subjects being studied include the nature of shale and shale oil, refining processes, solvent extraction and by-products. From the information which has been gained to date, it has been calculated that motor spirit from Colorado shale could be made available in California - which would be the natural market for the product - at from 16 to 19 cents per gallon, according to the process used. In all cases, it is assumed that operations would comprise mining, carbonising and viscosity breaking on the site, piping the oil to California and there refining it by a combination of hydrogenation and other processes.

Standard Oil Development has applied its fluid bed technique to shale carbonisation. Union Oil, also, is developing a retort which is, in effect, an inverted N.T.U. plant. Both these retorts, like the Gas Flow retort previously mentioned, give promise of high yields of oil and high capacities but have not yet been developed to a stage at which commercial application could be safely contemplated.

Because of its high nitrogen content, crude shale oil is not amenable to catalytic cracking. The hydrogenation process developed by Union Oil for the refining of Californian crudes of high nitrogen content is applicable, however, to shale oil. Because shale oil is of inferior quality and difficult to refine, it is considered that it should be utilised as far as possible for fuel oil, and that petroleum oil should be further refined to maintain the desired balance between "clean" and "black" products.

<u>Carbonisation of coal at low and medium temperatures</u> is being practised on a small scale in England and the United States, and is being further studied mainly for the production of smokeless fuel. In all cases, there is a wide margin between the cost of the raw coal and the price realised for the product at the works. The coke is usually worth rather more

han twice as much as the coal. The main demand in England is or a fuel for open fires, but hard, carbonised briquettes are lso in demand for enclosed stoves, foundries etc. In U.S.A., much attention is being given to the suppression of smoke, and supplies of low temperature coke, anthracite etc. are inadequate. The production of hard, carbonised briquettes is being investigated. In some cases, the raw coal is non-coking; in one case it is a coking coal which is too highly swelling for charging to a coke oven. The briquettes are needed for water gas plants, cupolas etc., because of the very high price of oven coke and anthracite.

In the field of complete gasification of coal, the main developments of recent years have been the application of the fluidised technique to the process and the gasification of pulverised coal in suspension. In the fluid bed process, coal is usually carbonised and the char gasified in successive stages, though the two stages may be incorporated in the same plant as practised by I.C.I. The other organisations currently working on the technique are Standard Oil Development, Pittsburgh Consolidation Coal, Hydrocarbon Research and the Anthracite Institute. For the gasification of anthracite, of course, preliminary carbonisation is unnecessary.

It would appear that satisfactory progress has been made in the carbonisation of fluidised coal; the gasification stage, however, still appears to be in an early stage of development. The authorities working on the problem are not yet ready to disclose results or the amount of progress made. Opinion is divided on the possibility of success with the process. It is pointed out that the progressive consumption of particles in the fluid bed introduces problems which are absent in other applications of the technique.

The main purpose for which fluidised gasification is being developed is the production of gas suitable for synthesis reactions. For such use, the gas is of considerably increased value if it is generated at reaction pressure or other high pressure. In America, accordingly, pilot plants operate, or are intended to operate, under pressure with oxygen and steam as reagents. The I.C.I. process, on the contrary, is devised to consume air instead of oxygen, on account of the high cost of oxygen for industrial purposes. The process, therefore, is restricted to application at atmospheric pressure.

The Bureau of Mines has studiously avoided fluidised gasification because the process is being adequately investigated elsewhere. Instead, it has undertaken to study three other methods of gasifying coal for synthesis purposes, viz. the Koppers method using powdered coal entrained in oxygen and steam, the F.R.S. vortex process also using pulverised coal, oxygen and steam, and a third process, devised in the Bureau, in which steam is superheated to a temperature so high that it carries with it the heat needed for the reaction. The Institute of Gas Technology, also, is working on complete gasification of pulverised coal with oxygen and steam. Even the most advanced of these processes, viz. the Koppers, must be regarded as inadequately developed.

The Bureau is investigating the production of synthesis gas by the complete gasification of lignite in an externally heated retort. The underground gasification of coal is also being studied, though in this case the gas is desired for firing boilers or perhaps gas turbines.

Further work, largely academic in nature, is being directed towards improving the capacity of water gas generators. The gas Research Board, however, is undertaking plant scale tests for a better utilisation of the waste heat available from the process.

There is little interest in the Lurgi gasification process in England and only mild interest in U.S.A. The Bureau of Mines has a small plant at Pittsburgh doing good work within its sphere of application. The Bureau contemplates installing a larger unit elsewhere. Opinions were sought on the application of the process to Victorian briquettes. The replies received were preponderantly in favour of the project.

Texaco Development states that it has ready for adoption a process with all the advantages of the Lurgi, but superior to it in certain respects. Little else is known of the Texaco process.

The Gas Research Board is working on a process for the manufacture of gas of high calorific value by the formation of methane from coal and hydrogen under high pressure.

Many of the complete gasification processes just mentioned are dependent for their economic success upon medium purity oxygen being available in large quantities and at relatively low cost. Since the war, many variants of the classical Linde Frankl process have been developed in U.S.A. and elsewhere. They are directed principally to the elimination of the high pressure circuit previously used to provide supplementary capacity for volatilisation of heat exchanger deposits. Many ingenious devices and circuits have been applied to that end, but the resulting improvement, if any, on the Linde Frankl process is quite small. Seven plants of a capacity in excess of 100 tons of oxygen per day are known to be in operation or course of construction for various purposes, including the generation of gas for the synthesis of oil, methanol or ammonia, the supply of oxygen to steel furnaces or ore roasters and the production of chemicals. Under American conditions, the cost of 95% oxygen from such plants is estimated to vary from four to nine dollars per ton.

For generating steam in quantities up to 200,000 lbs per hour in individual boilers, the spreader stoker is established as first favourite. For firing larger boilers, coal is generally supplied in pulverised form, in spite of the difficulties and expense incurred in recovering fine ash from the flue gas and subsequently disposing of it. The increased interest of local authorities in U.S.A. in the suppression of grit and smoke emission is likely to accentuate the fly ash problem in pulverised fuel installations. Partly for this reason, the cyclone burner recently developed by Babcock and Wilcox seems likely to challenge the supremacy of the powdered coal furnace. Only one such burner is at present in commission, but a large number is on order. About 85% of the coal ash can be tapped from the cyclone in the form of quenched slag which may be used as aggregate in concrete.

At the Fuel Research Station, work has been resumed on the Vortex burner, and an interesting study has been made of the suppression of smoke from Scotch boilers. There has been some revival of interest in down jet combustion and some furnaces applying the principle are now in service in England. B.C.U.R.A. is investigating the formation and prevention of deposits on the furnace side of boiler tubes, and in conjunction with the Fuel Research Station is studying the grindability of coal. Fundamental studies are being made of ignition phenomena and of combustion and gasification reactions at I.G.T., Battelle, Carnegie and other institutes.

The Locomotive Development Committee has been developing equipment for burning pulverised coal under pressure within the confined space of a locomotive. Good progress has been made on the combustion of the coal and removal of the ash, but the nozzle method of pulverising has proved unsuccessful.

Two gas turbine locomotives, to be fired with coal, are on order to the Committee, but it seems likely that their completion as coal burning locomotives will be delayed. A coal pulverizer, to operate under pressure, is on order from Babcock and Wilcox.

An oil-fired gas turbine locomotive was commissioned in U.S.A. early this year. Another is nearing completion and a third is believed to be on order. Two are in course of construction in England, one of which may now be complete. Switzerland has had a gas turbine locomotive in commission since 1941.

A marine gas turbine power plant of American manufacture is in service with the U.S. Navy, and three other such plants are under construction. In the development of gas turbines for service on land, Swiss manufacturers lead the rest of the world. Brown Boveri has installed or is installing gas turbines to an aggregate power of 90,000 K.W., the largest being one of 27,000 K.W. capacity now in course of manufacture for a Swiss power station. There is a difference of opinion between American and Continental manufacturers regarding the temperatures for which gas turbines may be designed. It is Brown Boveri's practice to restrict working temperatures to 600°C; gas turbines are being constructed in America for operation at 700°C and nearly 800°C. The experience of the next year or two with these turbines will be very interesting.

Outside the Bureau of Mines, there is little interest in the hydrogenation of coal as a possible commerical source of oil. The Bureau has ideas for improving the efficiency of the process from about 30%, as practised in Europe, to 50% or more. However, of the various refinements in plant design by which that improvement is to be achieved, few are to be tested in the demonstration plant just completed. A new technique for the partial hydrogenation of coal under mild conditions in a fluidised bed is believed to offer possibilities of improvement upon current practice.

The version of the Fischer Tropsch synthesis process which is receiving most attention is the fluid bed modification using an iron catalyst. The first commercial plant applying the process is due for completion in February, 1950. Natural gas is to be the raw material and the designed output of the plant is 7500 barrels of liquid product per day. A second and similar project has been abandoned, at least temporarily, in an early stage of construction. A plant of the same capacity, but using coal as raw material is being designed for South Africa. It is understood that the capital is not yet available for the enterprise.

The fluid bed synthesis process has been developed mainly by Standard Oil, Hydrocarbon Research and Texaco, but other organisations are working on the process. The petrol which comprises about 75% of the product has an octane number of eighty without lead. The remaining products are diesel oil of poor quality, a furnace oil and a complex mixture of alcohols, aldehydes, ketones and fatty acids. A comparatively high temperature, viz 320°C, is applied to prevent the formation of wax which would otherwise adversely affect fluidisation.

The Bureau of Mines has avoided fluidised synthesis and has chosen, instead, to develop German variations of the process using oil as the medium for dissipating heat of reaction. In one process being investigated, the oil is circulated through a fixed catalyst bed; in the other, both catalyst and oil are circulated in the form of a slurry. These techniques, particularly the latter, permit operation at temperatures of about 250°C which are close to the optimum for high yields of liquid hydrocarbons. The oxygenated product of fluidised synthesis is replaced by wax. There is less production of gaseous hydrocarbons and the diesel oil is of better quality. The petrol produced, however, is inferior in both quantity and quality. So far as can be judged from the small scale of the work so far by the Bureau, operating costs are not likely to be done substantially different for the fluid bed and oil cooling processes.

The Bureau has just completed a small pilot unit with an output of 4½ barrels of oil per day. A 200 barrel per day plant is being erected at Louisiana, Mo. The Fuel Research Board is investigating processes using the fluidised and the suspended catalyst, but the scale of study is small.

In America there is only academic interest in the synthesis of methane. The possible scope of application in England is regarded as very limited, mainly because of the loss of heat incurred in the process. The Gas Research Board is currently working on three phases of the problem. The first, as already mentioned in connection with water gas manufacture, is the better utilisation of waste heat in that process, so that good use may be found, if necessary, for the heat of the methane synthesis reaction. The second is the reaction itself, which has been carried out continuously over a period of four months without reactivation of the nickel catalyst used. The third is the purification of the gas. The required degree of removal of sulphur compounds has been achieved by converting organic sulphur to hydrogen sulphide over a copper-chromium catalyst, followed by oxide purification and a final treatment with a nickel absorbent. A difficulty experienced - though probably a minor one - is that the copper-chromium catalyst cannot be revivified. The Board is also working on improvements to the oxide process, applying a fluid bed in one case and extracting the spent oxide with a solvent in the other. The copper-chromium catalyst just mentioned is being tested on unpurified coal gas with a view to reducing the organic sulphur contained in the gas.

At I.G.T. and the Bureau of Mines station at Morgantown, work is proceeding on the estimation of sulphur compounds in coal gas and on their elimination.

Several applications of the fluid bed technique, other than those already mentioned, are being investigated. The Bureau of Mines is adapting the process to the drying of fine coal; the

Dorr Co, and the New England Lime Co. are both drying and burning lime in fluid beds; the Dorr Co. is adapting the principle to the roasting of various ores.

The flash drying of fine coal appears to be growing in favour. In one interesting application of the technique, lignite of 36% moisture content is being dried with air at 650°F in a pulveriser.

At the Peoria station of the Bureau of Agriculture, interesting work is in progress on the utilisation of corn cobs and other farm wastes. The object of the work is to convert pentosans and hexosans separately into desirable products, under conditions designed for maximum yields in each case. Although ethyl alcohol is a major end product of the treatment, it is desired to obtain, as far as possible, other and more valuable products such as furfural, sugars, acetone etc. The utilisation of ethyl alcohol as a motor fuel is being further studied. The alcohol is not blended with petroleum spirit, but is supplied separately to the engine through a twin fuel carburettor. It is hoped that the use of alcohol in this way, only when the engine requires a high octane fuel, will enable high compression motors to operate on low grade and, therefore, cheap petrol.

A technique for the storage of low rank coals has been developed at Golden, Colo. The coal pile is built up in layers, each of which is consolidated by driving a bulldozer over it before the next layer is added,

INTRODUCTION.

This report is a summary of information collected between November, 1948 and July, 1949 in the course of visits to the United Kingdom and the United States. Most of the inspections and interviews were made in the company of Mr. G. E. Baragwanath, Senior Research Officer in the State Electricity Commission of Victoria and Mr. R. W. Urie, Research Officer in the Commonwealth Scientific and Industrial Research Organisation. The main subjects investigated were the complete gasification of coal, particularly in respect of its application to Victorian brown coal, the production of oil by synthesis and the production and refining of shale oil. Information was sought on a considerable number of other interests in the field of fuel technology as the opportunity offered. The range of subjects investigated is so wide that the report should not be presumed to be based in all cases on expert knowledge. The authorities consulted, however, were invariably experts in their respective fields, and great care has been taken to record their information accurately. That information is offered with confidence as to its authenticity.

The report is a summary of recent developments and not an exhaustive study of the subjects mentioned. Where those developments have been published, references are quoted to the literature and no more is repeated in the text than is desirable to ensure a coherent narrative. A considerable mass of detail has been excluded but is available on record for further reference.

To avoid tiresome repetition in discriminating between British and American units, the practice has been adopted of recording tons and gallons just as they were originally quoted. Except where clearly indicated otherwise, the long ton and imperial gallon are implied in all information received outside the United States. In references to projects etc. within that country, the short ton and U.S. gallon are used. In the few cases where confusion might arise, the units are specifically stated.

OIL SHALE and SHALE OIL.

Although there is no active shale oil industry in the United States and no prospect of one being established in the near future, research work is being prosecuted steadily in that country in anticipation of the day - admittedly distant - when dwindling supplies of natural petroleum will need to be supplemented by production from other sources. The Bureau of Mines is investigating the mining of shale from a 70 ft. seam in Colorado, the production of oil from that shale in the laboratory and in two commercial retorts, the refining of the oil, fundamental properties of shale and shale oil and other subjects of interest. Samples of oil for refining studies have been made widely available by the Bureau, but little or no information resulting from that work has been published. It is generally accepted that the best method of refining consists in delayed coking of the crude oil and partial hydrogenation of the distillate, followed by further treatment by processes in common use in the oil industry. The Union Oil Company of California appears to have done most work and published most information on the hydrogenation of shale oil and similar stocks. The same Company is developing a process of considerable promise for the carbonisation of oil shale. The Standard Oil Development Company has applied to the same problem its fluid bed technique widely used in catalytic cracking. The American Cyanamid Company has studied the concentration of the low grade shales of Colorado by froth flotation and by heavy media separation.

UNITED STATES BUREAU OF MINES:

The Bureau of Mines publishes its work on oil shale and shale oil in great detail at yearly intervals. ++ For this reason, little purpose would be served by recording at length in this report the information obtained at the Laramie and Rifle experiment stations.

++ See U.S. Bureau of Mines, R.I. 4457.

SYNTHETIC LIQUID FUELS.

1948 Annual Report of the Secretary of the Interior,

Part 11. Oil from Oil Shale.

Mining Operations at Rifle comprise ; -

(1) The winning of selected shale in small quantities, and without regard to cost as a controlling factor, for the N.T.U. and other retorts. At present the shale is coming from a 10 foot section of the seam above the "mahogany marker" and has an assay value of about 30 gallons per ton.

(2) The winning, in large quantities, of shale from a 23 ft section of the seam, that section being one of three benches in which the best shale in the deposit would be mined in commercial operations. The shale won in these operations is dumped over the cliff, the purposes of the work being merely to obtain experience in suitable techniques and to determine costs of production.

The outstanding features of the shale mine at Rifle are the great height of the seam and the wide working places permitted by the mechanical strength of the shale. The bords from which the shale is being won are 50 and 60 feet wide. The experimental room for observation of roof sag is 100 feet long by 80 feet wide. It has been located deliberately at a point where there is a vertical crack in the roof from side to side of the room. The sag in the centre after 18 months' observatio is about 0.4 inch. So long as the sag increases slowly and uniformly the roof will be regarded as safe because the movement will be indicative of elastic strain. Any sudden or great increase in the rate of sag will be interpreted as a warning of impending failure. The record of rock noises, detected by geophones buried in the roof at various points, will be interpreted in a similar manner.

In mining operations, the shale is drilled to a depth of about 9'6" by two percussive air drills mounted on a jumbo. In a face 60ft by 23 ft, 84 holes nearly 3" in diameter are drilled in one shift, and are then charged with explosives and fired on the next shift. After the round of shots, loose pieces of shale are prised by hand from the roof and walls. A diesel bulldozer pushes the broken shale up to an electric shovel, which loads the two 15 ton diesel trucks used to transport the shale out of the mine. Ten men are employed in the operations briefly mentioned above and they win 800 tons per shift at a cost of 50 cents per ton exclusive of capital and surface charges. In the Bureau, hopes are entertained of effecting further, though small, reductions in cost, and of demonstrating that shale can be delivered in large quantities to a carbonising plant at an inclusive cost of 60 cents per ton. Some authorities outside the Bureau are inclined to emphasise administration and other charges not incurred in the present method of working. Probable all-inclusive costs of $1. and $1.15 per ton have been quoted by such authorities.

Carbonising Operations at Rifle comprise :-

(1) The production of substantial quantities of crude shale oil from two N.T.U. retorts chosen, not because of any preference for the process, but because it was known that they could be installed quickly and operated with little trouble. The oil was needed with a minimum of delay for examination and refining studies within and without the Bureau.

(2) The development of the "Gas Flow" retort chosen as one likely, in the Bureau's opinion, to prove superior to most, if not all, retorts now in commercial operation.

The N.T.U. retorts have fulfilled their main function in making available supplies of oil needed for other work. In other respects their performance has been disappointing. The oil yield has varied between 60% and 85% of the content by Fischer assay, losses in the form of oil fog being significant. Difficulties have been experienced with emulsions. A systematic investigation is being made into the effect of varying the air: recycle gas ratio.

The Gas Flow retort has been developed from the Lantz or G.P. process and differs from the latter mainly in three ways viz :-

(1) The circulating gas is heated in one of two pebble stove regenerators, which are alternately heated with natural gas and cooled by the circulating retort gas,

(2) The gases are cooled at each circulation to remove condensible vapours.

(3) There is no cooling zone in the retort.

The limited amount of work so far done on the retort has given results which may be regarded as generally encouraging. Difficulties have been experienced calling for modifications in design. The oil yield is substantially the same as given by the Fischer assay. The rate of carbonisation is fairly high, the throughput of the pilot plant, with a carbonising zone 10 feet high and 18 inches by 12 inches in cross section, being 1,500 lbs of shale per hour. Much more experience and information is needed, however, before the retort may be regarded as ready for commercial adoption.

<u>Solvent extraction</u> of oil shale has been pursued at Laramie to a point at which the process has been found technically attractive but commercially impracticable. The yield of oil is substantially greater than that obtained in the Fischer assay, and the quality is good for a shale oil. However, the rates of decomposition of kerogen and oil product respectively are such that, for Colorado shale as least, the process would not be self supporting in respect of solvent. A more important discouraging factor is the cost of filtering ash from the product — a cost regarded at the moment as likely to prove prohibitive.

The shale oil obtained by solvent extraction has been hydrogenated in a small, oscillating autoclave. It has been found that the nitrogen compounds present in the oil are removed in part only by this simple hydrogenation treatment. Because the solvent extraction product is similar in physical and chemical characteristics to retorted shale oil, it is inferred that substantially complete hydrogenation of crude oil is needed for the effective elimination of nitrogen and sulphur compounds. (Elsewhere in this report it is recorded that much more favourable results have been obtained by the Union Oil Company when hydrogenating, substantially in the vapour phase, coker distillates of high nitrogen and sulphur contents.)

Pyrolysis and assay studies at Laramie include the determination of the heat of decomposition of Colorade shale and a comparison of assay yields by the Fischer and Gray King methods respectively. The heat required to carbonise shale is measured in an adiabatic calorimeter, which has two jackets insulated from one another. Each is in contact with a thermo-couple, and a sensitive relay causes the outer jacket to be heated when its temperature falls below that of the inner jacket. It has been found that the total heat needed for carbonisation differs from sensible heat requirements, as determined from extrapolated specific heats, by an amount which is insignificantly small. It is concluded, therefore, that the heat of reaction is negligible. This deduction is contrary to previously published results of work on similar shales, but the sensitivity and accuracy of the method and apparatus in use at Laramie leave little room for doubt regarding the results now obtained. The work is to be extended to other shales in the near future.

The comparison between Gray King and Fischer assays is being made on Australian shale. The number of Fischer analyses so far made is too small to justify final conclusions, particularly with regard to reproducibility, etc., but it is tentatively indicated that the percentage yield by weight is slightly greater in the Fischer assay and the yield by volume slightly lower. It could be that the inclusion of naphtha in the Gray King yield, as practised at Glen Davis where these tests were made, is responsible for the lower specific gravity and greater volume of the Gray King oil.

The Fischer assay has proved very satisfactory at Laramie. The method appears simpler and more expeditious than the Gray.King. Three operators are able to complete eight assays per day, including the preparation of the samples. One weakness of the Fischer apparatus is that the seating surfaces of the retort and retort cap need to be turned down at intervals to keep the apparatus gas tight. Eventually the cap must be discarded and an oversize casting substituted for it. (At Golden, Colorado, where the Fischer assay is used by the Bureau of Mines in the analysis of coal, the taper plug type of seat has been abandoned in favour of a horizontal packed joint, using thin asbestos millboard and graphite as packing. The cap is held tightly in position by two clamps passing round and over the retort. The seating surface is recessed into the top of the retort so that the volume of the vapour space may be just as it is in the standard apparatus. This construction is said to eliminate the need for periodic refacing of the seating surfaces.)

<u>Laboratory and works instruments</u>, seen at Laramie and Rifle included a mass spectrometer for the analysis of gases, apparatus for the determination of oxygen in organic compounds by the Unterzaucher method and an instrument for registering the oxygen content of a gas stream. As mentioned later in this report, the mass spectrometer is now almost standard equipment in research laboratories working on fuel problems. The Unterzaucher analysis ++ has been developed as an alternative to the much distrusted "oxygen by difference" determination. The sample is completely decomposed in an inert atmosphere at 1100°C or 1200°C, and the gaseous products are passed through heated active carbon. Oxygen is then determined in the form of carbon monoxide using iodine pentoxide as reagent. At Rifle, the gas leaving the N.T.U. retorts is analysed continuously for oxygen content by an instrument which functions on the variation of magnetic susceptibility with oxygen concentration. The apparatus seen was the Pauling Gas Analyser made by A.E. Beckman of Pasadena, Cal. It appears rather delicate for ordinary works use, the suspension of the torsion balance being by quartz fibre. (An instrument, probably similar in principle, was seen recording the oxygen content of regenerator waste gases at Aurora refinery, Detroit. The manufacturer was the Hays Instrument Company of Michigan City, Ind. The instrument was giving good service but nothing was known at the refinery of its construction and maintenance. The instrument was still regarded by the makers as experimental; no description and instructions were issued with it, and all servicing is done by a representative of the Hays Company.)

<u>The Shale Oil refinery</u> at Rifle is a small Dubbs unit rated at 50 to 200 barrels per day according to the type of operation. The heater is so manifolded and the plant so designed

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Anal, Chem. Vol. 19 No.5, P.347.
U.S.B. of S. Journal of Research. Vol.40, No.6, P.443.

that the charge may be vis-broken, reformed, cracked to residuum or cracked to coke. Treatment of the raw petrol is to be in vertical towers with gravity settlement and re-circulation of the bottoms in each tower by centrifugal pump. The petrol will be washed in turn with caustic, dilute sulphuric acid and 98% acid in three counter-current stages. The concentrated acid treatment is to be at 35°F maintained by refrigeration. The refinery was completed early in 1949, but because of a shortage of steam had not been commissioned at the time of the latest advice in June, 1949.

Bench scale apparatus has been installed at Laramie for a close study of the catalytic cracking of shale oil. No results are yet available, however.

STANDARD OIL DEVELOPMENT COMPANY.

A few years ago at Bayway N.J., Standard Oil carried out some preliminary carbonisation tests on Colorado and Australian oil shale, applying on a relatively small scale the fluid bed technique which the Company had previously developed for the catalytic cracking of petroleum. Quite recently, at the suggestion of the Bureau of Mines, by whom the shale was provided, the Company resumed work on the process, using for the purpose the 100 barrel per day pilot catalytic cracker at Baton Rouge, La. The tests were only partially satisfactory, because the plant had not been designed specifically for the treatment of shale and because it had not been thoroughly overhauled after a period of idleness before commencing the work on oil shale. A flow diagram and a few major dimensions of the plant, as adapted to the carbonisation of oil shale, are given in Figure 1.

The mechanical difficulties experienced included -

(1) The blocking of circulating lines due to accumulations of scale which peeled of the plant.

(2) Erosion of gate valves which had been designed to pass cracking catalyst in the desired quantities in the full open position. Owing to the changed differential pressures when circulating spent shale, the valves were required to do some throttling and suffered undue wear in consequence.

(3). Because the feed was a mixture of particles some of which yielded oil and broke down to a fine powder on treatment, while others containing no oil suffered little disintegration, the size consist of the material in course of treatment was unfavourable to good fluidisation. For some time, therefore, difficulty was experienced in maintaining steady circulation and, on occasion, cyclone stand-pipes were suspected of being either unsealed or choked.

(4) Because of the very fine dust formed by the disintegration of oil yielding particles, and because the cyclones were designed to trap coarser particles of cracking catalyst, much dust was carried over into the oil recovery system. The second-hand Oliver filter installed to separate the sludge of oil and ash was found to be in poor mechanical condition and functioned most unsatisfactorily.

Difficulties (1) and (3) were solved by cleaning the plant thoroughly and by admitting plenty of fluidising gas into the circulating lines. It was found possible, eventually, to operate the plant continuously for long periods on automatic instrument control. The dust problem was never solved in these tests, however, and most of the product was collected contaminated with ash and fine shale.

To keep the product line free from dust deposits, it was found necessary to flush it with oil. Because the shale oil could not be filtered and re-circulated as rapidly as it was collected, extraneous petroleum oil had to be used for the purpose. Therefore, the product oil was contaminated, not only with dust, but also with oil not derived from shale. In these circumstances it was not possible to determine by direct measurement the volume of oil made from the shale. Instead, the yield was calculated from a carbon balance, using the carbon contents of the oil shale, the shale oil and the gases made in the tests, the volume of gas generated and the weight of shale consumed as determined by volumetric measurement in hoppers. In this manner, it was computed that the yield realised was roughly equal to the Fischer assay value of the shale, but as may be imagined from the method of calculation, no great order of accuracy attaches to the final result.

Again because of the contamination of the shale oil with petroleum used for flushing the product lines, it is uncertain whether information of great value will be obtained by inspection and analysis of the oil. So far, no information of this nature has been made available.

Shale oil was charged to the plant at the rate of rather more than nine tons per day. Ash, of which 60% was finer than ten microns, was circulated at a rate slightly in excess of ten tons per hour. If the capacity of a retort of this type be assumed to be proportional to the cross sectional area of the burner, the performance of the pilot plant would suggest a throughput of 2,000 tons of shale per day for a plant with the physical dimensions of a large catalytic cracker. If the rate of circulation of solids be regarded as the criterion of capacity, a large shale retort would be indicated as having a throughput of 5,000 tons per day. A plant designed specifically to carbonise shale would probably improve upon that throughput, and the Company itself considers that a capacity of even 10,000 tons per day is realisable.

The pilot unit at Baton Rouge was operated on shale from December, 1948 to May, 1949, with many interruptions for adjustment and modifications. The immediate programme of work is regarded as complete and the plant is to be returned to service as a catalytic cracker. If more work is undertaken on fluidised shale carbonisation - and it seems obvious that further development is needed before the process can be applied commercially - it is likely that studies will be resumed on a smaller scale in a plant specially designed for the purpose.

UNION OIL COMPANY of CALIFORNIA.

The Union Oil Company has been developing a shale [++] retort based upon the N.T.U. principle, but endeavouring to eliminate the weaknesses of that process, viz:-intermittent operation, dusty conditions, low throughput, and losses of oil due to entrainment and other causes. The Union retort is fed with shale continuously from the bottom and the ash and clinker spill over the top edge of the plant. In other respects the process has much in common with

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Development of Union Oil Retorting method of Recovery of Shale Oil, Petroleum Processing, December, 1948.

the N.T.U.

The Company's early work was carried out in a small retort, apparently no longer in existence, with a capacity of two tons of Colorado shale per day. During 1949, work proceeded intermittently on a five foot diameter retort which was found to have a capacity of fifty tons per day. About the middle of the year, modifications were made to certain mechanical devices, viz. the rabbles, the ultrasonic mist precipitator and the sludge scraper. Further tests were planned for the retort and it was intended, later in the year, to construct a unit capable of operating under pressure and, therefore, possibly suitable for the Lurgi method of gasification. The Company estimates that shale retorts could be constructed to its plans with a capacity of 4,000 tons per day per unit.

It may be doubted whether sufficient operating experience has yet been gained to establish the soundness of certain mechanical features of the plant, for instance, the unlined retort shell. As retorts of increasing size are designed and constructed, it will become necessary to ensure that the shale, charged through the bottom cone, will be spread sufficiently uniformly over the cross section of the retort. The ultrasonic mist precipitator appears to have been very successful and it is probably to this device that the high oil yield claimed, viz. 102% of the Fischer assay yield, must be, in a large measure, attributed. It is stated that 25% of the oil leaves the retort in the form of a mist, much of it in particles only one micron in diameter. The ultrasonic vibrations coagulate those very fine particles into drops ten microns in diameter, which are precipitated readily by the rotaclone incorporated in the plant. In this way, 99.8% of the mist had been removed from the gas stream.

The Union Oil Company's study of catalytic hydrogenation has been directed primarily towards the more effective utilisation of Californian crude oils of high nitrogen and sulphur contents. Reference is made to this subject on page 34 of this report, where a brief description of the plant and process is given. Crude oil from Colorado shale has been processed in the same way and with equally satisfactory results in the pilot plant. The sequence of operations is as given in Figure 2.

The purpose of the coking operation is to prepare a charge for the hydrogenation unit which may be treated substantially in the vapour phase. In typical pilot plant tests, the nitrogen content of 1.93% in the crude was reduced to 1.44% in the coker distillate and to 0.02% in the hydrogenated product. Sulphur was reduced from between 0.8 and 0.9% in the crude and the distillate to less than 0.05% in the finished product. The overall yield from the crude was about 85%. The hydrogenated oil was analysed for the production of jet fuel (62%) and diesel oil (38%). This selection of products was made, presumably, because it is considered that no early exploitation of shale can be contemplated except for reasons of defence.

The petrol which might be obtained from the hydrogenated product would be of low octane rating and would require reforming by some process to meet accepted specifications of quality. The cetane number of hydrogenated diesel oil was quoted in one instance as 49, which would be poor, and in another as 59 which would indicate a satisfactory fuel.

AMERICAN CYANAMID COMPANY:

American Cyanamid has undertaken work on Colorado shale in an attempt to concentrate the kerogen by its heavy media and froth flotation processes. For the former process, shale assaying 28 gallons per ton was obtained from the mahogany ledge at Rifle, crushed to 1" and screened over 1/10". This sizing was made to suit the 20" cone unit with a 2" air lift, which was used in the investigation.

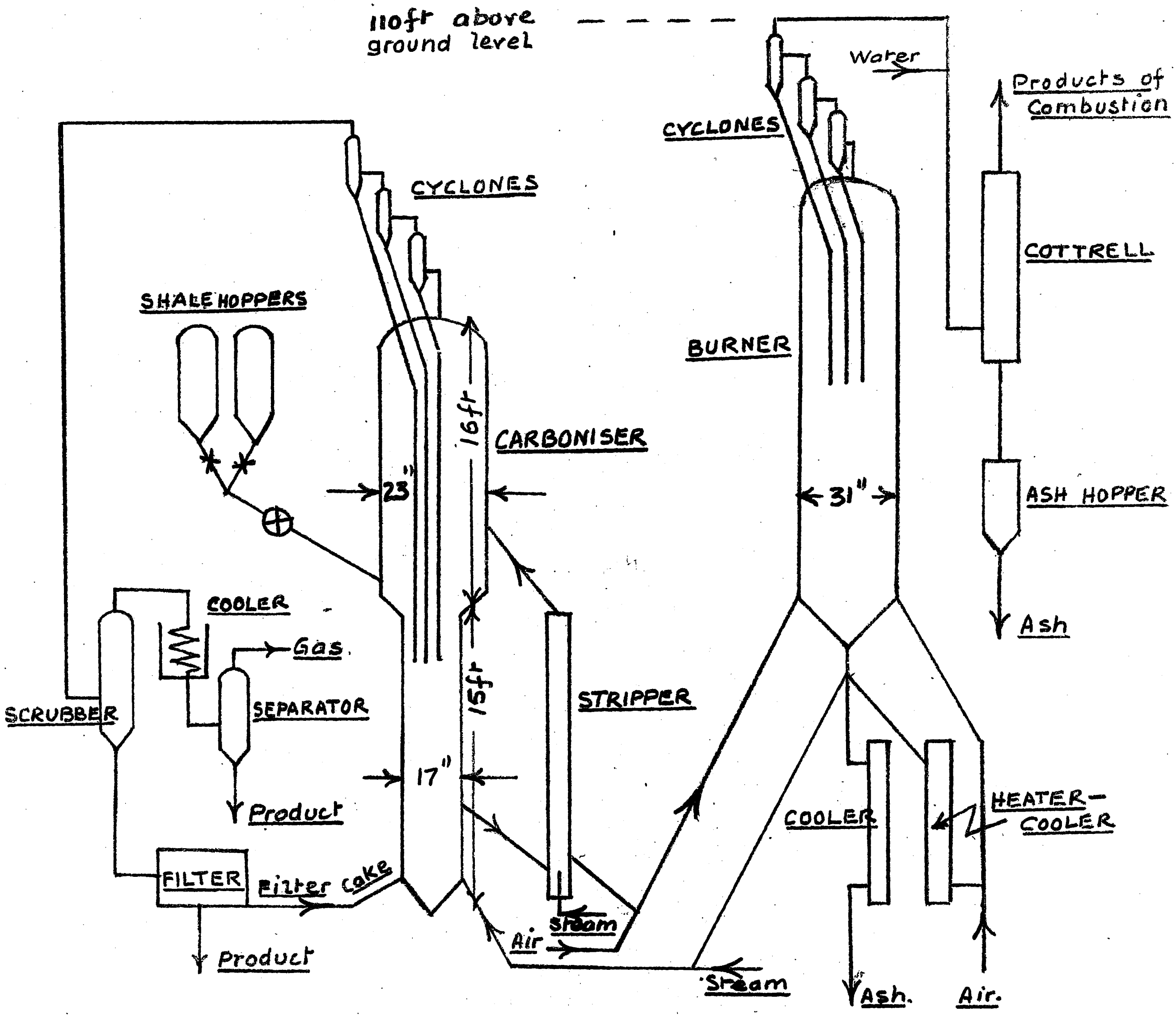

FIG. 1. FLUIDISED SHALE RETORT.

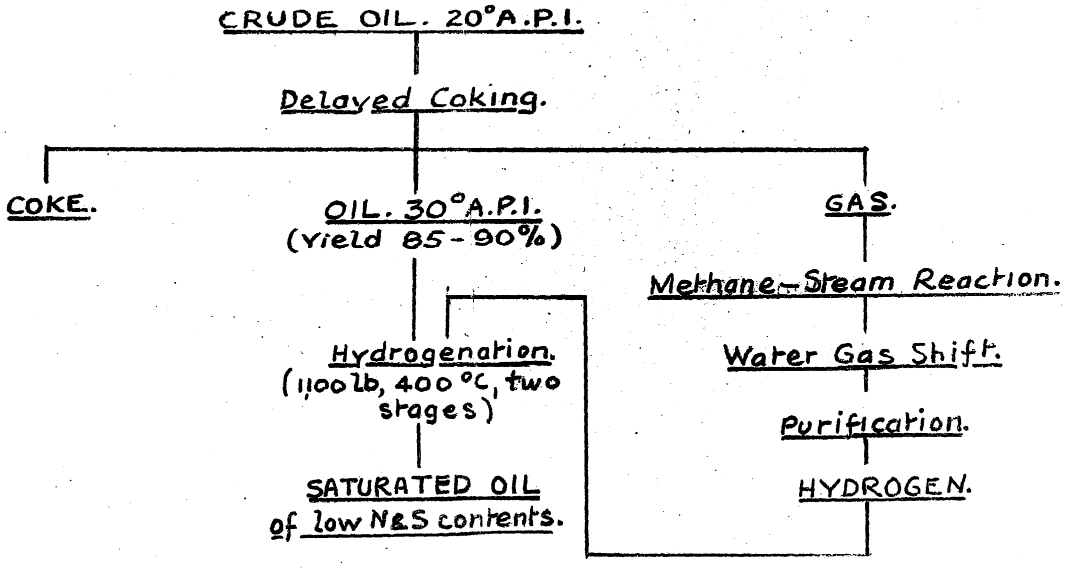

FIG. 2. REFINING SCHEME FOR OIL OF HIGH N. AND S. CONTENTS.

In computing the following results, the fines - whose assay differed little from that of the original material - are assumed to have been mixed with the concentrate.

Density of medium (water = 1)	Assay of Concentrate (gallons per ton)	Oil recovery. (per cent)
2.3	32.5	92.2
2.2	35.4	84.2
2.1	40	70
2.0	44.4	53

Froth flotation gave results of the same order. By crushing to 65 mesh, screening into six sized fractions, floating the fractions individually and lumping the concentrates, an oil recovery of 63% was realised for an appreciation in assay content from 28 to 39 gallons per ton. The richest concentrate, obtained from the +65 mesh fraction, assayed 57 gallons per ton.

Examination under the microscope disclosed that the concentrate contained at least two different "organic" constituents each of which would necessarily include much inert matter. The +65 concentrate contained 75% of dark and 25% of light "organic" matter. These observations indicated that in order to effect a close separation of organic constituents, the shale would have to be crushed to an exceedingly, and probably impracticably, small particle size. The whole investigation served to establish that no useful concentration of kerogen, as a treatment preliminary to carbonisation, could be effected by the most likely processes commercially available.

MISCELLANEOUS STUDIES.

Shell Development has confined its research work on oil shale to a study of Colorado shale itself, the oil obtained by carbonisation, and chloroform extracts obtained in the cold and after incipient pyrolysis. It has been found that as much as 25% of the organic matter in the shale can be recovered by repeated extraction with chloroform. The examination of the shale has disclosed that there are three different types of organic constituents in oil shale. The first is a bitumen soluble in cold chloroform, to the extent of up to 25% of the kerogen, the second is of humic nature and insoluble, the third is an insoluble bitumen. By heating at 350°C, substantially all the organic matter is rendered soluble in chloroform. The ultimate analysis of products obtained by extraction alone, pyrolysis and extraction, and by extraction alone are as follows -

		C,	H.	N.	S.	Cl.	O.(1)	H:C.
Extract from shale,	%	83.7	11.9	1.0	1.1	2.4(2)		1.71
Extract from pyrolysed shale,	%	82.4	10.4	2.3	0.8	0.9(2)	1.6	1.51
Oil by pyrolysis alone,	%	83.7	12.2	0.8	1.6	0.2(3)	1.5	1.75

(1) By difference, the result being considered accurate,

(2) Attributed to the solvent,

(3) Considered genuine and accurate.

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<u>California Research Corporation</u>, the research organisation of Standard of California, has studied the refining of shale oil under emergency conditions, in which the crude product would be supplied, regardless of cost, by some other organisation, and the Company would be required to make the best possible joint use of the petroleum and shale oils available. Because shale oil is so difficult to refine and its products of inferior quality, in these circumstances the Company would give that stock a minimum of treatment, commencing with vis-breaking to fuel oil specifications. The heavy product would give a bad odour to the black fuel oil into which it would be blended for disposal. In an emergency, however, that objection would probably be disregarded. If for any reason it should be necessary to reduce the odour of the black oil, the vis-broken stock would be fractionated and the gas oil treated with concentrated sulphuric acid for the removal of tar bases. Only 45% of the nitrogen compounds in Colorado shale oil are soluble in dilute acid. Concentrated acid removes 80% of the bases, the remaining 20% having a comparatively mild odour. The treated gas oil and the untreated residue would be blended into the black oil. The heavy bases in the shale oil residue are not strongly odorous.

To restore the balance between the distillate and residual fuel oils, catalytically cracked light oil, which is at present sold in admixture with black oil, would be treated by the Edeleanu process for the removal of those unstable components which constitute an objection to the sale of the oil as furnace oil. The extract, amounting to about 40% of the stock treated, would be blended into the black oil as before, but the raffinate would be sold as diesel oil or household fuel oil. Distillates at present used for those purposes would be catalytically cracked for additional petrol. This procedure would be preferable to the production of a distillate fuel from shale oil by fractionation and acid treatment. The Edeleanu plant needed in the one case would cost only a quarter as much as the acid plant required for the treatment of distillate shale oil in the other. The shale petrol resulting from viscosity breaking would be treated with soda and acid and mixed with the petrol produced from well oil. The deterioration in quality would be accepted as an inevitable consequence of the emergency. In any case, the depreciation of octane rating and the increase in sulphur content would not be great. For instance, 50,000 barrels of shale oil per day yielding 25% of petrol by viscosity breaking would not greatly affect the quality of the Company's daily output of 90,000 barrels of motor spirit.

OBSERVATIONS ON THE INDUSTRY ABROAD.

Inthe United States there is no commercial production of shale oil and no prospect of any early development in this direction. Most of the large oil companies, including Texas, Union, Standard of California, Pure Oil and Sinclair, have extensive reserves of oil shale property, but regard them only as insurances against the day when cheaper sources of oil shall have been exploited to the point at which depleted reserves are no longer adequate. Current work on oil shale and shale oil in U.S.A. is confined to research and the development of processes and techniques.

In Scotland, production continues at the rate of 5,000 tons [+] of shale per day, yielding 25 gallons of crude oil per ton.

[+] The ton quoted for mining operations by Scottish Oils Limited is the "miners ton" of 25 cwt.

The oil is not cracked and the principal product is diesel oil. Other important products are petrol, wax and sulphate of ammonia. The average wage in the industry is £6. 9. 0d for a week of 44 hours. The cost of mining is 15/-d per ton and of carbonising and refining also 15/-d per ton approximately.

Some shale is being won from an open cut and about 400 tonsper day are being reclaimed from a dump of material discarded fifty years ago as too lean to be worth carbonising. Conditions in the mines do not favour extensive mechanisation, because there are few working places free from faults and at the same time not dipping steeply. Electric drilling of shot holes is usual, but all loading into skips or conveyors is by hand. Miners are paid 37/-d per shift and the output per man at the face is 5½ tons.

Carbonising operations are concentrated at four works. It has been possible to close down some older works because of :-

(1) the increase from four to ten tons in the throughput of existing retorts each day, when air is injected at the base, a practice adopted about twelve years ago.

(2) A new bench of retorts was erected at Westwood about seven years ago. The Westwood retorts, 104 in number, are of the traditional Scottish type, but with Woodall Duckham extractor gear and belt conveyors for handling shale and residue. They carbonise 1,200 tons of shale per day and the whole works, including the sulphate plant, is operated by 110 men. The works is neatly laid out, clean and comparatively free from dust.

The yield of sulphate of ammonia has been reduced somewhat to about 30 lbs per ton of shale by the practice of injecting air into the bottom of the retort. The product is of superfine quality.

Two innovations have been made comparatively recently at the Pumpherston refinery. From the acid washings from the diesel oil there is obtained, after neutralisation with caustic soda, a mixture of ester salts, mainly aliphatic, which are sold in solution form as a soap substitute under the name of Iranopol. Detergents prepared from oil and oil products are mentioned later in this report. The second development is the discarding of wax sweating houses in favour of a more efficient process, in which the wax is heated to a gradually increasing temperature in a vessel fitted with a large number of tubes through which hot water is passed.

A good market persists for the bricks made at Pumpherston from spent shale and lime. The present output is 40,000 per day. Girls are used for all operations not involving attendance on machinery.

Brazilian shale is a "paper" shale, soft and wet when mined, but drying to a hard, brittle, finely laminated material when exposed to the air. The yield of oil is 20-30 gallons per ton and the product is paraffinic. The seam is some 200 feet thick below a cover of 40 or 50 feet. Its extent is not known. The deposit is conveniently situated, about thirty miles from San Paulo astride the railway and road leading to that city. The retorts at present in use are Henderson units, in a bad state of disrepair, installed at a time when the Henderson retort was the latest development in the Scottish shale oil industry. Refining operations are almost primitive. Foster wheeler is planning a new carbonising and refining plant for a capacity of 6,000 barrels of crude oil per day. It is believed that there is sufficient shale in Brazil to justify several plants of that size. For this project, the Union Oil Coy's. retort is receiving attention, but doubt is entertained regarding the ability of so brittle a shale to resist the crushing load which is applied by the underfeed charging device, A retort of the Lurgi Spulgas type may eventually be recommended. The scheme is at present delayed because of difficulty experienced in finding dollars for purchasing machinery in U.S.A. The project is being considered by Government and private interests in co-operation.

In Sweden, where the shale industry is reported to be flourishing, the main technical interest is in the Ljungstrom method of carbonisation in situ, by drilling the deposit at seven foot intervals and inserting electric heaters. It is said that the current has to be turned on for three months, before the first oil appears. Yields are rather less than 90% of those obtained from retorts, but the product is thinner and of higher value.

The shale is highly sulphurous and the odour of hydrogen sulphide in the vicinity of the plant is most unpleasant.

Reflections on Australian Practice. (CONFIDENTIAL)

The main difficulty being experienced in the Australian shale oil industry is the restriction of output arising out of limitation practices and demarkation of activities. On such matters it is not possible to offer comment beyond observing that estimated costs of 16 to 19 cents per gallon for petrol produced from Colorado shale and delivered in California are based upon tasks greatly in excess of those being fulfilled at Glen Davis. The Scottish oil industry which is owned and operated by private enterprise could not survive if the effort per man employed were no higher than it is in the Australian industry.

In the production of petrol from shale, there appears no outstanding improvement in technique ready for adoption at Glen Davis. At the Westwood works, certainly, some practices are obviously superior to our own. Shale and spent ash are transported exclusively on rubber belt conveyors with every satisfaction; the retorts and conveyors give good service without protection from the weather; the Woodall Duckham water-sealed extractor is an improvement upon the screw extractor adopted at Glen Davis. These theoretically possible improvements, however, are matters of detail rather than of principle. They are, indeed, of academic interest only, for the alternative arrangements installed at Glen Davis either cannot, or need not, now be changed. The possibility of the Westwood retort itself satisfactorily carbonising 55 gallon shale is, also, of academic interest only. The Glen Davis retorts were designed to treat 100 gallon shale and they may be called upon some day to do so. The Westwood retort definitely could not handle that rich shale economically. No new retort was seen developed to a stage at which its commercial adoption in Australia could be soundly recommended. The Gas Flow, Standard Oil, Union Oil and Renco retorts all give promise of producing from the same shale considerably more oil than does the Fell retort at Glen Davis. They must all be regarded, however, as in an early stage of development.

For the refining of crude shale oil in the quantities available locally, there is no alternative to the processes already adopted at Glen Davis. Catalytic cracking must be disregarded on two counts; firstly the nitrogen content of the stock, secondly the small volume of crude oil available. Viscosity breaking and hydrogenation, followed by thermal cracking, could hardly be entertained for operations on so small a scale, because of the high capital charges which would be incurred. The Scottish method of refining by straight distillation is not applicable at Glen Davis because of the small scale of operations, and because the assistance given to the industry by remission of excise duty calls for a maximum production of petrol.

A technical problem which may arise in the refining of Australian shale oil in a few years' time is the production of petrol of higher octane rating to meet competition from imported motor spirit of improved quality. Octane numbers appear to be on the rise in the United States after a long period of stability In England, ratings are being kept low as a matter of policy, so as to reduce the value, and perhaps even the volume, of imports. The early installation of four catalytic crackers of moderate capacity in Great Britian and one large unit at Abadan may, however, cause that policy to be modified. If the octane number of petrol in England and Australia should rise to 75, as it may well do in a few years, difficulty would be experienced at Glen Davis in meeting the challenge. No improved crude oil treatment process is available for application under local conditions. Some increase in octane number could be effected by pre-fractionating the crude so as to make more naphtha available for reforming. It would probably be necessary, however, to consider the further elimination of sulphur from the raw petrol so as to improve its response to lead. Current authoritative opinion on catalytic de-sulpharisation in U.S.A. is to the effect that the process is not applicable to cracked petrol. Refrigerated acid treatment seems more likely to provide an answer to the problem.

THE PETROLEUM INDUSTRY.

In the course of a number of visits to oil refineries and interviews with personnel in the industry, generally on matters only incidentally connected with refinery practices, interesting information was gained on various phases of the industry.

The Supply Position.

In 1948 and for some years previously, oil production in U.S.A. was so far short of demand that until recently the country was importing, on balance, about half a million barrels per day. The somewhat surprising increase in consumption since the war, to its present level of over six million barrels daily, is the result of the increased use of motor vehicles, a wide substitution of oil-burning for coal-burning equipment in domestic furnaces and the extended adoption of diesel electric locomotives. Production was unable to keep pace with consumption because, during and immediately after the war, the steel needed for drilling equipment etc. was not made available in adequate quantities. These were the conditions which, a few years ago, inspired keen interest and considerable activity in the production of oil from other sources viz. shale, coal and natural gas. These conditions, too, were responsible for the action of the Secretary of the Interior, Mr. Krug, in advocating a nine billion dollar scheme for the production of synthetic fuels by the Government on a scale of two million barrels per day. Mr. Krug was persuaded to recommend this scheme by the defence implications of an oil shortage within the frontiers of the United States, a subject upon which official and private opinion were at variance. The oil industry regarded without undue concern a situation in which large volumes of oil would have to be imported from Venezuela, and possibly the Near East, for the defence of U.S.A. The Government was perturbed by that same prospect. It entertained no hope of maintaining shipping channels across the Atlantic and considered that it would be difficult to protect even the route to Venezuela.

Those conditions have now disappeared and are unlikely to recur for many years. The intensive drilling campaign since the war has so increased potential production in the United States that it has become possible to shut in about 500,000 barrels of daily production in Venezuela, Texas and elsewhere. The defence position in particular has been fundamentally changed by the discovery of vast oil reserves in Alberta. Insufficient work has yet been done to indicate with any accuracy the extent of the field. It is considered possible, however, that the Alberta field and its likely extensions along the Rocky Mountains may contain as much oil as has been discovered in Texas, or even in the whole United States. With such resources just outside the borders of the country, within the control of a friendly power and not subject to the hazards of ocean transport, considerations of defence can no longer be regarded as outweighing purely economic issues.

Now that there is a reasonable assurance of adequate oil supplies being forthcoming from natural sources for many years, at a price with which substitutes from coal and shale cannot compete, interest in these substitutes has faded very considerably. Such interest as remains is devoted to processes rather than production.

It is generally considered that when the time does come for oil to be produced in large quantities from alternative raw materials, there will be a broad division of the United States into three areas according to the resources upon which they will draw. The west coast will exploit tar sands and oil shale. From the Mississippi to the Rocky Mountains low rank coals will be converted to oil by synthesis. The eastern states will rely upon coking coals for oil production, again by synthesis. This division of interests is responsible for the selection of different subjects for study in connection with fuel research in those respective regions.

Few authorities in America appear to have confidence in the ability of the hydrogenation process to survive in competition with other processes.

It is now considered unlikely that natural gas will contribute largely to future production of synthetic oil. Reasons for this opinion vary widely. On the one hand there is the rather academic argument that natural gas is a fuel already in a highly convenient form, and that it is illogical to destroy more than half of it to effect a slight improvement in the form value of the remainder. Practical critics point out that even the smallest, commercial synthesis plant calls for large quantities of cooling water and enormous volumes of gas, the latter being of the order of seventy million cubic feet per day for twenty or thirty years. It is commented that there are few locations, even in the United States, where both those supplies are available in the quantities required. Finally, the price of natural gas has risen to a point at which the margin of profit to be earned by synthesising oil is rather small.

The improvement in the supply of petroleum has not brought with it the change in prices which might normally be expected from plentiful production. The new oil now being brought in has been obtained at costs never before experienced in the industry. Holes have to be drilled deeper and the percentage of failures is greater. The deepest well is now beyond 20,000 feet, and the average depth of a new producing well exceeds 8,000 feet. According to whether holes are sunk on a well established property or in a speculative area, a productive bore may cost $80,000 to $500,000. Shell recently lost by fire a well on which $500,000 had been spent. It is considered that the charge against each barrel of new production which must now be allocated to the repayment of development expenses is in excess of one dollar. The high cost of development means that drilling intensity will in future be very sensitive to fluctuations in the market price of oil, and will in effect, keep that price reasonably stable. The present price of good average crude is $2.65 per barrel. For the same reasons it is considered that petrol prices are likely to remain fairly stable. The cost of petrol production from new crude at the refinery has been quoted by Dr. Murphree of Standard Oil Development at 14 cents per gallon.

While the industry was in the happy position of not being required to seek markets for petrol, refiners were content with prevailing specifications and did not respond to an invitation by the automobile industry to produce petrol of higher octane rating. A departure from this attitude now seems likely.

Ethyl Corporation has recently announced a rise from 80 to 84 in the octane number of petrol to which the name "Ethyl" may be attached. Cadillac has in production a car engine designed to make full use of a 91 octane petrol; Buick, also, is understood to be prepared to produce a super-compression engine.

Appreciation in octane rating is being resisted in England on economic grounds, in order to restrict imports. It has been calculated that if car engines and fuel are accurately adapted to one another, the maximum mileage from a given volume of crude oil is obtained by refining to an octane number of 70. That is to say, the additional loss incurred in producing petrol of higher quality is greater than the resulting gain in car mileage per gallon. It is not clear whether this conclusion is based upon the refining technique at present in use in England, and whether it will need modification as a result of the installation of four new catalytic cracking units on order for British refineries.

CATALYTIC CRACKING:

Cracking by the fluid catalytic process continues to gain ground at the expense of thermal processes and other methods of catalytic cracking. New units of all sizes are on order, but it would seem that the physical dimensions of large plants are approaching the limit at which little or nothing is to be gained by further enlargement. Reactors and regenerators 40 feet and more in diameter are being fabricated in the field because of the impossibility of lifting and transporting them in one piece. At the other end of the size range, there is no indication of it being found economical to extend the lower limit of unit capacity. The smallest plant yet designed by Universal Oil Products Co., which specialises in small installations, is one of 1,500 barrels daily capacity now in course of construction.

The Thermofor catalytic cracking process has gone out of favour, originally because of the mechanical weakness of the bucket elevator installed to handle hot catalyst. An air lift has now been substituted for the elevator with satisfactory results, but the Pneumofor as it is now called is still at a disadvantage in respect of the catalyst: oil ratio which may be maintained in the reactor.

In consequence, these units cannot crack a stock as heavy as those which may be charged to a fluid cracker. This weakness acquires significance under current conditions, for at present the supply of heavy products is in excess of the shrinking demand.

The design, control and operation of catalytic cracking units have been greatly simplified. These plants are now little more expensive than thermal crackers. In particular, the number and range of instruments required for automatic control appear to have been substantially reduced. The plants are very reliable in operation, periods of a year on stream being commonplace. It is standard practice in these units, as in other refinery equipment in U.S.A., to provide few or no spares, and not to operate a plant until a shut-down becomes necessary by reason of a mechanical failure. Instead, inspection and replacements are carried out in anticipation of failures, to a schedule which is based upon the extensive experience available in the industry.

Cracking catalysts are susceptible to poisoning by sulphur and nitrogen compounds in crude oil. The effect is not permanent, and full activity is restored in the normal process of revivification. This characteristic of catalysts in respect of sulphur compounds is, indeed, beneficial. It makes it possible to produce from a crude of high sulphur content a petrol to satisfactory sulphur specifications. The effect of nitrogen compounds on cracking catalyst is understood to consist in neutralising the acidic silico-alumina complexes which are responsible for catalytic activity. A certain tolerance or compensation for nitrogen compounds may be acquired by reducing the charge rate while maintaining a normal circulation of catalyst. For obvious reasons, it is uneconomical to adopt this expedient to any considerable extent. At one of its refineries in California, Shell catalytically cracks, by this device, a stock containing 0.25% of nitrogen. This is indicated to be the highest nitrogen content permissible in an oil to be charged for catalytic cracking.

Inorganic compounds in crude oil, such as vanadium salts, have a minor poisoning effect on cracking catalyst. Texaco Development has carried out work successfully on the extraction of crude oil with furfural for the removal of nitrogen bases and with other solvents for the removal of inorganic salts. No commercial application of these processes was brought to notice.

It is an interesting commentary upon the chemical engineering prowess of the American petroleum industry that when the first three fluid catalytic cracking units for Bayway, Baton Rouge and Beaumont were urgently required because of the war, they were designed from results obtained on a laboratory bench scale unit, Construction of those plants with a capacity of 25,000 barrels per day each, was commenced without awaiting further experience. Some minor modifications only were made towards the end of construction, based upon information which had in the meantime become available from a 100 barrel per day pilot unit. Those three plants, though now obsolete, are still in satisfactory service. In all, fifty catalytic crackers were in process of design or construction before the first came on stream.

CATALYTIC HYDROGENATION of OIL:

Some of the crude oil produced in California is asphaltic and of such high sulphur and nitrogen contents that it cannot be catalytically cracked. The Union Oil Company handles much of that crude and at present refines it by simply vis-breaking to a residue which can be sold as fuel oil. The yield of fuel oil is 60%, aggravating the disproportionate yields of heavy and light products respectively, which are general throughout the country and are particularly embarrassing in California. The local market for fuel oil is dull because of a reduced demand for bunkers, the replacement of many oil burning locomotives with diesel electric units and the extended use of natural gas for heating purposes. The raw petrol produced from this crude contains 2 to 3% of sulphur and still retains much sulphur after drastic treatment with thirty pounds of acid per barrel. The final product is finished at the unusually low end point of 300°F and is blended into other stocks for disposal. The value of a crude oil which gives such indifferent products and calls for such refining operations is obviously very low.

Aspreviously indicated on page 21 and illustrated in Figure 2, Union Oil has developed a catalytic hydrogenation process for the elimination of nitrogen and sulphur from coker distillate.

The catalyst employed is cobalt molybdate deposited on alumina, and is revivified by burning off carbon deposits about once per week. It is prepared in ~~for~~ the form of pellets of such mechanical strength that they are expected to have a useful life to be measured in years. The pilot plant at Wilmington in which the process has been developed has a charging capacity of three gallons per day. By a single stage of hydrogenation in this plant, at 1,600 lbs. pressure, coker distillate from Santa Maria crude may be purified to about 0.05% each of nitrogen and sulphur. In the original crude, nitrogen and sulphur are present to the extent of about two and five per cent respectively. The consumption of hydrogen is 1050 cubic feet per barrel and the production of gas, excluding hydrogen sulphide, ammonia and propane, about 50 cubic feet per barrel. The Octane number of the hydrogenated petrol is 53 clear and 76 with 3 c.cs of lead per gallon. There is no spread between the motor and research ratings. The liquid volume yield for hydrogenation only is about 104% and for coking only about 84%.

In a commercial plant, hydrogenation would be effected in two stages, reducing the sulphur content of the product to 0.02% and the nitrogen content to a few thousandths of one per cent. The hydrogenated product yields on simple distillation about 50% of petrol and 50% of mixed kerosene and diesel oil. The latter fraction would be amenable to catalytic cracking, but under local conditions further conversion to petrol would probably not be desired. The low octane number of the petrol could be corrected by hydroforming or some other suitable treatment. Because of the greatly superior quality and value of the products, this scheme of refining is easy to justify on paper; it should earn in profits the capital cost of the installation in two or three years. Union Oil has recently announced[+] its intention of spending forty million dollars in the next two years on a commercial plant to apply the scheme to 25,000 barrels of Santa Maria crude per day. The money is needed for :-

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San Francisco Examiner, July 22nd, 1949.

(1) Delayed coking plant.

(2) Hydrogenation and de-sulphurisation plant,

(3) Adaption of boilers to consume the 15% of coke made, instead of the heavy fuel oil now being used.

BY-PRODUCTS.

There is much activity in the industry in the utilisation of waste products, and of products of low market value because of potential surplus production. There is less and less disposition to burn refinery gases to get rid of a nuisance. The wide range of by-products now available from refineries includes special fuels, raw materials for rubber synthesis, solvents, refrigerants, organic intermediates, carbon black, soap substitutes etc.

The propane, isobutane and butane in excess of primary requirements for petrol production in natural gasoline and catalytic cracking plants are usually separated from one another, and shipped to the market in liquid from under pressure in rail or road waggons. The liquefied petroleum gas (L.P.G.) widely used for heating homes outside the distributing areas of gas companies now consists almost exclusively of propane. It is the exception, rather than the rule, to find a home in the country not supplied with L.P.G. The practice is growing of servicing customers from tank waggons rather than with transportable bottles. The bulk container at the home is then a spherical steel shell approaching four feet in diameter. The retail price of LPG is about 8½ cents per gallon and the normal wholesale price in tank waggons 4 cents per gallon. There is over-production at the moment/because in the last few years steel has been made available for manufacture more freely than for storage and distribution.

The price of butane and isobutane for rubber synthesis and other purposes is fairly steady at four cents per gallon. The manufacture of synthetic rubber is expected to continue indefinitely, for the product can compete on its own merits with natural rubber. It is considered that the cost of synthesis operations will set a ceiling to the price of natural rubber and so act as a stabilising influence.

Ethylene from refinery gases is widely used for the manufacture of alcohol, about one quarter of the U.S. market being now supplied from this source. At Baton Rouge, ethylene from the Standard Oil refinery is used in the preparation of tetra ethyl lead. The residual gas from the oil synthesis plant at Brownsville, Tex. is to be stripped of its ethane and ethylene by United States Chemicals, and the products used in the manufacture of organic chemicals.

The market for natural gas has appreciated considerably in the last few years as a result of the construction of numerous long distance pipe lines of considerable diameter. Many more such lines have been planned and approved, and are waiting only on deliveries of steel. It has been claimed by pipe line operators that they can deliver natural gas to any location in the United States at a cost lower than that of locally producing gas from coal. In Illinois, recently, there existed an anomalous situation in which natural gas was available at a lower price per B.T.U. than that of coal! The increased facilities for transporting and distributing natural gas have resulted in an increase in the price at the well. A few years ago, prices ranging from three to five cents per thousand cubic feet were the general rule; today, prices of ten and eleven cents per thousand are most common.

The solvents and other oxygenated organic products obtained from refinery gas, or from gas manufactured for the purpose by vapour phase cracking of oil stocks at high temperatures, include glycerine, various alcohols, ketones, ethers, etc. The gases are used also for alkylating aromatics. Ester salts to be used as substitutes for soap and soap powders are manufactured from similar cracked gases. Production of all these and other by-products is on a very large scale in America and is now being extended in Britain. The Shell Company, I.C.I., and Scottish Oils have been producing synthetic detergents for a market which is still unsatisfied. Some housewives in England however, use the synthetic substitute mainly because it is not rationed and prefer soap because they find it "softer" on the skin. The Shell Company has recently commissioned an extensive plant at Thornton, Lancs, for the manufacture of various chemicals; a plant jointly owned by Anglo Iranian and Distillers Ltd., is being installed at Grangemouth to prepare a different range of chemical products.

LABORATORY EQUIPMENT:

The most outstanding recent development in laboratory equipment and technique within the petroleum industry is the wide use now being made of physical methods for the analysis of hydrocarbon gases and vapours and for examination of catalysts and of ashes from lubricating or crude oils. In spite of its high cost ($35,000) the mass spectrometer seems the most popular of the new analytical instruments and is to be found in all research laboratories of note. Infra red spectrometers are utilised to a considerable extent and some laboratories are equipped with Ultra violet, Raman, arc emission and diffraction spectrometers.++ ++

The mass spectrometer will analyse gases for oxygen, nitrogen etc. and for hydrocarbon gases up to C5 for 21 unknowns. The analysis takes one hour and the calculations about 1½ hours. In one laboratory where simpler gases containing eight constituents are commonly analysed, forty samples may be handled per day with one man operating the instrument and two women the calculating machines. Because of the comparatively short time needed for analyses, the mass spectrometer has superceded the Podbielniak apparatus in laboratories where there is sufficient work to justify the high cost of the instrument. Constituents of petrol up to and including C5, may be determined completely. Closely cut fractions from C6 up to and including C8 hydrocarbons have been analysed after saturation with hydrogen, in which way it has been found

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++ Brummage K.G., Cooke P.W. and Gordon R. R.
New Physical Methods in the Petroleum Industry.
Institute of Petroleum Review, Vol.1, Pp.33 and 65.

possible to identify and estimate all the structures present, though not to assign them individually to saturated or unsaturated hydrocarbons. The instrument estimates simple hydrocarbons more accurately than they can be determined by combustion, absorption in caustic etc. Operators of matriculation standard are able to use the spectrometer for routine work, but a graduate supervisor is needed to ensure that the instrument is in adjustment.

As a further illustration of the application of physical methods of analysis in refinery operations, the water content of by-product acetone is being determined in a recording instrument as a calibrated function of the dielectric constant of the mixture. The same method of analysis has been applied to crude oil, but in this case the technique fails, partly because the mixture is heterogenous and partly because the same calibration does not normally apply to different crudes.

Many laboratories are equipped with bench-scale, catalytic cracking units for the evaluation of oils and catalysts, as distinct from the larger pilot plants maintained by only a few organisations to obtain data for the design of commercial units. Much useful information can be gained from those plants, which in view of the complexity of the commercial process are surprisingly simple in construction and operation. In a typical unit, the reactor is a stainless steel cylinder about three feet long enclosed in a heavy mechanite jacket for even heat distribution. The jacket is in four sections, individually heated by electricity. For each section there is one thermo-couple in the jacket and one inside the catalyst bed in the reactor. The reactor to be used is selected, according to the process being studied, from a range in which the diameter varies from ½" to 1½". The catalyst is not circulated and for use in catalytic cracking tests, alternate periods of about ten minutes duration are devoted to cracking and regeneration. The test is continued in this manner until the number of observations made is sufficient to give results to the desired degree of accuracy.

There is an increasing tendency today for motor fuels to be valued in part according to their octane ratings as determined by the research method. This method gives a measure of the performance of the fuel when the engine is accelerating or labouring at low speeds. The motor method octane number becomes the significant rating when the engine is running at normal or high speeds. It is common practice to base the value of a petrol on its motor octane rating and to assign it a premium value in virtue of its "spread". For petrols of 72 octane, for instance, a spread of four numbers is valued at about ½ cent per gallon.

MISCELLANEOUS.

Much of the natural gas produced in Kansas and piped northwards to Colorado and elsewhere contains about 18% of nitrogen. Because pipe lines are now operating at the limit of their capacity, investigations have been made into the possibility of removing that nitrogen by liquefaction of the more volatile methane or by adsorption in active carbon. It has been established that removal is practicable and economical where the original nitrogen concentration is in excess of 12%. The adsorption process is preferred.

It has been found that naphtha stocks for reforming make more coke in the process if they have been stored. This tendency has been traced to atmospheric oxidation which occurs to a small extent even with straight run, paraffin base naphthas. The trouble may be corrected by contacting the naphtha just before reforming with hydrocarbon gas in a small bubble deck column.

LOW and MEDIUM TEMPERATURE CARBONISATION.

The carbonisation of coal for the production of a freely burning, smokeless fuel is being practised in England on a scale somewhat smaller than that obtaining during and immediately before the war. Two plants are understood to have been closed down and no new installations have taken their place. Supply does not meet the demand, due in part to the diversion of some coke to the export market. There is a considerable margin between the cost of the coal used and the price obtained for the coke, without which the industry could not survive. As it is, a large number of people in England find real or fancied advantages in the smokelessness, cleanliness, bulk etc. of low temperature coke, mostly for use in open fireplaces. They buy the fuel, without compulsion in the form of anti-smoke ordinances, and keep a small industry operating profitably.

In the United States there is believed to be only one low temperature carbonisation plant in active commercial operation. It is likely that, in the near future, more will be established. Authorities in America are very conscious of the pollution of their city atmospheres and of the toll which it takes in the health of citizens and deterioration of property. In Pittsburgh recently, drastic legislation was enacted to reduce smoke emission.

Medium temperature carbonisation is being studied both in U.S.A. and England for the production of smokeless briquettes for slow combustion stoves, water gas plants and low shaft furnaces. The interest is inspired by similar circumstances in the two countries: viz. the high price of the fuels now used (Pennsylvania anthracite $15. per ton on the field, oven coke $15 - $22 per ton delivered to the works; in England, L.T. coke £5. and H.T. coke £3.10/-d per ton both at the works,) and a desire to utilise coals of a quality not suitable for charging to coke ovens.

In the following account of operations witnessed and information gained, carbonisation by the newly developed fluidised technique has been omitted. The fluid process does not yield a lump fuel suitable for domestic and miscellaneous industrial purposes. It is applied to coal so far only as the first stage in the complete gasification of the fuel. For these reasons it is best considered later in conjunction with the second stage, viz. the fluidised gasification of the Char.

Rexco Process.

The Anglo-American Carbonising Corporation has at Mansfield, Notts. two Rexco retorts which were tested and reported upon by the Fuel Research Board in 1935.[+] The retorts are practically identical with N.T.U. plants as employed in the distillation of shale, They are 10 feet in diameter and 20 feet high, taking a charge of 35 tons of coal, which is carbonised by lighting a fire at the top and drawing hot products of combustion downwards through the charge. The two retorts at Mansfield carbonise twelve charges between them in a six day week. Two men are employed on each shift and the total number of employees, including staff, is 14. The coal is a mixture of non-coking "hards" and moderately coking "brights" obtained from an adjacent colliery at a price of 48/-d per ton delivered.

The coke yield is 68% of the charge. It has 6% of volatile matter and is hard enough to stand transport well. It is screened into four sizes which are separated by 1½", 3/4" and 3/8" screens. The coarsest product is used in open fires, the next in Esse and A.G.A. stoves and the third in A.B.A. cookers. The fines are sold to power stations. The average price realised at the works is £5. per ton for the home market and 5/-d more for export.

The gas made, amounting to about 20,000 cubic feet per ton, is wasted. The calorific value is 120 B.T.U. per cubic foot when part is circulated and burned in the retort. Towards the end of the cycle of operations when

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[+] Report of Test by the Director of Fuel Research on a Retort of the Coal Research Syndicate Ltd., at Mansfield Colliery, Nottinghamshire. H.M.S.O.

the circulating gas is not burned, the calorific value rises to 300 B.T.U. per cubic foot. The total loss is about 29 therms per ton of coal.

The tar yield is about 19½ gallons per ton and amounts to 2,000 tons per annum. The tar is difficult to dehydrate and as sold still contains 9% of moisture. Difficulty is being experienced in disposing of the tar, because, it was stated, distillers do not like mixing it with high temperature tar and do not have sufficient of the low temperature product to treat on its own.

At one time, the liquor was concentrated for the preparation of ammonium chloride, but the operation was found uneconomic. The effluent is now merely poured into the ground which is sufficiently porous to absorb the liquor as fast as it is made.

The plant has been functioning for about 15 years. It was acquired by its present owners for £12,000 and is now valued at £30,000. The annual profit is £5,000.

<u>Disco or Wisner Process.</u>

++ The operating Disco plant + at Champion, Pa. comprises three rotating, horizontal retorts about 8 feet in diameter and 100 feet long, with a charging capacity of 100 tons per day each. The plant has been worked steadily beyond that capacity, yielding large, smoky lumps of coke with incompletely carbonised cores. This method of operation had been adopted deliberately because of the unsatisfied demand for the product. Customers have not complained about the quality of the coke.

The coal charged is a washed and dried coking slack, screened through a 3/8" sieve. Before entering the retorts, the coal is heated to 600°F to control, by oxidation, its agglutinating behaviour. The retorts are heated mainly by products of combustion

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++ Trans. Am. I.M.M.E. Vol.139, P.328, (1940) from a furnace, with a re-circulation of waste gas for temperature control. The heating flue is the annular space between the retort and a surrounding, externally-insulated shell. Additional heating results from the leakage of air through the seal between the rotating retort and stationary coke discharge pipe. It is likely that that leakage is deliberate. The gas make is 3,000 or 4,000 cubic feet per ton, with a nitrogen content of 40% and a calorific value of 400 B.T.U. per cubic foot.

The coke is cooled, crushed to 6" and screened at 3" and 1/2". The two lump fractions are sold at an average price of $11.80 per ton as compared with a purchase price of $4.85 per ton for the coal. The screenings below 1/2", amounting to 25% of the coal feed, are returned to the retort together with 10% of high temperature coke breeze, thus providing an added measure of control over the coking properties of the coal.

The operating staff comprises ten men on each shift. The plant has been working for fifteen years and was due to be closed down in the middle of 1949, as soon as new works adjacent were completed.

The new plant comprises seven retorts 120 feet long by 9 feet in diameter with a charging capacity of 150 tons per day each. The agglutinating value of the coal is to be reduced by preheating and blending with breeze as in the old unit. The installation of further retorts on the same site is unlikely because the mines supplying coal to the plant are nearing the end of their productive life.

An interesting adjunct to the new retorts is a flash drying plant which is described later in this report.

Chemical Construction Process.

In collaboration with the Chemical Engineering department of Yale University, Chemical Construction is developing a process based upon the long abandoned

Carbocoal process, but concentrating in one stage of operations all the essential functions of the original two-stage treatment. A small, five ton per day retort was operated for some years at Newhaven, but the work was interrupted by the war. In January, 1949, a 100 ton per day plant was commissioned at the steel works, Bethlehem, Pa. Because of certain patent applications pending, no announcement has been made regarding the process and visitors have not been admitted to the plant.

The object of the process is to produce a fuel strong enough for use in water gas generators, cupolas etc., from coal which is too strongly swelling to be carbonised in coke ovens. The coals to be used at Bethlehem are of a B.S. swelling index greater than 6½. For coking in ovens the swelling index should be between 4½ and 6½. At Newhaven, the swelling of the coal was reduced by blending it with non-coking coal or iron ore. Satisfactory carbonised briquettes were made from a 50:50 mixture of Pocohontas coal and anthracite, and from a 30:70 blend of crushed iron ore and Pocohontas coal. The iron ore briquettes were made for a steel company but nothing is known by Chemical Construction of any further tests to which the briquettes have been put.

At Bethlehem, the swelling and agglutinating properties of the coal are being modified by a preliminary oxidation. For the subsequent briquetting and carbonising operations to give a satisfactory product, the swelling index of the coal must be 1 or less, but the oxidised material must have a slight tendency to cohere. A weakly caking coal meeting this specification may be briquetted and carbonised without the preliminary oxidation. A useful test of the prepared coal is to heat it as rapidly as possible in a crucible over a gas burner and to cool it after all the volatile matter has been evolved. If the char can be poured out of the crucible like dry sand, the coal is useless for the process on its own. If, however even a small portion of the char is agglomerated, the coal may be used.

In the plant at Bethlehem, pulverised coal is pumped by the Fuller Kinyon method through a tubular heater to effect the oxidation. The temperature required varies with the coal to be treated; for Pocohontas coal it is 750°F. The oxidised fuel is cooled, mixed in a pug mill with about 10% of tar, and then briquetted in a roller press to form pillows about 2" x 2" x 1". The binder for the briquettes is part of the tar recovered from the retort. It is prepared by treatment with concentrated sulphuric acid, the consumption of which is 15 lbs per ton of coal. The purpose and effect of the acid treatment are to reduce the solvent action of the tar on the coal at the time when it is passing through the plastic stage in the retort. The action of the acid may be a sulphonation or extraction, but it is not at present understood. The control of the solvent power is said to be essential for the production of a satisfactory carbonised briquette.

The retort itself is an internally heated firebrick shaft about 20 feet high. The top section is 10 feet in diameter; the middle section tapers from 10 to 6 feet in diameter in conformity with the shrinking of the charge; the bottom section is 6 feet in diameter. Heating gases are burned in a combustion chamber surrounding the 6 foot section and are then admitted to the retort through tuyeres at the top of that section. Cold gas is recirculated to the coke discharge pipe and in cooling the product, returns to the retort about 65% of the heat needed for carbonisation. The temperature to which the briquettes are heated is 750°C to 850°C, according to the purpose for which the coke is intended. For a fuel to be used in open fires, the lower temperature is recommended; the briquettes for foundry work, slow combustion stoves, water gas plants etc., should be heated to the higher temperature.

The raw briquettes are hard and dense, and smell strongly of naphthalene. The carbonised briquettes are very hard, of uniform fine texture and shrunken to the extent of about 30%. They contain less than 1% of volatile matter and their bulk density is 45 lbs per cubic foot as compared with about 35 for oven coke. In a cupola charged with the carbonised briquettes, it has been found possible to put 40% more iron into the charge and to operate with a 6" blast instead of the usual 20".

The gas made is 40 therms per ton, but the calorific value is low. The tar yield, viz. 30 gallons per ton, is three times as high as from a coke oven. It is thin tar of high quality because of the repeated carbonisation of the heavy fraction.

Miscellaneous.

In conjunction with the University of Wyoming, the Golden station of the Bureau of Mines has prepared from local sub-bituminous coals a carbonised briquette hard enough for metallurgical purposes. The procedure adopted was to mix 100 parts of lignite char with 5 of local coking coal and 7 to 9 of petroleum pitch. The mixture was briquetted at pressures ranging from 3000 to 10,000 lbs per sq. in. and the briquettes were carbonised at 1,600°F. The briquettes were hard, though not as hard as oven coke. They had a granular texture characteristic of coked blends and usually indicative of a reactivity higher than that of normal coke. The best briquettes were those made at the highest pressures and with the greatest admixture of coking coal.

In England, Powell Duffryn has produced similar briquettes by carbonisation at 900°C in a coke oven. It has been established that intermittent vertical retorts could be used for the purpose equally well. The briquettes are an excellent fuel for domestic purposes and are sold at a retail price of £7. per ton.

COMPLETE GASIFICATION.

Many interesting developments are in progress abroad in the field of gasification of coal, mainly directed towards the production of synthesis gas. The fluid bed technique, best known for its application in catalytic cracking, is being tried in gas generators with a measure of success which is not yet clear. Coal is being gasified in suspension on a large experimental scale by the Koppers process and on smaller scales using other techniques. Lignite is being gasified in an externally heated retort in a large pilot plant. The well established water gas process is being further investigated in the laboratory and gas works. In England gasification of coal under pressure in an atmosphere of hydrogen receives continued attention. The Lurgi process is being studied in U.S.A. and plans are well advanced for work to be done on the process in England. In the United States, Belgium and elsewhere, further experiments are proceeding on the gasification of coal in the seam.

FLUIDISED CARBONISATION and GASIFICATION:

It was probably in the metallurgical industry that the first application was made of the technique of handling finely divided solids in the form of a dense suspension in gases. Neither at that time, however, nor later when the principle was adopted in the Fuller Kinyon system for transporting pulverised fuel, was the significance of the technique properly appreciated. Fritz Winkler patented his gasification process in 1922, likening the conditions in his producer to those of a "boiling bed". Although he understood the characteristics and advantages of a fuel bed densely suspended in the gasifying medium, it would seem that he had no idea of the wide range of uses to which the principle could be usefully applied. It was left to W.W. Odell to appreciate the full significance of the discovery and, in 1929, to patent its application to a considerable number of industrial operations, excluding, of course, gasification. Lewis and his associates at M.I.T. took an early interest in fluidisation and instituted studies of the mechanics of the process, which are still in progress. It is to this school that we owe most of the published information on reaction kinetics, material and heat transfer etc. in fluidised solids.

By its application to catalytic cracking in the years, the fluid bed technique has virtually revolutionised the petroleum industry. Since that time, the process has been used on an experimental or commercial scale in a great variety of applications including lime burning, oil synthesis, gasification and carbonisation, ore roasting and the manufacture of phthalic anhydride. There was at one time, indeed, a tendency for the virtues of the technique to be unduly exaggerated and for fluidisation to be advocated as a solution for all technical difficulties in chemical industry. A more temperate opinion has now become manifest it and is generally realised that the characteristics of fluid beds which are of almost inestimable value in many applications are actually undesirable in others. The gasification of coal is a case in point. Opinion is sharply divided on the prospects of the process achieving any greater measure of technical success in the gasification of bituminous coal than has been achieved by Winkler in the gasification of lignite. Those authorities who are, or have been, working on the process appear reasonably confident of ultimate success; critics are inclined to emphasise the following difficulties and disadvantages :-

(1) Because each piece of fuel becomes progressively smaller as gasification proceeds, the size consist of particles in the fuel bed cannot be conducive to good fluidisation, with freedom from slugging and undue entrainment.

(2) For the same reason, some carry-over of fine particles, difficult or impossible to trap with cyclones, is almost inevitable.

(2) (To minimise these losses, coal for fluidised gasification is desired to be crushed to pass a 20 mesh sieve with nothing passing 325 mesh. The nearest approach to this specification attainable with pulverisers now in common use is 99.9% through 20 mesh with 18-20% through 325).

(3) Because of the uniformity of the fuel bed, much combustible matter must be rejected with the ash.

(4) Because of the uniform temperature conditions throughout the fuel bed, the temperature of the gas leaving the plant must be high and the efficiency of the process correspondingly low, unless great care is exercised in the recovery and effective utilisation of waste heat.

The authorities now engaged in commercial, as distinct from academic, studies of fluidised carbonisation and/or gasification include Standard Oil Development and Pittsburgh Consolidation Coal working together, Hydrocarbon Research Inc., the Anthracite Institute and Imperial Chemical Industries Ltd., Some work was undertaken at the Institute of Gas Technology on a pilot plant scale about three years ago, but has now ceased.

Standard Oil Development first interested itself in fluidised gasification by instituting process studies in a small plant at the Bayway laboratories of Standard of New Jersey. That work is still being continued, the subject at present under investigation being the kinetics of reduction reactions. Results are not published. The plant is intended purely for academic studies and not for the development of a process.

Pittsburgh Consolidated Coal first became actively interested in fluidised carbonisation when the Company acquired the Disco plant near Imperial, Pa. by amalgamation of various interests, including the Disco Co., which now form P.C.C.. For two years, Disco had operated a small fluidised carbonising plant, believed to be about two feet in diameter, to supply char for blending with coking coal fed to the main retorts. The fluid bed retort was closed down before the amalgamation, because it was regarded as having no future, but it was returned to service by Pittsburgh Coal late in 1947. The plant is understood to have been operated continuously since that time, but visitors are not admitted. The coal charging rate is known to be 1,800 lb. per hour, and the plant has not been operated under pressure. When coal of 35% volatile content is carbonised at 600°C, the yield of coke is 65% and its volatile content 10%. The tar yield of 28-30 gallons per ton is close to the Fischer assay yield. Much trouble has been experienced with the clogging of cyclones and product lines by a sludge of dust, tar and water. The gas is discharged to atmosphere carrying much dust with it.

Early in 1947, Standard Oil and Pittsburgh Consolidation entered into an agreement for a joint investigation of fluidised gasification and it was published that, in due course, a mammoth commercial plant would be constructed in Pittsburgh. In summer, the major product of gasification and synthesis operations would be petrol; in winter, a rich gas to supplement inadequate supplies of natural gas for domestic and industrial heating. As a preliminary step towards the achievement of that goal, a $500,000 pilot unit would be installed at Library, Pa. and operated for perhaps two years to obtain information needed for the design of the large plant.

The pilot unit was commissioned in November, 1948. Some visitors have been admitted to the plant, but the inspection permitted is incomplete and only meagre information is disclosed on the scale and nature of operations. In early tests, the medium used for fluidising and gasifying the fuel was air at substantially atmospheric pressure. The fuel was char from the fluidised carbonisation unit at Imperial. More recently, oxygen has been employed instead of air, but at the time of the last advice received, the high pressure for which the plant was designed had not yet been applied.

When the plant was operated with air, the main problem appeared to be the entrainment of finely divided fuel in the product gas, which was blown to waste. The cyclones installed were incapable of precipitating the very fine dust resulting from partial combustion of fuel particles. The quantities escaping appeared sufficient to constitute both a serious loss of fuel and a nuisance in the further utilisation of the gas. From the estimated dimensions of the generator (diameter about two feet), the gas velocity in the fluidised bed (probably rather less than one foot per second), the stated ash content of the fuel bed (up to 40%), and the ash content of the feed (probably not less than 12%) it was conjectured that the fuel supply rate was not more than 100 lb. per hour and the dust loss not less than 20% and probably much higher. It was conjectured, also, that to maintain a fuel bed with an ash content approaching 40%, no residue was withdrawn from the bottom of the plant. It was believed, instead, that all the ash was discharged overhead in the gas, a method of operation which, by accident, had been found quite satisfactory for Winkler generators.

The dust problem was greatly aggravated at the time of inspection by the conditions of operation. When the fuel is gasified, as it is intended it ultimately shall be, with oxygen and under high pressure, the volume of gas, measured under plant conditions and calculated to each ton of fuel consumed, will be greatly reduced. Its carrying capacity for dust will likewise be reduced, probably to a point at which the resulting loss of fuel will not be significant. However, the cleaning of the gas so that it may be used for catalytic synthesis will still require considerable study. It seems likely that in due course it will be found possible to withdraw most of the ash from the base of the generator. That ash will contain much combustible matter, because the fuel bed cannot be improverished too much if gas production is to be maintained at a high rate. It is intended by Pittsburgh Consolidation, apparently, that the "ash" shall be used as a fuel for boilers.

It is understood that in early operations with oxygen, trouble was experienced due to leakage of oxygen and entrained fuel between the brick lining and steel shell of the generator. Similar troubles have occurred in other types of plant. The remedial measures are structural in nature; they are well understood and easy to apply.

The operation of the plant under pressure calls for the development of a satisfactory method of feeding and metering powdered coal into a vessel under pressure. This problem is being studied by other authorities also, in connection with other fuel activities.

It is considered by Standard Oil and Pittsburgh Consolidation that it will be two years at least before the fluidised gasification process, if successful, will have been developed to the stage at which a commercial plant can be designed. In the meantime, it appears that no further thought is being given to the project which received so much publicity in 1947. There may be two reasons for this abatement of interest; firstly, the technique of fluidised gasification probably calls for much more development than was at first anticipated; secondly, new discoveries and increased production of petroleum have deferred the day when petrol from coal will be able to compete economically with the natural product.

Pittsburgh Consolidation considers that there is a prospect of profitably carbonising coal by the fluidised technique, without subsequently gasifying the char for a synthesis operation. The presumed market for the solid residue would be public utility and other power stations. To evaluate the char for this purpose and to gain experience of its utilisation, work has been initiated on fundamental combustion studies at Battelle and on practical boiler firing tests at a local power station. The capital cost of a 1000 ton per day fluidised carbonisation plant is estimated at $1,000,000 to $1,500,000. Current prices for coal, tar and gas in Pittsburgh are reppectively $4.50 per ton, 12 cents per gallon and 30 cents per million B.T.U. If prices of coke and tar be depreciated somewhat, for the sake of being conservative, a condensed profit and loss account for a commercial project would appear as follows :-

Expenditure.	$	Revenue.	$
Coal, 1 ton,	4.50	Coke, 0.65ton at $4.50 less 10%	2.63
Operating costs per ton	0.75	Tar, 28 gal,@ 8c.	2.24
Balance, available for capital charges, and equivalent to 8% on $1,500 for 330 tons per annum.	0.37	Gas, 2.5 million B.T.U. at 30c. per million.	0.75
	$5.62		$5.62

Hydrocarbon Research Inc. has studied the fluidised carbonisation and gasification of coal, the work being financed, it is understood, by Standard Oil and Texas. The small pilot plant used in the carbonisation tests is illustrated in figure 3. The heat required for the process is generated by burning some of the circulating char, so raising the temperature by 100°F or a little more. The heated char is mixed with the coal, 25% of the carbonisation being effected in the inlet pipe to the cyclone and retort, the rest being done in those vessels. No difficulty is experienced in carbonising in this way a coking Pittsburgh coal and in reducing its volatile content from 30% to 5%,

H.R.I. appears pessimistic regarding fluidised gasification. Work has been done on the subject and it was implied that some progress has been made towards a solution of the problems involved. In discussing the subject, however, more emphasis was laid on difficulties than on achievements. No information was divulged regarding the results of work done or the direction in which solutions to problems were being sought. The considered opinion of the organisation on fluidised gasification is, no doubt, reflected in Mr. Keith's announcement that for the South African synthesis project he is recommending his firm's version of the Lurgi process.

The Anthracite Institute is the research organisatio of the anthracite industry in U.S.A. The anthracite mines of Pennsylvannia produce screenings below 1/8" in size at the rate of 6½ million tons per annum in excess of consumption. Stocks already accumulated amount to 200 million tons. Large quantities of anthracite have also been deposited in the form of silt on river banks. It has been calculated that stocks of screenings are sufficient to supply a 50,000 barrel per day oil synthesis plant for thirty years.

With a view to the possible development of a process to convert this waste material into useful gas, the Institute has constructed a small fluid bed producer and has operated it since the middle of 1948. Anthracite fines passing 95% through 14 mesh and 55% through 60 mesh are gasified by air and steam at atmospheric pressure. Fuel is charged at the rate of 300 lb. per hour. The gas velocity in the producer is 10 f.p.s. and in the disengaging zone 2 f.p.s. These high velocities would appear to be permissible because :-

(1) The specific gravity of the anthracite is high (about 1.6).

(2) The particles are coarse.

(3) Losses in the outgoing gas are of little significance because the fuel is a waste product.

Only 50% of the anthracite is gasified; 30% is entrained in the gas and separated by the cyclone without being returned to the producer; about 10% is lost in the ash and the remaining 10% is accounted for as moisture present in the coal as charged. The calorific value of the gas made appears to be usually 80 or 90 B.T.U. per cubic feet, but has been reported as high as 150 B.T.U. The temperature in the fuel bed is only 1800°F approximately instead of the

2,300°F desired; for some reason not yet understood, the temperature cannot be raised further by mere adjustment of air:steam ratios.

Imperial Chemical Industries has devised, and is developing its "moving burden" process with the object of producing gas for ammonia synthesis and hydrogenation, using comparatively cheap coal not suitable for charging to coke ovens. It is hoped that the gas so made will be cheaper than water gas, not merely because of the use of non-coking coal but also because a single integrated process should be more efficient and economical than the successive steps of carbonisation in coke ovens and gasification in intermittent generators. I.C.I's. patent on the process, B.P.582,055 is admitted to conflict with Standard Oil's patent BP.586,391, and the latter is conceded to have prior rights. The confusion arose out of the practice of suppressing publication of specifications for security reasons during the war. The respective applications were both lodged in 1944, but it was not until late 1948 that it became known to I.C.I. that vital features of its process had been anticipated by Standard Oil by only two or three months. The Company is continuing development of the process because it may eventually transpire that the experience which will have been gained in the technique of operation will prove of considerable value even without the patent rights.

The main feature of the moving burden process, which distinguishes it from the fluidised gasification process being actively developed by Standard Oil, is that air is used instead of oxygen for the exothermic gasification reaction. The plant is arranged to separate the nitrogen - containing blow gas from the nitrogen-free water gas. It is I.C.I's contention that separation of these two gases by simple mechanical operations must be more economical than the separation of atmospheric oxygen and nitrogen by liquefaction and fractionation. Oxygen is regarded as too expensive a commodity for general

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consumption in large scale industrial operations.

The process has been studied in the laboratory in vessels 4" and 12" in diameter. The construction of a pilot plant was commenced early in 1949, but at the time of the last advice received, it had not yet been completed. The flow of materials through the pilot plant and its main dimensions are indicated in figure 4. In the portion of the plant so far constructed, it has been possible to carbonise coal in a fluid bed, but not to gasify the char with steam. A satisfactory circulation of the char, used as the internal heating medium for the retort, has been achieved. Some trouble has been experienced from leakage of air and entrained fuel between the firebrick lining and the shell of the combustion vessel. The remedy for this trouble is not difficult to find and apply. It has been found that the carbon monoxide in the blow gas may be kept satisfactorily low - lower, probably, than the 5% to 8% present in the waste gas from an intermittent water gas generator. It is not yet known to what extent it will be economic to reduce the ash content of the residue which must be withdrawn from the circuit. Some thought is being given to the use of a small subsidiary gasifier to consume the carbon in the ash purge.

The process is regarded by the Company as one for operation at atmospheric pressure only. Where it is desired to gasify coal under pressure, it would be better to use oxygen in another type of plant. This preference is based on the argument that if large volumes of nitrogen are to be compressed, the compression might as well be done in an oxygen separation plant and some advantage gained by using oxygen for gasification.

The moving burden process must be regarded as being in a very early stage of development. It seems likely that some years must elapse before the process will be ready for commercial adoption.

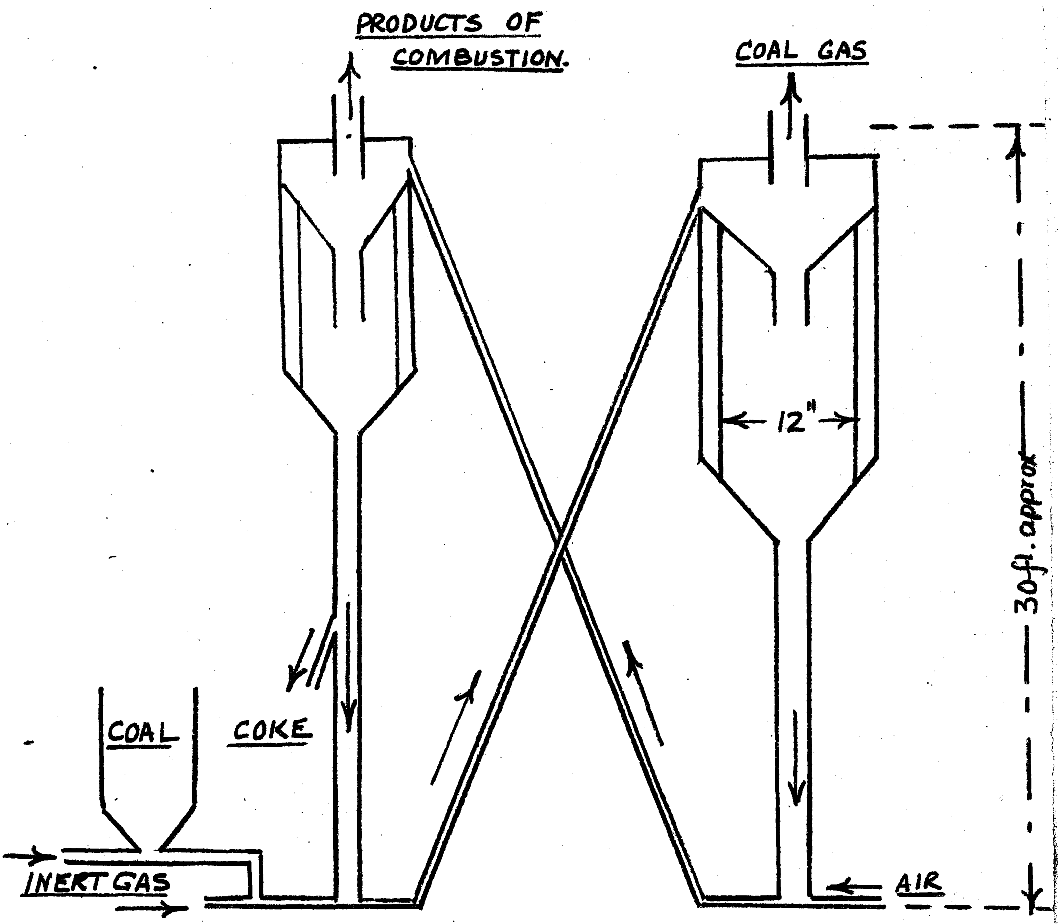

FIG 3. FLUIDISED COAL CARBONISATION HYDROCARBON RESEARCH INC.

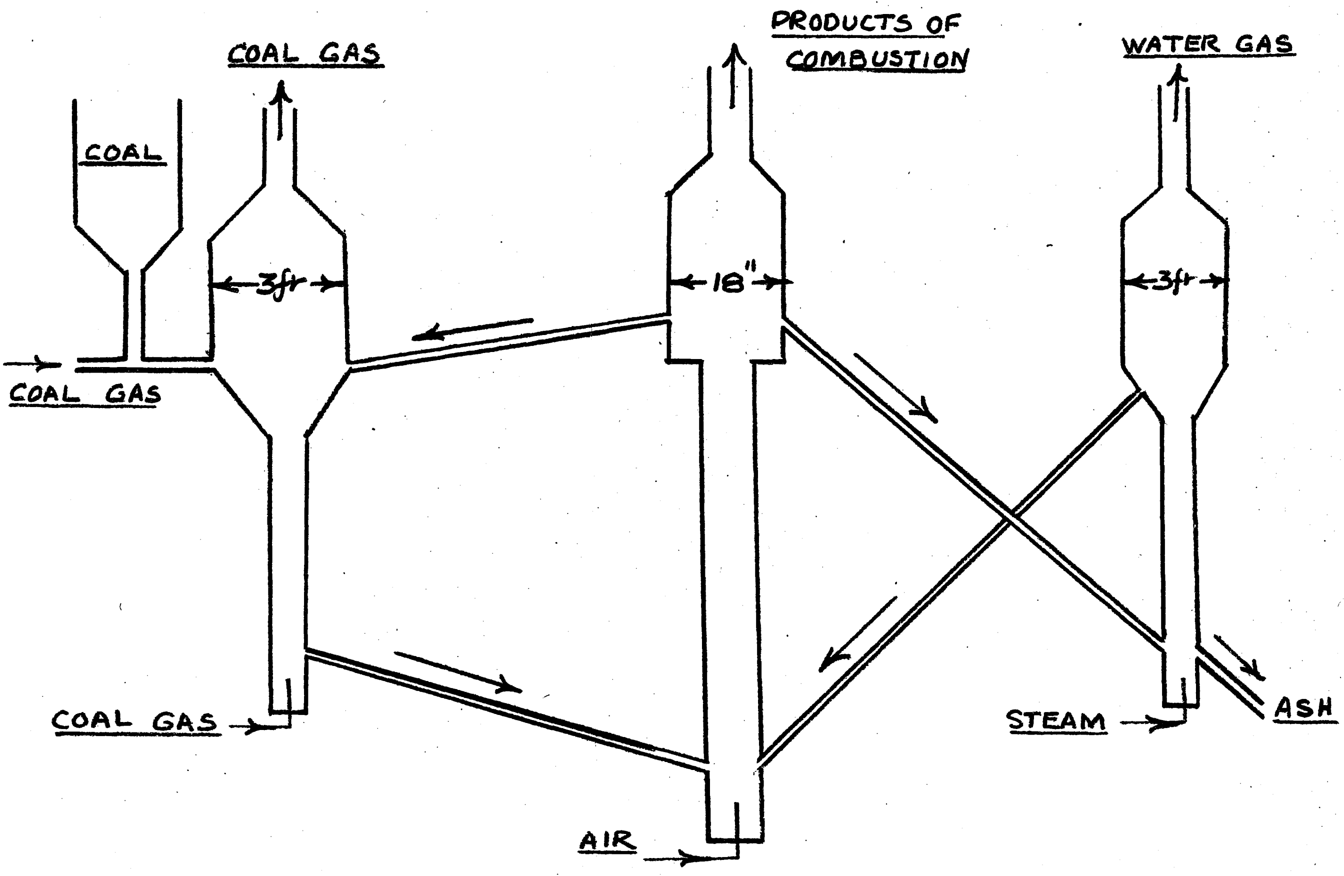

FIG 4. FLUIDISED COAL GASIFICATION IMPERIAL CHEMICAL INDUSTRIES.

Miscellaneous. Very little is known of the actual results of the work done over a period of twelve months at the Institute of Gas Technology, and opinion appears to be divided on the interpretation to be placed upon them. The Director is sceptical of the prospects of fluid bed gasification, but Dr. A. D. Singh who carried out the work at I.G.T. and is now in private practice as a consultant, is optimistic so far as the carbonising operation is concerned. He considers the high production cost of oxygen an economic bar to complete gasification, but seems to have no doubt regarding the technique of the process. He is reported to be endeavouring to interest State authorities in an ambitious project of fluidised carbonisation with the object of reducing smoke emission.

From other sources, it was learned that the results of the work done at I.G.T. had failed to indicate any improvement in the cost of gas production as compared with normal water gas manufacture.

I.G.T. is continuing fundamental studies of the gasification process in a small, fluid-bed unit.

It has been proposed to undertake work on fluidised carbonisation and gasification at the Golden Station of the Bureau of Mines. At the Fuel Research Station in England investigations on that subject have just commenced.

GASIFICATION in SUSPENSION:

The Koppers process using oxygen and steam, the Bureau of Mines process using mainly steam heated to very high temperatures, the I.G.T. process using oxygen and steam and the Bureau of Mines adaption of the F.R.S. vortex process have all been devised for the production of gas consisting substantially of carbon monoxide and hydrogen. Combustion of coal suspended in air is reviewed elsewhere in this report. To the extent that combustion is itself a gasification process, this division of the subject is scientifically illogical.

Gasification, however, as here understood, is directed to the production of gases for use as materials; in combustion it is the incidental heat of the reaction which is the desired product. The division of the subject as indicated is, furthermore, a great convenience in preparing a report intended, as this one is is intended, to be of practical rather than academic interest.

<u>Koppers Process</u>.+ Most of the documentary evidence of the experience gained in the early development of the Koppers dust gasification process in Germany during the war has been lost by reason of war damage or other causes. For the information required in planning the resumption of investigations in U.S.A. and Germany, it has been necessary to draw extensively upon the memories of engineers - notably Totzek - associated with the original project. At the time when the Koppers Company of America was planning a pilot plant for further studies of the process, the Bureau of Mines became interested in it for the production of hydrogen and synthesis gas at its demonstration station, Louisiana, Mo. A joint investigation was therefore planned. The Koppers Company has designed and installed the plant without guarantees except in regard to mechanical performance. The Bureau has borne the cost and is to undertake the actual operation.

The process has been adopted advisedly by the Bureau of Mines, in spite of its inadequate development, because the Bureau urgently needs a process which will gasify a coking coal in one stage of treatment. The coals which must be used for any synthetic fuel plant that may be installed east of the Mississippi are strongly coking. If the two stage process of carbonisation in coke ovens and orthodox water gas production can be replaced by a single, efficient, gasification operation, a considerable saving would be effected in what is the

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See Newman L.L. "Oxygen in the Production of Hydrogen or Synthesis Gas". Ind. Eng. Chem. <u>40</u> p.559.
See also F.I.A.T. Report 1303.

greatest single item of cost in the production of oil from coal.

The plant is as illustrated by Newman (loc.cit. p.568) Pulverised coal is to be suspended and gasified in a stream of oxygen and steam, the latter superheated to 2,500°F in a continuous pebble bed heater. The designed capacity of the plant is 28 tons of coal per day to yield 90,000 cubic feet of gas per ton. Oxygen consumption is expected to be one ton per ton of coal or 25% of the volume of gas. Steam consumption is expected to be 2,900 lb. per hour. The period of retention of gas in the furnace is 1 to 2 seconds, a high temperature being necessary to complete gasification in that time. The gas is expected to issue at 2,300°F and is to be cooled immediately to about 1800°F by re-cycling cold gas. This recirculation of gas is a safety measure designed to prevent explosions in the event of the fuel supply failing. In such an event, the oxygen still admitted to the generator will be consumed promptly by the recycle gas and will not pass forward, as oxygen, to the gas holders etc. where it could form an explosive mixture. The gas to be made with oxygen and steam is expected to contain 12-15% of carbon dioxide and about 40% each of carbon monoxide and hydrogen.

The plant at Louisiana, Mo. was commissioned in May, 1949. Little is known of its performance, but it is understood to have operated for a short period at full design capacity and to have produced a gas containing 47% of hydrogen, 32% of carbon monoxide and 16% of carbon dioxide. Consumptions of coal and oxygen were 30 lb. and 240 cubic feet, respectively, per thousand cubic feet of unpurified gas. These figures suggest that an appreciable amount of unconsumed carbon was carried forward in the gas.

It is understood that two Koppers plants are being erected by the French national collieries organisation and that Koppers is building an experimental unit at Essen, in which the reaction is to be carried out at about twenty atmospheres pressure. The plant is expected to be completed late in 1949.

<u>The F.R.S. Vortex Process</u> is being adapted at the Pittsburgh station of the Bureau of Mines for a gasification operation as distinct from combustion. The experience gained to date has indicated the need for certain modifications, particularly in respect of the method of introducing the fuel. The Vortex burner is mentioned at greater length in this report in connection with combustion.

<u>Gasification in highly superheated steam</u> forms the main subject of study at the Morgantown laboratories of the Bureau of Mines[+] Pulverised coal is gasified while suspended in steam superheated to such a temperature that most of the heat for the endothermic reaction is contained in the reactants themselves. To the present time, only preliminary work has been described in publications. Using oxygen and steam at normal temperatures, entrained pulverised coal has been gasified in its downward passage through a generator six inches in diameter and seven feet high. The fuel is consumed in a period of about one second. The efficiency of gasification is stated to be 65% for the clean gas or 80% if credit be taken for the unconsumed carbon leaving the generator entrained in the gas. Correlation with other information indicated that the "gasification efficiency" was calculated from the heating value of the cold gas and the sum of the heating value of the coal and the total heat of the steam supplied. The rate of fuel supply is 30-50 lb. per hour.

A larger unit with a capacity of 200 lb. of coal per hour, and designed to utilise highly superheated steam, has just been built. It is hoped to preheat the steam to 4,000°F; if this temperature is achieved, there will be little or no need for supplementary oxygen. In preliminary tests, steam has been superheated to 3600°F

++ Schmidt L.D. McGee J.P. and Slone M.C. A Pilot Plant for Gasifying Powdered Coal Entrained in Oxygen and Steam Chemical Engineering Progress, Vol.44, No.10, P.737.

in pebble stoves, two of which are installed for alternate heating with natural gas and preheating of steam. Dolomite pebbles have been found satisfactory at 3600°F though not at 3000°F. The extent to which steam preheat may be used instead of oxygen is determindd by the respective costs. It has been calculated that if oxygen were to cost $8 per ton, it would pay to spend $4. per ton on superheating steamto 4000°F.

The Institute of Gas Technology, Chicago, is working on a small scale to develop a process for the gasification of pulverised coal in an atmosphere of oxygen and steam or air and steam. Coal pulverised to pass an 1/8" screen is carried in a stream of air at 50 lb pressure through a 1/4" nozzle in which the air is expanded substantially to atmospheric pressure. Steam at 700°F and oxygen are added, and the mixture is admitted at high velocity to a "cyclonizer". This piece of equipment resembles a fan casing and its function is to provide the opportunity for further attrition between coal particles and between particles and the casing. About half the fine pulverising is effected in the nozzle and half in the cyclonizer. After addition of more steam and oxygen, the gas from the cyclonizer is admitted tangentially to a furnace. The period of detention in the reaction zone, which is 3 feet high by 21 inches in diameter, is about five seconds. About 70% of the coal is gasified and the outlet temperature is 2,100°F. The gas obtained is of the following approximate composition when only oxygen and steam are used :-

Nitrogen	3%
Hydrogen	40%
Carbon Monoxide	40%
Carbon Dioxide	17%

Calorific value 280 B.T.U. per cub. ft.

It is intended at some later date to use steam superheated to 2,000°F so as to reduce oxygen consumption.

At Battelle, some work has been done on the gasification of lignite in suspension, using an externally heated, alloy steel tube. The object of the work was to compare the respective merits of external heating and gasification with oxygen.

GASIFICATION in an EXTERNALLY HEATED RETORT.

At the Bureau of Mines Station, Grand Forks, N.D., work is proceeding on the gasification of lignite in the Parry retort,[++] in which the fuel passes downward through the narrow annulus between two concentric vertical cylinders. The outer cylinder, about four feet in diameter, is heated to a temperature of about 1950°F and a useful life of about 10,000 hours is considered necessary to justify the process. The plant may be used either for dry carbonisation of non-coking coal or for complete gasification in which case steam is passed through the charge. The heated outer tube has been made of various materials, viz :-

1. Mild steel with a sprayed-on coating of refractory metal.
2. Mild steel clad with a thin sheet of 26% chrome alloy in each side.
3. A cast alloy tube containing 20% Ni. and 10% Cr.
4. A fabricated alloy steel of high nickel and chromium contents.

Cylinder No. 1 was found unsatisfactory after a very short period of service. The clad tube suffered distortion considered due to the unequal expansions of the mild and alloy steels. After reversal, so as to bring the previously unaffected section of the tube into the high temperature zone, the plant was again operated for a short time. At the end of 3,500 hours

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[++] Bureau of Mines, R.I. 4120.

of service, however, the tube had distorted beyond all possibility of further use. The cast tube was designed to give an annular space for the coal passage 2¼" in width. Because of slight eccentricity in cross section and considerable surface roughness, it was found impossible at first to move lignite through the retort. After grinding the surface as smooth and as true as was practicable, the tube was returned to service. However, the two sections in which the cylinder was cast parted completely at the weld and the tube had to be discarded. The fabricated cylinder is now in service and gives promise of easily meeting the requirement of a life of 10,000 hours.

The weakness of the Parry retort, which would seem to preclude its adoption in any large scale enterprise, is the one experienced in connection with the cast alloy tube. The design calls for the construction of two concentric tubes of such accurate finish that there will be a substantially uniform annulus only 2¼" wide between them. This requirement places a severe limitation upon the diameter of the tubes and, therefore, on the capacity of individual retorts.

A small, externally-heated pilot plant for fundamental studies of the steam-carbon reaction has been installed and operated at the Battelle Institute[+] with funds supplied by Bituminous Coal Research Inc.

WATER GAS.

There is a tendency in America for the proportion of water gas distributed by public supply authorities to become progressively smaller. Because of the greater demand for distributed gas, however, the actual volume of water gas manufactured is probably not

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[**] Foster, John F. Production of Water Gas from Pulverised Coal. Industrial and Engineering Chemistry, Vol.40, P.586.

reduced. Natural gas is now more generally available than it has ever been before; since the middle of this year even New York has been served in part with natural gas transmitted by pipe line from Texas. The cost at the outlet of the pipe line is understood to be 40 cents per thousand cubic feet, for gas of 1,000 B.T.U. quality. Natural gas, so far as it is available, is the first choice of the supply authorities. In a large area of the United States, it is distributed as such, and on its own, for domestic and industrial purposes. Where supplies of natural gas are unavailable or inadequate, coke oven gas is the next choice of the authorities. Carburetted water gas is used only for peak load operation, in which service the advantages of low capital cost and low standby charges are able to outweigh the disadvantage of a high fuel cost..

The choice of gases for public distribution in England is somewhat similar except that, of course, natural gas is not available. Economic conditions in the British gas industry favour the production of coke as well as gas. Coke is worth as much as £5. per ton in some works, and there is an export market which the country is anxious to exploit to the best advantage. Carbonisation is regarded as the first choice of processes for base load gas; water gas is manufactured to meet normal peak loads, at which time the mixture would comprise about 30% of coal gas, 65% of blue water gas and 5% of oil gas; oil cracking, as distinct from carburetting, is justifiable only at times of abnormal peaks of short duration, which would constitute in the sum only 2% of the total operating time. The arguments in favour of this allocation of functions are as follows :-

1. Even if coke were no more valuable than coal and even if one therm of coal gas were no more valuable than one therm of blue gas, under conditions of continuous operation it would pay to carbonise coal rather than to gasify coke.

(2) Capital charges on a water gas plant are only half those on carbonising equipment. Standby losses are low and the unit can be put on load in half an hour.

(3) Capital charges on an oil gas plant are insignificant.

Conditions may favour enrichment by the synthesis of methane, in preference to carburetting, at times when the load is within the capacity of the plant. At times of heavy load, however, enrichment should be effected by oil cracking, which increases both volume and therms instead of reducing them as does methane synthesis.

<u>Current practice and developments</u>. In the United States, it is general practice for water gas plants to be used at very high ratings, with steam and blast pressures which are possible only in closed generators. Under these conditions, units with an internal diameter of ten feet are guaranteed to deliver ten million cubic feet of gas daily, of which about 20% would probably be oil gas. In England on the contrary, it is general practice to operate water gas plants with a water seal and continuous, mechanical extraction of ash and clinker. The limitations of pressure in such plants restrict the gasification rate. The largest water-sealed generators in service in England are believed to be those at Billingham with an internal diameter of 13'6" at the grate. They produce eight million cubic feet daily, of a mixture of blue gas and nitrogen, of which about three quarters would be straight water gas. At Heysham, Lancs., however, there are several closed generators with an output of ten million cubic feet per day per unit.

In American plants, the high gasification rates employed are conducive to high fuel losses in the blow gas. At the moment of shutting off the secondary air to the carburettor, a pressure surge tends to occur in the generator, lifting the fuel bed and discharging as much as 200 lbs of fine fuel in the blow gas. Equipment is now available for maintaining a constant flow of air to the generator during the blow period, so eliminating that loss and nuisance.

Recent and projected developments in water gas manufacture include continuous production using a mixture of oxygen or enriched air and steam. At Traill, B.C., coke has been gasified in this way for several years. The standby plant for the Koppers gasification unit at Louisiana, Mo. is a Korpely producer arranged to utilise an oxygen-steam blast. A large oxygen plant being installed for Dupont is understood to be for the generation of water gas to be used in the synthesis of methanol. At the Dutch State Mines, an oxygen plant and Power Gas Corporation generators are being installed for the production of a water gas - nitrogen mixture for the synthesis of ammonia.

<u>Current Research in U.S.A.</u> In spite of the high gasification rates in American practice, it is generally believed that plant capacity is still limited by mechanical considerations and not by the velocity of the reaction. Current research work, therefore, is directed towards further reaction studies between carbon, steam, etc. and to methods of further increasing output. At the Institute of Gas Technology there are two small units for studying gasification reactions at atmospheric pressure and one for further investigations under pressure. Mechanical troubles with the pressure plant have not yet permitted much progress in this very interesting field of study. It has been determined, however, that oxygen is completely consumed within 1 mm of the inlet, when using finely divided pitch coke as the test fuel.

At Battelle, work is being done on the kinetics of the water gas reaction, as distinct from its mechanism which is being studied at I.G.T. Both programmes are sponsored by the American Gas Association. The producer at Battelle is of ½ ft. square cross section and four feet high. It is designed to operate at 100 lbs. pressure. By sampling gases from different levels it has been found that at the top of the fuel bed there is a reversion of carbon monoxide and hydrogen to carbon and steam. The steam decomposition percentage is reduced in this way from perhaps 90 to 80. It has been discovered, also, that there is an equilibrium between the nitrogen in the blow and that held in some way on the surface of the carbon. The facts are not explicable by the mere adsorption of nitrogen. The first gas made from the steam blast may contain as much as 5% of nitrogen. If this phenomenon be ignored and if nitrogen be added to the steam as a tracer, then the steam decomposition so calculated reaches the absurd figure of 120%.

In the work so far done at the Institute on the water gas reaction, nitrogen has been used as a tracer and corrections have been made by running blank tests with steam free from nitrogen. In future work, argon is to be substituted for nitrogen.

The <u>Gas Research Board</u> in England has just installed a small, stock model, water gas plant intended for an interesting investigation into the further possibilities of waste heat utilisation. The investigation is inspired by two possible future developments in the industry. The consideration of minor importance is the likelihood of a fuel efficiency campaign arising which would result in power being generated from high pressure steam, and exhaust steam being used for water gas generation and other process purposes. In such circumstances there would be no further use for the steam now raised at moderate pressure in water gas plants, and consumed in those plants with little or no production of mechanical power. The main incentive, however, is the elimination of the great objection to the enrichment of water gas by the synthesis of methane, viz the evolution of reaction heat equivalent to about one quarter of the heat value of the water gas converted. At present that heat would be a total loss in plants which are already fully supplied with waste heat.

Part of the blow gas from this plant is to be used for preheating air and steam to 600°C in a tubular heater with internal fins. The hot end of the heater, which will be subjected to a temperature of about 1,000°C, is made of cast nickel-chromium steel. The remaining blow gas and the hot, enriched water gas (if methane synthesis were to be practised) would be available for steam raising. The preheating of the blast and blow will have incidental advantages as follows :-

(1) The consumption of coke is expected to be reduced by 10%.

(2) The capacity of the unit is expected to be increased by 10%.

(3) Production will be more nearly continuous, because of the extension of the make period and shortening of the blow period.

(4) Because the air will enter the generator at a temperature above the ignition temperature of the fuel, the possibility arises of dispensing with the down run and so simplifying controls.

Elsewhere in this report, reference is made to laboratory work in progress on the oxidation of coal or carbon with steam or carbon dioxide. Although this fundamental work has a bearing on water gas production, it is included for convenience under oxidation studies.

LURGI HIGH PRESSURE GASIFICATION PROCESS. +

There is little interest in the Lurgi process in England, because the highly reactive coals to which it is most applicable are not available. It has been established in Germany that the capacity of Lurgi generators and the quality of the gas made very greatly with the rank of the coal used, as indicated by the following approximate but typical figures :-

Coal.	Capacity of Unit thousands of cubic feet per hour.	Calorific Value of Gas B.T.U. per cub. ft.
German brown coal,	83	480 maximum
Bituminous coal,	65	420 "
Coke,	36	380 "

+ See B.I.O.S. Report No. 521.

However, some work on a Lurgi unit is planned by the Gas Research Board to determine, amongst other things, whether its performance on coals of high rank may be improved.

There is greater interest in the subject in the United States, though even in that country the process is not regarded as one with early prospects of adoption. The only active plant is a very small unit at the Pittsburgh station of the Bureau of Mines. The Bureau is anxious to instal a larger generator at the Louisiana, Mo. Station, but the project has not yet received official sanction. A proposal has been made that the process shall be studied on a large pilot plant scale at Seattle, with funds contributed partly by the U.S. Treasury and partly by the State of Washington. It is believed that the Federal Government has not acceded to the request for financial assistance. The purpose of the project is to demonstrate that gas can be manufactured from Washington coals, in quantities and at a price which would make it unnecessary to import natural gas by pipe line from Canada.

The Bureau of Mines has a small Lurgi plant at Pittsburgh which was installed and is being operated at the cost of the Southern Natural Gas Co. The main business of that Company is the distribution and sale of natural gas from Texas, but it also has coal properties, probably of Texas lignite. Its work on the Lurgi process is inspired by the knowledge that some day it will become necessary to supplement natural gas with a manufactured product.

The plant is of one square foot internal area and is about 13'6" high. It is divided by flanges, valves and the grate into three sections of equal height, which serve as charging pouch, generator and discharge pouch respectively. The stainless steel liner of the generator is separated by an air space about 1/2" wide from the surrounding water jacket. The plant has been designed for a pressure of twenty atmospheres and all work has been done at that pressure. Oxygen and steam are admitted as usual through the grate.

Anthracite has been tested with some measure of success, producing a gas with a calorific value of 400 B.T.U. per cubic foot on a carbon dioxide - free basis. Very hard clinker was formed, leading to difficulty in discharging and to destructive channelling of oxygen through the fuel bed. Coking coal could be used after destruction of agglutinating properties by low temperature oxidation. The preferred fuel in these tests is a low temperature coke made at 600°C in a small intermittent version of the Disco retort. The feed to the retort is a pre-oxidised coking coal and for the Lurgi unit the coke is screened between 3/4" and 1/4". The gas made from this fuel is of 420 B.T.U. quality. The ash is reasonably free from unconsumed fuel, and the content of hard clinker is not high. It has been found possible to operate the plant on low temperature coke at a gasification rate of 5,000 cubic feet of crude gas per hour.

Because of the exaggerated wall effect in a generator of such small diameter, and because also of its low height, the value of the work done is somewhat limited. It has been shown that after suitable treatment high rank coals may be successfully gasified, at rates previously thought unlikely to be achieved. The plant, however, is not capable of evaluating the efficiency of gasification to be expected in a commercial unit, the maximum calorific value of the gas, the mechanical behaviour of the fuel and other important factors.

To repair these omissions, the Bureau desires to instal a Lurgi generator five feet in diameter at the Louisiana, Mo. demonstration station. The plant would operate at thirty atmospheres, gasifying non-coking, sub-bituminous coal from Wyoming or elsewhere, and would produce nearly two million cubic feet of scrubbed gas daily. In addition to providing fundamental information not obtainable from the Pittsburgh unit, it would serve as a standby for other gas plants at Louisiana and would afford an opportunity for work on the synthesis of oil from Lurgi gas. It would enable a long range study to be made of what is regarded as the most likely field of application for the process under American conditions. In the opinion of the Bureau, California's gas requirements are not likely to be met for many more years by pipe line transport from Texas. The most obvious source of supplementary supplies at that time will be Colorado and other western coals which would be gasified on the site. This service would seem eminently favourable to the adoption of the Lurgi process, which effects complete gasification of the coal, producing a gas of good calorific value at a pressure suitable for pipe line transmission.

Research and development. The feature of the Lurgi process which offers most scope for further research and ultimate improvement is the low rate of gasification or methane formation, whichever is the controlling factor. Although pressures are twenty times as high in the Lurgi generator as they are in a water gas plant, and although the gas making process is continuous instead of intermittent, the outputs from plants of similar size are substantially the same. The Bureau of Mines hopes to improve the rate of gasification in the Lurgi process by operating with a higher temperature at the base. It is not intended to slag the ash, but to operate under conditions producing clinker as hard as can be comfortably handled by clinker crushing grates of the Lymm Rambush type. The fuel bed will be deepened to suit the higher bottom temperature, so that the zone in which temperatures are favourable to methane formation will not be reduced in volume.

The improvements which Hydrocarbon Research has suggested for the Lurgi generators planned for installation in South Africa are mainly in matters of detail. It is hoped to improve the thermal efficiency of the plant by better utilisation of the waste heat available.

Elsewhere, it has been suggested that the Lurgi process might be improved by operating generators under slagging conditions or by fluidising the fuel bed. Neither proposal seems attractive on a superficial appraisal. The former would be difficult to apply for mechanical reasons; the latter is open to objection on the score that uniformity of temperature in the fuel bed is contrary to the principles of the process. The temperatures needed for rapid gasification of carbon and for effective production of methane are entirely distinct. In theory, the two reactions could be carried out in a generator with two separate, fluidised beds. This development, however, would add materially to the cost of the plant, which, considered in relation to the low gasification rate, is already undesirably high.

At the Carnegie Institute an interesting investigation is in progress to determine the source of the methane generated in the Lurgi process, that is to say, to decide whether it is synthesised from hydrogen and carbon monoxide (in accordance with the belief of German and some other authorities) or whether it is the product of a reaction between hydrogen and coal (as it is considered to be by British authorities). Isotope carbon is to be used in the experiments.

<u>The application of the Lurgi process to Victorian brown coal</u> was discussed with a number of authorities on the subject in U.S.A. and England. Because all commercial Lurgi high pressure gasification plants are in the Soviet controlled area of Europe, no opportunity offered for seeking the desired information at its source. In the period which followed immediately upon the armistice, many British and American technologists had access to some German installations. It was from them, mainly, that opinions were canvassed as to the advantages and disadvantages of the Lurgi process and the possibility of better processes being, or becoming, available. Other authorities who had made a close study

of the Lurgi or alternative processes were consulted also.

Of about a score of opinions obtained from various organisations or individuals, more than half were unequivocally in favour of the Lurgi process being adopted in Victoria. The reason given for this preference was that there is no satisfactory alternative process for producing from brown coal a gas of sufficiently high calorific value for general distribution. Of those who subscribed to this opinion, all had considered and rejected fluidised gasification. Some merely though the fluid bed process insufficiently developed; others frankly entertained no hope of the fluidised process ultimately proving superior to the Lurgi; others again thought that the gas produced from a fluid bed generator would be of too low calorific value without enrichment, which for various reasons could not be recommended. They advised against the enrichment of water gas, or its equivalent from a fluid bed plant, by synthesis of methane because of the heat loss in the process and the high cost of the preliminary purification of the gas. In some quarters, the indifferent calorific value of Lurgi gas was regarded as an objection though the process itself was otherwise approved. Enrichment with methane by synthesis or by the G.R.B. process was recommended in those two cases.

Some authorities rejected the Lurgi process on the score of the high capital cost of the plant, the use of expensive oxygen, or the indifferent calorific value of the gas made. They recommended instead low temperature carbonisation by the fluidised technique, by the Lurgi spulgas method or without naming any specific process. In each case the char was to be consumed under power station boilers. It appeared that the difficulties associated with the handling and storage of a highly reactive coke, made in a base load plant and intended as fuel for boilers meeting a fluctuating load, had not been adequately considered. Nor had it been possible to assess the economic implications of carbonising a relatively expensive dried brown coal when selling the residue in free competition with cheap raw brown coal.

Because lignite and lignite briquettes produced in U.S.A. tend to decrepitate when carbonised, one authority consulted was doubtful whether the Lurgi process could be applied to Victorian briquettes, while another was convinced that it could not.

The remaining gentlemen consulted had full confidence in the Lurgi method of complete gasification, but were non-committal in respect of its application to Victorian brown coal. They declined to express a clear opinion, either because of their unfamiliarity with the coal to be used or because they doubted the advisability of trying to distribute 450 B.T.U. gas in Melbourne.

The point was made more than once that, in so far as the technique of gasification is being constantly improved, it is useless and illogical to defer decisions so that a promising but undeveloped process may receive consideration at a later date. At that later time, there is likely to be another equally promising process, again not quite developed to the point of being ready for commercial application. Deferment, therefore, never solves the problem of finding the perfect process; it merely adds urgency to the need for a solution.

A general review of the advice given and of the circumstances in which it was offered, discloses an overwhelming preponderance of opinion in favour of the Lurgi complete gasification process. This conclusion is subject to one reservation only, viz. that the authorities consulted had little or no knowledge of the Texaco process and were unable, therefore, to consider it as a possible alternative to the Lurgi.

TEXACO PROCESS:

Texaco Development Corporation has been developing a complete gasification process for two or three years, and is reported to have spent two million dollars on the investigation in 1948 alone. The process was originally an adoption of the fluid bed technique, but the technique was abandoned some considerable time ago as not applicable to gasification. The process has this much in common with the Lurgi that it uses oxygen and steam under pressure and can produce a gas of about 450 B.T.U. calorific value. In other respects it is said to differ fundamentally from the Lurgi. Little more is known of the process because of the secrecy in which Texaco has been developing it. It is believed, however, that it is a two stage process in which carbonisation and gasification are effected separately, that the ash is slagged and that it has been developed to the stage of producing about one million cubic feet of gas daily at Montebello, Cal.

It is stated by the Company that the process is applicable to all types of coal, that it is indifferent to the size of the coal feed and that moisture contents up to 25% can be tolerated. The rate of gasification is very much higher than in a Lurgi generator and the cost of the plant is reduced to a corresponding extent. The Company is now ready to recommend and license the process, and to issue the data necessary for the design of the plant. Any organisation genuinely interested in applying the process commercially may, by applying formally to the Company, obtain the information needed for the confirmation of claims made for it.

The G.R.B. PROCESS.

The Gas Research Board has been working on a process for the production of gas of high calorific value by the substantially complete gasification of coal. Development is not being pressed intensively, because it is realised that there is no prospect of the early application of the process.

The G.R.B.process[+] consists essentially in gasifying coal in an atmosphere of hydrogen under pressure, under which conditions, methane is produced extensively. A temperature of 500°C is needed to initiate the reaction which is highly exothermic. In a static bed, therefore, the temperature tends to rise rapidly until the charge becomes unreactive by reason of high

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Communication G.R.B.45. Report of the Director for the Year, 1948.

temperature coking. In an autoclave intermittently charged and discharged, there are normally, therefore, three successive zones in the charge, viz.

(a) A zone in which only coke remains and in which the hydrogen is merely preheated to reaction temperature.

(b) An intermediate zone in which the reaction proceeds at temperatures progressively reduced from 1200°C approximately at one boundary of the zone to about 500°C at the other.

(c) A final zone in which the coal has not yet reached reaction temperature and in which it is being preheated by the hot gases leaving the reaction zone.

In a typical test with a fuel bed a foot in diameter and 9 feet deep, and with hydrogen supplied at 50 atmospheres pressure, the whole charge is exhausted in about 35 minutes. The average calorific value of the gas made is about 450 B.T.U. per cubic foot. It has been calculated that if a plant of this type could be charged and discharged continuously, the rate of reaction would be equivalent to the consumption of a 6" column of coal per minute, A coking coal moving uniformly at that rate would not agglomerate sufficiently to cause trouble, and it would be possible in theory, therefore, to operate the process continuously. Only part of the coal would be gasified of course.

It has been found possible to exercise sufficient control over the temperature by carrying out the reaction in a fluidised bed 4" in diameter by 10" in depth. Purified and preheated hydrogen is admitted at a pressure of 25 atmospheres and a temperature of 800°C. A catalyst in the form of carbonate of soda is added to the charge, which is usually Coalite. By adding methane to the hydrogen supplied to the reactor, conditions may be simulated for an upper section, 10" high, of a fluid bed exceeding 10" in depth. With a range of such mixtures, results may be predicted for a bed of maximum effective thickness. At 25 atmospheres pressure, the maximum concentration of methane in the product gas had been found in this manner to be 35.8%; at 50 atmospheres, it is 40%. Provided that sufficient hydrogen is available, the whole of the coal may be converted to methane in the fluid bed reactor. If sufficient coke were to be withdrawn from the reactor to provide the hydrogen needed for the gasification of the rest of the charge, the mixed gas would have a calorific value of 600 B.T.U. per cubic foot and the total yield of gas would be about 220 therms per ton. Less favourable results have been given by other fuels.

An interesting suggestion made for the further development of the C.R.B. process is to use the Lurgi gas at generator pressure as the source of hydrogen. It differs from the Lurgi process itself - in which some methane is formed, according to one school of thought, by contact between hydrogen and coal - in that a second stage of methane formation would be added after the removal of carbon dioxide and excess steam. In the absence of those two gases, it is believed that a higher equilibrium content of methane would be possible with a welcome improvement in the calorific value of the gas. The Board's work indicates that the second stage of methane production would need to be carried out in a fluid bed. The whole process, therefore, would call for the adoption of the fluidised technique.

It is conceivable that the same end might be achieved by circulating some scrubbed Lurgi gas through a hypersorption unit. The residual gas, high in hydrogen an low in methane, would be recycled to the generator; the overhead concentrate would be returned to the final gas stream.

UNDERGROUND GASIFICATION:

The Bureau of Mines is investigating under-ground gasification in co-operation with the Alabama Power Company, with a view to producing directly from the seam a gas which may be used as a fuel for boilers or gas turbines. For this limited purpose, it is sufficient to produce a gas which is rich enough to burn. The site of the operations is Gorgas, Ala. where the Company has two steam power stations adjacent to a colliery and a river. The coal seam being gasified is under virgin, wooded country and there is no town of significant size in the vicinity.

The results of the first experiment in 1947+ were encouraging in that they indicated satisfactory gasification. Certain conditions, however, had proved unfavourable. In particular, the thin cover above the seam had permitted leakage of gas through fissures in the ground. Provisions made for observing conditions in the fire zone were of little value and actually undesirable. Accordingly a second experiment has now been commenced, taking advantage of the experience gained in the first.

Two tunnels ten feet wide have been driven in a coal seam about four feet thick, with a barrier twenty feet wide between them. The overburden varies in thickness from fifty to two hundred feet. The twin tunnels are 1,241 feet long and one has been extended for 300 feet. Brick stoppings have been erected 141 feet from the portals and at intervals usually of 300 feet thereafter, the tunnels are connected by cross cuts. Each cross cut and the heading are pierced by a bore hole driven from the surface. The first and fifth boreholes are 18" in diameter and unlined; the other three are 28" in diameter, cased and lined. All boreholes are connected by valved pipes to both a 20" air manifold and to a waste gas burner. The method of operation planned is to ignite the seam at the heading, blow air down at that point and withdraw gas from No.4 borehole. When the coal in the vicinity is satisfactorily consumed, operations will be transferred to Nos.4 and 3 boreholes and eventually in this manner to the stoppings. The flow of air and gas can be reversed. It is not planned to use steam

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+ U.S. Bureau of Mines, R.I. 4164.

and/or oxygen.

Gasification was commenced in March, 1949. At the time of the last advice on the project, encouraging indications had been reported regarding the completeness of gasification. Collapses have occurred in the tunnel, however, dur to the width prescribed by state mining laws being much greater than was desired for the experiment. It was proposed to fill the cavities with sand or other material and to resume operations.

The Union Carbide Company has been working on the underground gasification of coal at Charlestown, W.Va. It is understood that horizontal bore holes are drilled for air supply and gas withdrawal, and that it has been found possible to drill a hole 30" or 36" in diameter, 700 feet long, in a seam of coal four feet thick.

It is in Russia that most application is being made of under ground gasification. However, little or nothing is known to the outside world of recent experience and developments within the Soviet Union.

One preliminary experiment has been made in France, and investigations are understood to be in progress in French Morocco. Interest in the subject is very active in Belgium, probably because of the steeply inclined seams in that country which are difficult to mine by normal methods, and which may be, perhaps, more easily exploited by gasification in situ. One large scale test was carried out in 1948, and a second is now in progress. French authorities are believed to be collaborating in, and contributing towards, the present experiment.

Some work has been done in Italy on the underground gasification of lignite.

MISCELLANEOUS.

A slagging producer is being developed at the Anthracite Institute for the utilisation of anthracite silt, which is a fine material accumulated in stream beds in admixture with clay, cinders and other refuse. It contains 25% of ash with a fusion temperature of 2,000° F.

The producer is 18" in internal diameter and about 8 feet high. It is lined with firebrick, finished with carbofrax on the inside and enclosed in a water jacket. It is supplied with preheated air and steam. It has no grate and is fired with extruded briquettes made at the station. At the time of the last information, little work had been done on the producer. An earlier and smaller unit had gasified 120 lbs of fuel per hour per square foot of cross section and had produced a gas of 135 B.T.U. quality.

The extruded briquettes were of poor mechanical strength. Their likely break-down in the fire would probably result in a high resistance to the flow of gas through the producer and a high loss of combustible material in the ash or slag.

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LARGE SCALE OXYGEN PRODUCTION.

Industrial Applications.

Since the war, there have been widespread interest and considerable activity in the manufacture and utilisation of oxygen of "medium" or 90-98% purity, as distinct from the "high" purity oxygen, containing less than ½% of inerts, which is needed for welding and other purposes. The uses to which medium purity oxygen is being put, or for which it is contemplated, include the manufacture of steel, production of nitric acid and other chemicals, roasting of sulphide ores and gasification of coal for towns' supply, or for synthesis gas to be used in the production of ammonia, methanol or oil. In the steel industry, the demand is largely for air enriched to about 30% of oxygen content, although for certain applications oxygen of 90% or higher quality can be used. In the gasification of coal, particularly for oil synthesis gas, it is desired to reduce the content of inerts as far as is possible without incurring disproportionately increased charges. For these reasons, the plants installed or now being erected are designed to give a product of about 95% quality. At higher concentrations, involving the separation of argon, costs are increased by reason of the small difference between the vapour pressures of oxygen and argon respectively.

The development of medium purity oxygen production has been most marked in North America,[*] where at least seven plants have been, or are being, installed with capacities expressed in terms of hundreds of tons per day. The largest of these plants is designed to separate 2,000 tons of oxygen daily for the production of oil synthesis gas, and is due to be commissioned early next year at Brownsville, Tex. At Johnstown, Pa., Bethlehem Steel has erected in collaboration with Koppers and Air Reduction a 150 ton per day unit which has been used for enriching blast furnace air. Early reports indicated that the furnace operation was satisfactory but that minor adjustments were needed in the oxygen plant. Other authorities consider that oxygen could be used economically in many open hearth applications with a substantial increase in production per furnace, and in the basic Bessemer process with advantages in output and the range of raw material suitable for charging. As an interesting by-product of oxygen manufacture, the Ford Motor Company at River Rouge is using liquid nitrogen for the shrinking of cylinder liners and valve seats into position. In Europe, argon produced in the separation of high purity oxygen is extensively used as a blanketing medium in the welding of aluminium.

FUNDAMENTAL PRINCIPLES.

The wide publicity which has been given to these developments in the last few years has tended to convey

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[*] Downs C. R., Impact of Tonnage Oxygen on American Chemical Industry.
Chemical Engineering, Vol.55, No.8, P.113,

For information in the production of oxygen in Germany,
See B.I.O.S. Reports 199 and 591.

the impression that there has been a revolutionary, rather than evolutionary change in the technique of oxygen separation. As aptly stated by one authority, "It has been difficult to see the subject for the searchlights". In actual fact, most of the numerous processes now available for the separation of medium purity oxygen in large quantities still use the fundamental principles developed many years ago by Linde, Frankl, and Claude. Recent technical innovations have been in the nature of refinements and the resulting improvements in performance of fractional value and significance. These improvements should not be under-rated; on the other hand, they should not be regarded as constituting fundamentally new processes.

The broad principles of all processes for the separation of oxygen from air comprise :-

(1) The production of cold by expanding compressed air (or nitrogen) through a throttle valve (Linde) or engine (Claude) or both.

(2) Heat exchange between the incoming air and the outgoing oxygen and nitrogen.

(3) Purification of the air to remove water vapour and carbon dioxide.

(4) Fractionation of the liquid air resulting from (1) and (2).

Linde's great contribution to the art, in 1906, was the double fractionating column, with the condenser of one column serving as the reboiler of the other. Air is partially fractionated under pressure in the lower column providing a liquid of about 40% oxygen concentration and liquefied nitrogen which, after expansion, can be used as reflux in the upper low pressure column. In this way Linde greatly reduced the escape of oxygen in the effluent nitrogen and to a corresponding extent improved the efficiency of the process. In 1926, Frankl introduced alternate cold accumulators as substitutes for the continuous exchangers previously used. The improved heat transfer efficiency, so achieved, resulted in a substantial reduction in costs. At the same time, pressures were reduced because of the lower frictional resistance to the flow of air, and impurities were removed in part by alternate freezing out of the air and subsequent re-sublimation in the product oxygen or nitrogen. The purification incidentally effected in this way made it possible for the first time to introduce some low pressure air into the upper column, with an improvement in thermodynamic efficiency.

At the temperatures obtaining at the cold end of a Frankl accumulator (or an exchanger) the specific heat of the incoming compressed air is higher than that of the outgoing low pressure oxygen and nitrogen. Therefore, in the absence of any compensating factor, the temperature difference at the cold end must be greater than it is at the warm end. To transfer the desired amount of heat in equipment of economical size, it is necessary to maintain a temperature difference of 5°F or 10°F at the warm end, corresponding to a difference of about 20°F at the cold end. At the lower temperature, the product oxygen and nitrogen become saturated with smaller quantities of water vapour and carbon dioxide than are carried by, and deposited from, the incoming air. The exchangers or accumulators cannot, therefore, completely purify all the air which is to be condensed and fractionated. In Linde Frankl plants this difficulty was overcome by separately treating a small portion of the air, amounting to five or ten per cent of the whole according to the size of the unit. This stream of air was cleaned chemically, compressed to 200 atmospheres, cooled in an ammonia refrigerator and expanded to the temperature and pressure of the lower column. In this manner, the carrying capacity of the outgoing gases for carbon dioxide and water vapour was improved in two ways, viz. by an increase in volume and by a slightly higher temperature at the cold end of the accumulator. The Linde Frankl process, so applie was representative of the state of the art in the thirties.

RECENT PROCESS DEVELOPMENTS:

During and since the war, ideas have been advanced and developed for alternative forms of heat exchange equipment. The first of them was the Collins reversing exchanger which had three channels, in good thermal contact, for oxygen, nitrogen and air respectively. The nitrogen and air channels were interchanged, at intervals, without reversal of flow, while the oxygen stream remainded unaffected. The idea was applied in large numbers of mobile oxygen units constructed for war purposes. The plant had the advantage shared with other units using exchangers instead of accumulators, of delivering an oxygen uncontaminated with carbon dioxide and water. The equipment was cumbersome however; it was out of balance for the clean-up of the exchangers; and because no auxiliary purification plant was provided, it operated only for short periods of time before becoming choked. The exchanger is now obsolete.

More recently, heat exchangers have been devised with modifications permitting complete purification to be effected in the exchanger and so obviating any need for a separate high-pressure, compression system. The principles upon which these various modifications are based are illustrated in figures 5 to 13. Processes have been devised, also, to achieve in other ways what was previously effected in the Linde double column. It is in these respects that the new processes differ in detail, rather than in principle, from the classical Linde Frankl.

Trumpler of the M. W. Kellogg Co. has devised a simple method of providing additional capacity for re-subliming impurities. In what may be regarded as a Collins exchanger, a fourth pass is provided in which cooled air is returned in the same direction as the product oxygen and nitrogen. The effective temperature of those gases is thereby raised, and their vapour carrying capacity increased, to the necessary degree. Because increased carrying capacity is needed only at the cold end of the exchanger, the additional or Trumpler pass is only half the length of the others. The same effect can be achieved by circulating through the Trumpler pass a stream of nitrogen suitably cooled in each circulation. The whole principle may be applied to separate exchangers for oxygen and nitrogen - in which case the oxygen and air channels would be periodically interchanged - and to accumulators of the Frankl type as well.

Frankl, himself, proposed to achieve the same objective by tapping a small stream of air from the middle of the accumulator and completing the cooling of this stream by external means. In the process devised by the Arthur D. Little Co., two exchangers are connected in series and the compressed air is cooled by external means before entering the second exchanger.

Of those processes which do not use the classical Linde double column, the Esscol process of the British Oxygen Company and that of the Elliott Company have much in common, in that they use nitrogen as the refrigerating medium independently of the air which is to be liquefied and fractionated. This separation of functions adds to the flexibility of the plant and simplifies the clean-up problem. Because the air enters the single fractionating column at atmospheric pressure, and at a correspondingly lower temperature than in those processes which use air for refrigeration at about 90 lb. pressure, the residual impurities carried into the plant are greatly reduced. It is claimed for the Elliott unit that instead of requiring to be shut down at intervals of two or three months, as does the Linde Frankl plant, for thawing out accumulated ice etc., de-riming will hardly be necessary within the useful life of the plant.

The feature of the Oxyton plant offered by L'Air Liquide, which mainly serves to distinguish this process from others, is the three stage fractionation effected in a double column followed by an independent single column.

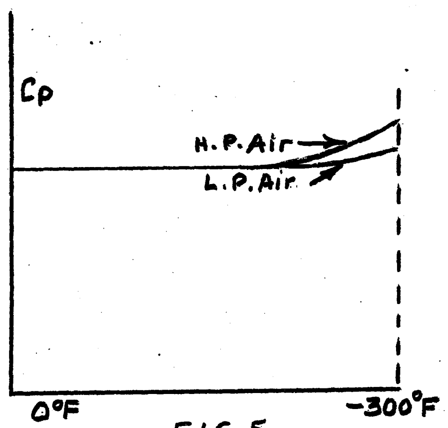


FIG 5.
SPECIFIC HEAT OF AIR.

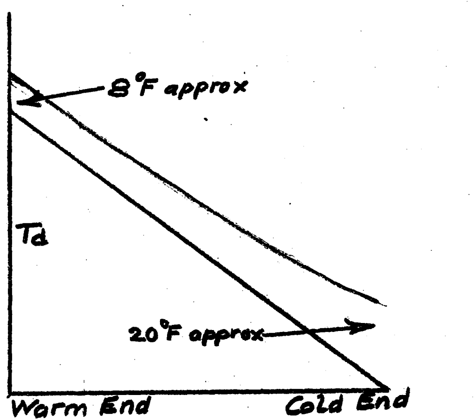


FIG 6.
TEMPERATURE GRADIENT
IN UNCOMPENSATED EXCHANGER

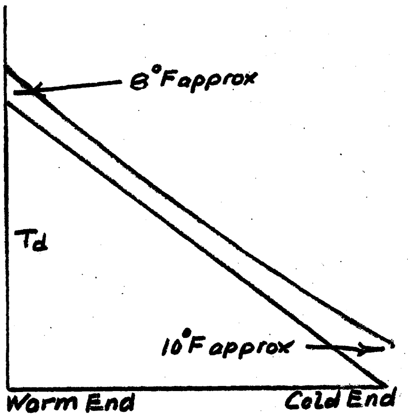


FIG 7.
TEMPERATURE GRADIENT
IN LINDE FRANKL
COMPENSATED EXCHANGER

TRUMPLER COMPENSATION.

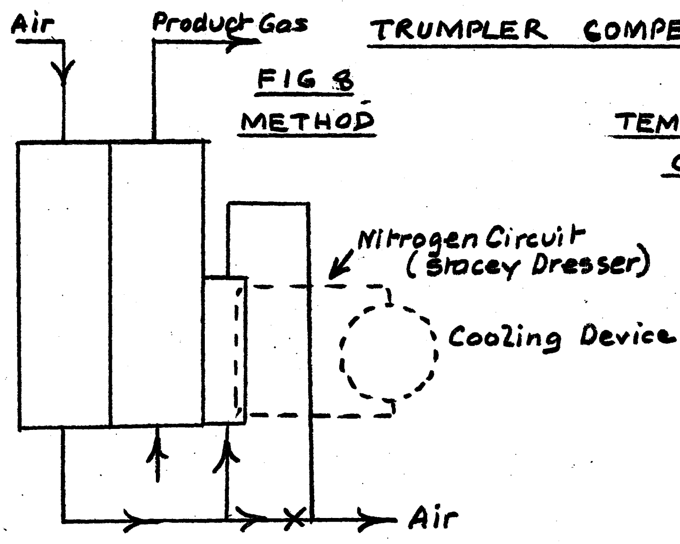
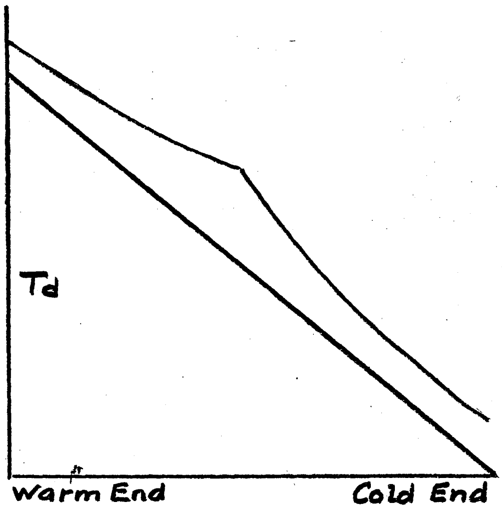


FIG 8
METHOD

FIG 9.
TEMPERATURE
GRADIENT



ARTHUR D. LITTLE COMPENSATION.

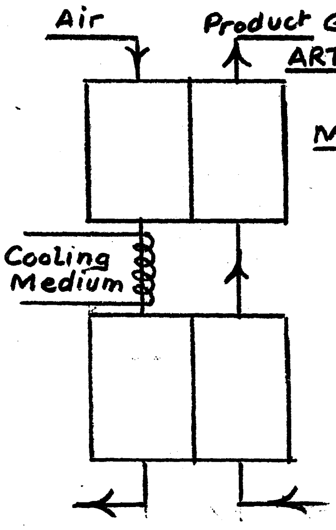
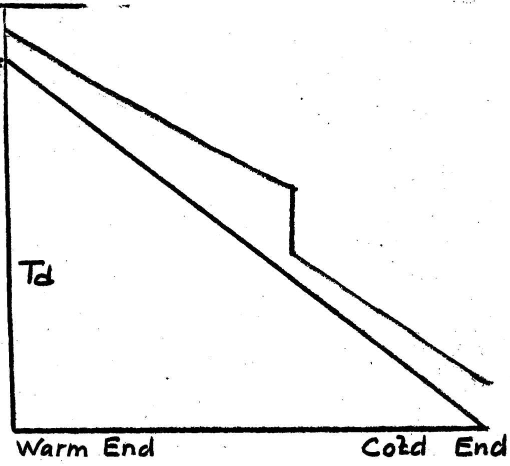


FIG 10
METHOD

FIG 11.
TEMPERATURE
GRADIENT



FRANKL COMPENSATION

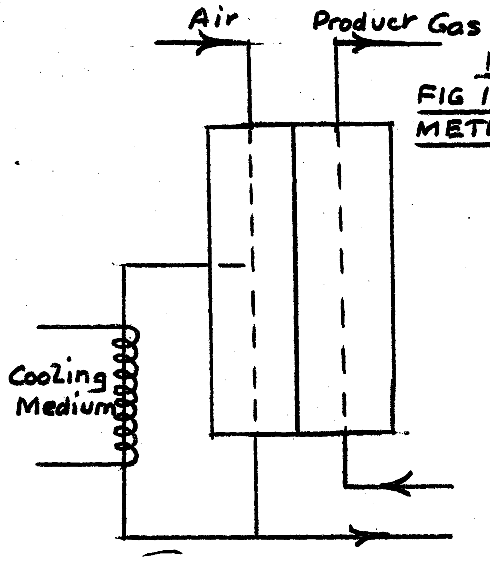
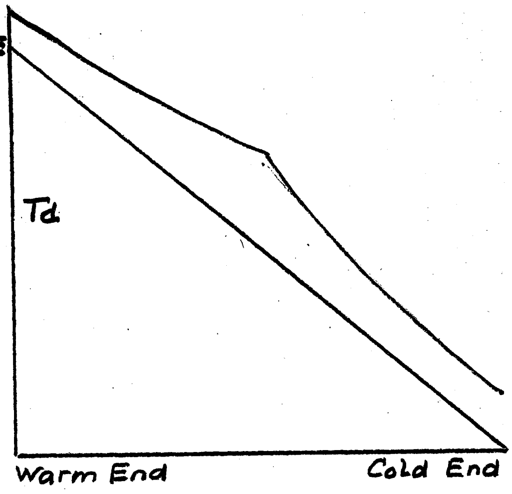


FIG 12
METHOD

FIG 13.
TEMPERATURE
GRADIENT.



This arrangement is said to reduce the height of the fractionating equipment and its capital cost. The Air Reduction Company's installation at the Bethlehem steel works also utilises three successive stages of fractionation,

It is understood that in the medium purity process adopted by Linde Air Products Company, the licensee of the Linde Frankl patents in U.S.A., the high pressure circuit has been eliminated but portion of the air is still cleaned chemically. Linde has one large central plant at East Chicago and one in course of construction for the Dupont Company.

Little was learned of the Air Products process incorporated in a 400 ton per day plant being built for Weirton Steel. It has no high compression circuit.

Stacey Dresser plants comprise one in operation for McCarthy Chemicals and one nearing completion for Carthage Hydrocol at Brownsville, Tex.

L'Air Liquide is currently building two large units, one for the Dutch State Mines to produce ammonia synthesis gas, the other for International Nickel to be used in the roasting of Canadian ores.

Nothing is known of any large scale application of the Elliott, Kellogg, Little and Rescol processes.

ELIMINATION of ACETYLENE:

One refinement to which much thought has been given by manufacturers of oxygen plants is the removal of hydrocarbons, considered to be predominantly acetylene, which tend to accumulate in explosive proportions from the minute traces present in free air. Before the application of adsorptive methods of purification, it was common practice in Germany to construct long and expensive pipe lines to convey air to the separation plant. According to the direction of the wind, one or other of the intake pipes would be closed, so as to get the purest possible air into the plant. Accumulations of hydrocarbons tend to occur in the base of the low pressure column, and in spite of all precautions taken, explosives used to occur with such frequency as to justify the use of armour plating in some plants.

An early method of purging acetylene was to withdraw a slurry of solid carbon dioxide in liquid oxygen and acetylene at intervals and in quantities calculated to keep the concentration of hydrocarbons within safe limits. Another expedient is to withdraw the liquid oxygen and acetylene into an external vessel continuously evaporating most of the oxygen at the same time. The residue is not allowed to reach hazardous limits of concentration. At intervals, the vessel is disconnected from the supply line and the residue is flashed into the product, so allowing no opportunity for further concentration during the process of evaporation. In some processes, including recent versions of the Linde Frankl, the Oxyton and the Stacey Dresser, the liquid, oxygen-enriched air, proceeding from the H.P. to the L.P. (or I.P.) column is passed through a silica gel filter. In the Elliott plant, an adsorptive filter is installed in the air inlet to the single column used. The M. W. Kellogg Company has studied the elimination of acetylene by catalytic oxidation in the compressed air. It is understood that during the war, when many oxygen plants were required to operate for the longest possible period without cleaning, acetylene explosions were more frequent than previously. Since the adoption of silica gel filters in 1942, no explosion has occurred in any oxygen plant so equipped

COSTS:

Some manufacturers of oxygen separation plants do not sell the unit even when it is erected for the use of a single customer and on his works. They retain ownership, operate the plant and sell the oxygen instead. Other manufacturers are prepared to sell either the plant or the product. They may also contract for the operation and/or maintenance of the unit.

The main items of cost in the production of oxygen are capital charges and power consumption. In current practice, about 15 K.W. hours are needed for each 1000 cubic feet of 95% oxygen, though hopes are entertained in some quarters of reducing the power consumption to 12 K.W. hours. The following approximate estimates of cost are intended as a rough guide only. Capital charges are assessed at 5% for interest and 7½% for depreciation. Power is charged at 0.3d per unit, and for every 0.1d by which the actual rate exceeds 0.3d, the total cost should be increased by 1½d per thousand cubic feet. The plant is assumed to operate for 95% of the time, an important condition because for most processes the efficiency falls at reduced loads and satisfactory operation becomes impossible at less than 60% or 70% of full rating, according to the process. The total cost shown includes 5% for contingencies.

Capacity of unit, tons per day,	100:	200;	400;	1000:
Capital cost, dollars,	1500000,	1800000,	2600,000,	4000,000,
Total Production cost, :— dollars per ton.	8.80,	6.57,	5.22,	3.95,
Aust. d. per 1000 cub. ft. (at $ 3.2 per £1.)	27.5	20.5,	16.3,	12. 3,

So far as can be gathered by one without special knowledge and experience in the oxygen industry, there is little difference between the various processes offering in respect of costs, reliability in operation and incidental advantages or disadvantages. The largest single item of capital expense is in respect of heat transfer equipment. Because regenerators are normally smaller and cheaper than exchangers for the same duty, a preference might then be given at first thought to processes still using the Frankl system of heat exchange. However, such ingenuity has been applied by American firms in designing exchanger packing for cheap, mass production, that it would be rash for a layman to express an opinion on the subject. Some processes are advertised to be capable of producing a proportion of the oxygen (up to 20%) in 99.5% concentration, which would be a considerable advantage where welding oxygen is needed. However, an intensive study of the subject is needed for the proper assessment of these and other features of individual processes.

COMBUSTION and OXIDATION STUDIES.

The spreader stoker is the favoured mechanism for firing boilers evaporating up to 200,000 pounds of steam per hour. The elimination of dumping bars in favour of a slowly moving, reversed, travelling grate - an innovation introduced in the early years of the war - has further added to its popularity. The largest boiler yet built for firing with a spreader stoker is one of 300,000 lbs. per hour capacity. The limitation is determined by the length of the grate which can be covered with reasonable uniformity when using a cheap fuel containing much fine material. Spreaders can handle coal as fine as 0 - 3/8", but in such a case the fuel must contain a substantial amount of 3/8" material. The coarser particles are needed to cover the back of the grate; fines cannot be thrown that far and are deposited, instead, on the nearer section of the grate. About 50% of the fines, indeed, may be burned in suspension without reaching the grate at all. Because of the thin and, perhaps, somewhat uneven fuel bed, more excess air passes through the fire than is usual in pulverised coal fired boilers. The resulting greater loss of heat in the flue gas is outweighed by the reduced power consumption of auxiliaries.

Users of pulverised coal are much concerned at the difficulties experienced in the collection and disposal of fly ash. At one time it seemed that the slagging, well-type furnace would provide the answer to this problem, but wastage of tubes exposed to the molten slag proved so severe that this method of firing had to be abandoned. Increased pressure exerted by local authorities to reduce the emission of grit from boiler chimneys has further emphasised the problem. Mechanical and electrostatic precipitators installed in series, the former designed for peak load conditions, the latter to meet the average load only, have proved to be insufficiently effective. In recent installations, the Cottrells also have been designed for peak load service and it is hoped in this way to suppress the discharge of fly ash to about 1%.

It is almost as difficult to dispose of fly ash as it is to collect it. The material is difficult to wet, it does not compact, it runs freely and has little or no bearing value. It is useless, therefore, for aggregate, foundations or even back-fill. Present practice is to dump it at sea or on waste land, a method of disposal which involves transport charges without any credits. Some thought has been given to the treatment of fly ash so that it may be used as an aggregate in concrete, but it is understood that no encouraging progress has been made in the investigation. Fly ash remains an embarrassment both in its collection and in its disposal. It seems likely that the Babcock cyclone burner, to be mentioned later, will prove the best solution of the problem.

It is accepted practice for turbines not to be connected to a steam range supplying other machines as well, but for each to be coupled to one or more boilers serving that turbine alone. Now that boilers approach turbines in reliability and availability (95%) this practice is entirely justified. The improved performance of boilers in this respect is attributed to the elimination of much brickwork, and reduced maintenance on what remains, as a result of the use of water screen tubes. Boilers are commonly operated continuously for a year at a time and are then closed down voluntarily, to a pre-determined schedule, for repairs, replacements and insurance inspection. It is considered in some quarters that a good case may be made for extending the period between statutory inspections of modern boilers beyond the present limit of a year.

Impact pulverisers are favoured for grinding soft coals, ball and bowl machines for coals of medium hardness and ball mills for anthracite, slatey coal etc. When pulverising petroleum coke, which is used to only a small extent for steam raising, the mill is commonly swept with air preheated to about 600°F, so as to prevent agglomeration due to the traces of pitch left in the coke.

Because of the increased cost of coal, high ash fuels are commonly used in power stations, with further aggravation of the fly ash problem. In one plant, middlings from a washery constitute a considerable portion of the coal supply; in another, a "boney" coal of 37% ash content is being consumed.

A further consequence of the high price of coal is the tendency in recent power plant designs to strive for greater efficiencies by adopting high pressures and temperatures. At Twin Branch Station, east of Chicago, a plant has been operating since 1940 under the following conditions:-

	Original.	Reheat.
Pressure, lbs per sq.in.	2,300	400.
Temperature, °F,	950	915.

Additions now in course of installation will operate as follows :-

Pressure,	2300 lbs. per sq. in.
Temperatures, -	original 1050°F ,
"	reheat 1,000°F. Flue, 275°F
Efficiencies, -	boiler, 90%, overall, 37%
Evaporation, -	900,000 lbs. per hour.

Because of the low outlet temperature and the high sulphur content of the coal to be fired, the last section of the air heater is made of aluminium.

BABCOCK CYCLONE BURNER.

In its horizontal form, the cyclone burner[+] is a cylinder 8 feet in diameter, of the same length approximately, and inclined 5° from the horizontal. The steel shell is lined internally with chrome bricks, in which are embedded studded cooling tubes with water passing through them. The air for combustion is preheated to 400°F. The coal is crushed to pass a 1/4" screen and is admitted tangentially into the burner suspended in the primary air. Secondary air also is admitted tangentially and, as a result of the swirl, the furnace lining is covered and protected from erosion by a layer of slag. Large particles of coal thro

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[+] Transactions A.S.M.E. August, 1947. p.613.

out by centrifugal force adhere to the slag until they are consumed. Products of combustion issue through a throat 3' or 3'6" in diameter and then enter the boiler proper. Because of the high temperature at the point of coal entry, this part of the burner is subject to erosion and is fitted, therefore, with wear plates. For further protection, tertiary air amounting to 5 to 8% of the total supply is admitted centrally at the entry. This is said to be the only part of the burner requiring maintenance. The connection of the burner to the boiler calls for special shaping of screen tubes around the orifice.

Ten years experimental work was done on the burner before the first commercial unit was installed at Calumet Station, Chicago, in September 1944. In four years experience from that date till the time of the last advice, the burner had given complete satisfaction and no modification had been found necessary. Without special attention, it fires a boiler evaporating 180,000 lb. of steam per hour. It is shut down with the boiler at yearly intervals for inspection and maintenance.

The burner requires air at 30 to 40 inches of water gauge pressure, which is considerably higher than the air pressure in orthodox pulverised fuel installations. However, because the coal needs only coarse crushing, the total load due to auxiliaries is in favour of the cyclone burner. Its great advantage, however, is the removal of 85% of the ash in the coal in the form of a molten slag. At Calumet the slag is tapped intermittently from the burner in a thin trickle which is allowed to drop straight into water. The slag is thereby quenched and disintegrated into glassy particles like coarse sugar. This material may be used as an aggregate in making concrete. Of the other 15% of the ash, 5% is volatilised and 10% leaves the plant in the form of ash particles much less in quantity, therefore, than the fly ash normally discharged from pulverised fuel boilers. Because of the small quantity, that ash can be sufficiently removed by mechanical separators and electrostatic precipitation is regarded as unnecessary.

The cyclone burner has many incidental advantages. Because of the substantial elimination of ash in the burner itself, soot blowing is necessary only once per shift. Since the burner is installed in front of the boiler proper, and not under it, the total headroom required is considerably reduced. The high rate of heat release, viz 42,000 B.T.U. per cubic foot per hour, permits a reduction in the width of the boiler to such an extent that the ground space saved is greater than that occupied by the burner. A substantial saving is effected, therefore, in the capital cost of the whole equipment. If the burner could be operated satisfactorily in a vertical position, still greater economies in space and cost would be possible. An experimental, vertical unit has been installed at Calumet, but its performance is regarded as not yet good enough to justify commerical application.

Lighting up the burner is done simply and rapidly in a period of 30-60 seconds,

At the time of the last advice, Babcock and Wilcox had 51 cyclone burners on order, including eight 9 ft. diameter units for the Fisk Street, station. Four were required for each of two boilers designed to evaporate 750,000 lb. of steam per hour per boiler. In the opinion of many competent observers, the cyclone furnace is the most outstanding development for many years in the firing of large boilers.

F.R.S. VORTEX BURNER:

In the vortex burner, finely pulverised coal and air are admitted tangentially at the periphery of a shallow, circular, combustion chamber. The outlet for the products of combustion is at the centre and the flame is directed downwards. The diameter of the chamber and its outlet are so related to one another and to the rotational velocity of the gases that within the burner there is an equilibrium radius for each particle of coal. That radius is determined by the inward sweep of air or other gas and the outward centrifugal force due to the swirling motion in the cham. In theory, the coarsest particle should just fail to touch the containing wall of the burner, and no unconsumed particle should be able to escape through the central aperture. The relative radial motion between coal and air results in rapid combustion and a high rate of heat release.

The burner was first investigated at the Fuel Research Station about twenty years ago. Because of the persistent deposition of fuel within the chamber, that early work was suspended. It was established, however, that a phenomenal rate of heat release, viz. two million B.T.U. per hour per cubic foot, could be achieved. The idea has been revived recently because of new possible applications being envisaged. As already mentioned in this report, the Bureau of Mines is interested in applying the principle of the vortex burner to the production of synthesis gas from coal. At the Fuel Research Station, new interest has been aroused by the prospect of applying the burner to a coal fired gas turbine. Work has been resumed at the station in collaboration with B.C.U.R.A. The complementary investigation of gas turbines is being undertaken at the R.A.E. establishment, Farnborough.

The burner recently fabricated at the Fuel Research Station is 45" in internal diameter, 15" deep at the centre and 12" at the circumference. The top and bottom plates are of 18:8 alloy; the cylindrical side wall, in which the main air ports are formed, is of cement fondu. Air is admitted first to a narrow space between the top outer casing and the upper plate of the combustion chamber. Thereafter, it is conducted to a similar space below the lower bottom plate of the burner. Having thus cooled the top and bottom plates in turn, and having been itself preheated in the process, the air is divided into two streams. The larger volume is discharged to waste; it is intended ultimately to be mixed with the products of combustion, to cool them to a temperature at which they may be safely admitted to a gas turbine. The smaller stream of preheated air is conducted to a manifold surrounding the refractor wall of the burner and thence, through tangential ports already mentioned, into the combustion chamber itself.

Pulverised coal is admitted to the burner in streams of cold air, through six equally spaced stainless steel pipes. The pipes pass through the outer mild steel cover and are welded to the inner top plate of the burner. The coal and air enter tangentially so that they may join the main stream of preheated air with a minimum of shock.

At the time of the last advice on the subject, the burner had been operated for an hour approximately at 40% load - i.e. at 200 lb. of coal per hour - for the purpose of drying the brickwork and annealing the fabricated steel work. From the colour reactions of the paint applied, it was determined that the maximum temperature experienced by the alloy cover plates was 800°C at the edges. At the centre, the temperature was about 500°C.

LOCOMOTIVE DEVELOPMENT COMMITTEE'S BURNER.

The Locomotive Development Committee was formed by Bituminous Coal Research (representing colliery interests) and several American railroad companies to develop a gas turbine locomotive burning pulverised coal. The railroads prescribed that the locomotive must draw its fuel from normal stocks and bunkering facilities. Coal of low ash content could not be specially selected for the purpose; nor could the fuel be dried and pulverised in advance. The Committee, therefore, had to pioneer work on many problems apart from the obvious one of designing a vehicle to accommodate all the component parts of a gas turbine power plant. The operations which had to be carried out within the confined space of a locomotive were as follows :-

1. Drying and grinding coal to a fineness preferably greater than that desired for power station boilers.
2. Feeding and metering coal into a pressure vessel.
3. Burning the pulverised coal in a small combustion chamber.
4. Separating and disposing of the ash.

Early work at I.G.T. gave promise of leading to the development of a suitable method of pulverising coal, viz. by suspending it in a blast of steam or gas under pressure, suddenly releasing the pressure by expansion through a nozzle and directing a high velocity stream of gas and coal on to a target. As mentioned on page 62, the same device is used for part of the pulverisation effected in the I.G.T. gasification process. Because of its superficial simplicity and the absence of mechanical equipment otherwise necessary, the nozzle pulveriser seemed ideally suited to the locomotive project. The Committee adopted the method, therefore, and persevered with it for two or three years before ultimately discarding it as totally unsatisfactory. The reduction in size of the coal particles was found to be due not to disruption on release of pressure, but to impact and attrition entirely. The system did not permit a classification and re-circulation of oversize only, and no reasonable amount of re-cycling would reduce the + 100 fraction to the extent deemed essential. In the attempts made to pulverise to the fineness indicated by combustion tests to be necessary, the power consumption of the compressors increased so much as to become prohibitive - not because of cost which was relatively unimportant, but because of space considerations. Even the simplicity of the method proved illusory because for variable load operation, the nozzle had to be adjustable.

When the decision was made to abandon the nozzle method, manufacturers of pulverisers were invited to develop equipment for grinding coal under pressure. The problem was one which could not be solved by the use of impact pulverisers because of the greatly increased windage losses. Manufacturers of other types of mills were satisfied, however, that they could meet the Committee's requirements. A pulveriser is now on order from Babcock and Wilcox and is expected to give a product of which only 2% will fail to pass a 100 mesh screen.

Two devices investigated by the Committee for feeding coal into a pressure zone proved reasonably satisfactory. As metering devices, their accuracy was found to be affected by the moisture content of the coal. Now that pulverising is to be done under pressure, however, accurate metering is no longer necessary. Variation of coal supply with load will be automatic, for each is a direct function of the volume of air passing through the mill.

The Committee's first type of combustion chamber was based upon the design of aircraft jet combustors. They were made of 18:8 alloy, titanium stabilised, and were 6'6" long by 2' in diameter. Two were intended for each 4,000 H.P. locomotive. In spite of numerous variations in the arrangement of cooling air ports, none of these burners proved satisfactory. Scale and slag collected inside the burners, particularly at the fuel admission end. Largely as a result of that deposition, distortion was rapid. The best of the burners operated for only thirty hours intermittently followed by thirty hours of continuous service. The scale etc. were found to flake off the metal when the burner was allowed to cool, so introducing a possible operating hazard for the turbine.

The best combustion efficiency realised in metal burners was 90%. Objection was not taken to the 10% loss of fuel, but the resulting high carbon content of the ash could not be tolerated because of damage resulting from combustion in the ash separators. It has been found necessary on this account to reduce the carbon content of the ash below 20%.

The Committee is now developing a refractory burner of the same size, built up from silicon carbide tiles. Silicon carbide has been chosen for its heat resistance, low thermal expansion, good conductivity and therefore its excellent resistance to thermal shock. The tiles are insulated by air pockets between them and the outer, stainless steel casing of the burner. The casin itself is cooled externally by arranging for good contact with the secondary air. The burner is mounted vertically with the flame directed downwards. The secondary air is directed across the burner outlet so as to chill the slag dripping from the refractory cylinder. In this manner, nearly 50% of the ash is caused to separate in comparatively large particles of slag. Nearly all the remaining ash is larger than 325 mesh in size and is suitable for separation in multiple small cyclones. The temperature in the combustion zone is 3,400°F. The combustion efficiency is 98% and the heat release at full load is one million B.T.U. per hour per cubic foot.

A similar burner is being constructed for horizontal mounting. Aerotec cyclones about 18" high and 3" in diameter have been found satisfactory for service under the conditions expected to obtain in a gas turbine locomotive. They pass very little dust finer than 20 microns and most of the escaping dust is finer than 10 microns. Coarse particles are not separated efficiently because of interference effects in tubes of such small diameter. The 168 aerotec cleaners needed for the locomotive are to be installed between the burner and the turbine. They are made of stainless steel and their weight is estimated at 20,000 lbs.

No decision has been made regarding the method of storage and discharge of ash.

The temperature of the gas entering the turbine is to be adjusted by mixing hot gas from the burner with cool air. Control is to be exercised automatically by instruments. Over a range of loads which is determined by the performance of the compressor at low speeds, the gas temperature will be kept constant and the turbine speed varied to suit the load. At lighter loads, the gas inlet temperature also will be reduced. For starting the turbine, a 150 H.P. diesel generating set is provided. The power from that unit will be applied to one winding of the main generator designed to function as a motor winding. In this way, the turbine will be run up to a speed of 1,200 r.p.m., when oil fuel will be admitted to the burner. Under the combined power of turbine and diesel engine the set is accelerated to its idling speed of 2000 r.p.m., at which the power generated by the turbine just exceeds that absorbed by the compressor etc. The diesel set is then disconnected and the main plant further accelerated under its own power.

The Committee's programme was to have been completed in a period of five years, of which four have now elapsed. It seems most unlikely that the work still remaining to be done will be completed within a year.

Some information on the turbines which are proposed for the Committee's locomotives is given in a later section of this report.

COMBUSTION in SCOTCH BOILERS:

During the war, the Fuel Research Board was consulted on the suppression of smoke from shell-type marine boilers, and recommended the admission of secondary air in liberal quantities immediately after firing. A Scotch boiler was installed at the station for appropriate tests. It was found that by admitting secondary air over the fuel bed, in large quantities after firing and in small volumes thereafter, combustion could be made practically smokeless and efficiencies improved by from 10 to 20%. The technique was applied under sea-going conditions and under F.R.B. observation on a round trip to South Africa. The results indicated satisfactory suppression of smoke and a reduction in fuel consumption of about 5%. In view of the inexperience of the stokehold crew and other circumstances, these achievements were regarded as satisfactory.

Furnace fronts were designed for boilers operating in one case on forced draft, and in the other on natural draft. It was arranged that the auxiliary air door should open automatically when the firing door was opened but that the subsequent closing of the door should be a manual operation. Consideration was given to devices for shutting off the auxiliary air after a suitable time interval, but the need for flexibility precluded the use of such devices.

Instead, a coarse adjustment of the door setting was provided to suit different coals, and the fireman was instructed to close the supplementary door as soon as the faint smoke, emitted after firing, disappeared. A smoke observing device was installed for the fireman's convenience and assistance. As a result of the work done by the Board, about 800 furnace fronts for forced draft and approximately 400 for natural draft have been installed to date.

DOWN JET COMBUSTION.

The down jet furnace was first considered in England during world war 1 without any commercial application of the principle resulting. During the second world war, the idea was revived by the British Coal Utilisation Research Association and some work on the subject was also undertaken at the Battelle Institute in U.S.A.

The principle of down jet combustion is that the air supply to a fire shall be directed downwards onto the front exposed surface of the fuel bed. Where the fresh air meets the incandescent fuel combustion is intense and high temperatures result from the formation of carbon dioxide to the substantial exclusion of the monoxide. The front of the fire is kept free of ash, most of which is slagged and gravitates to the bottom of the fuel bed. The rest of the ash is carried into the interior of the fuel bed by the products of combustion and is there filtered out of the gas by freshly applied fuel. The hottest portion of the fire is, therefore, that exposed to the object to the heated, so ensuring rapid and efficient heating.

The composition of the combustion gases is not uniform across the fire. At the top there is a tendency towards incomplete combustion and at the bottom towards excess air. The gas outlet is a cylindrical, refractory lined passage designed for a velocity of 50 f.p.s., whereby sufficient mixing is provided to ensure completion of combustion with carbon dioxide contents of the order of

12% and temperatures of about 1,900°C. Because of the high temperature the unconsumed carbon in the clinker is quite low. For any one furnace, the combustion products are determined by the depth of penetration of the air jet and, therefore, by its velocity. Consequently, the down jet furnace is essentially a constant load furnace, which is rarely a disadvantage in application since most furnace loads are constant. The down jet may be operated intermittently to accommodate a change in load, but if it is to function continuously at a light rating, the jet must be changed.

Coke is the ideal fuel for down jet combustion because it will feed out of a hopper by gravity. Because of the high cost of the fuel, however, it is considered in England that the largest scale on which the principle may be applied is one of 200 lbs of coke per hour. Because coal does not feed evenly downwards by gravity, it must be supplied to the fire mechanically and therefore in a horizontal direction. The slag is moved forward also and must, therefore, be removed in a molten condition rather than as clinker. The need for a mechanical stoker and for the tapping of liquid slag means, in effect, that furnaces burning coal in this manner should be large enough to consume at least 1,000 lb of coal per hour.

Although down jet combustion has been developed mainly for high temperature furnaces, it has been applied with complete satisfaction to a low temperature operation, viz. the drying of foundry cores and moulds. One English company which previously used for this purpose conventional, manually operated coke stoves, is now converting completely to down jet furnaces. The advantage of the furnace in this service is that it requires no attention other than the filling of the hopper and removal of clinker at long intervals.

The principle has been applied experimentally to a household fire, using chimney draft to draw air down into the fire.

At Battelle, it is considered that the principle has prospects of application in kitchen ranges and in small boilers, but apparently not elsewhere. A combustion rate of 180 lb. of fuel per sq. ft. of grate area per hour has been attained at the Institute. Several domestic ranges equipped for down jet combustion are being tested under service conditions. Special heat resistant lids have had to be provided over the fuel compartment. Two jets of air are provided for combustion, one vertically downwards onto the fuel the other horizontally across its surface. Heating up from cold is said to be almost immediate, as fast, indeed, as over a gas burner. When the air is shut off, the fire goes cold with equal rapidity. Because of the intense heat, ashes are clinkered freely, but clinker needs to be removed only once per week. The range has the disadvantage, of course, of needing a forced draft.

MISCELLANEOUS STUDIES:

At Carnegie Institute, the ignition of coal particles has been studied in apparatus designed to avoid the complicating and obscuring effect of flames. Coal particles are dropped individually down an air gap between two, heated, parallel, carborundum tiles 1" apart. Because of the dispersion of the particles there is no flame and their temperature is that of the carborundum surfaces. Even at 700°C, the particles do not ignite but cool as they issue from the furnace and are recovered in the form of irregular, swollen particles of coke.

In a different apparatus, similar investigations have been made under pressure and in atmospheres of varying oxygen and nitrogen contents. It has been found that the effect of pressure is to retard ignition. This observation is explained by the fact that the coal particle is heated by radiation and then transfers heat by convection to the gas with which it is in contact.

The reactivity of fuels to oxygen is determined by passing air at a suitable fixed rate and temperature through a sample of the fuel contained in an adiabatic calorimeter. The rate of temperature rise is recorded and interpreted as a measure of the reaction velocity at progressively increasing temperatures. The reaction rate so determined is an exponential function of the temperature. It has been found too, that the volatile contents of coal, high temperature coke and petroleum coke may be correlated with the reactivity of those fuels, if the reactivity be expressed in terms of the temperature to which the fuel must be heated for low temperature oxidation to proceed at a standard rate. Low temperature coke, as made or after de-volatilisation by subsequent heating, does not behave in this respect as do the other fuels mentioned. The correlations determined are indicated in Figure 14. below:

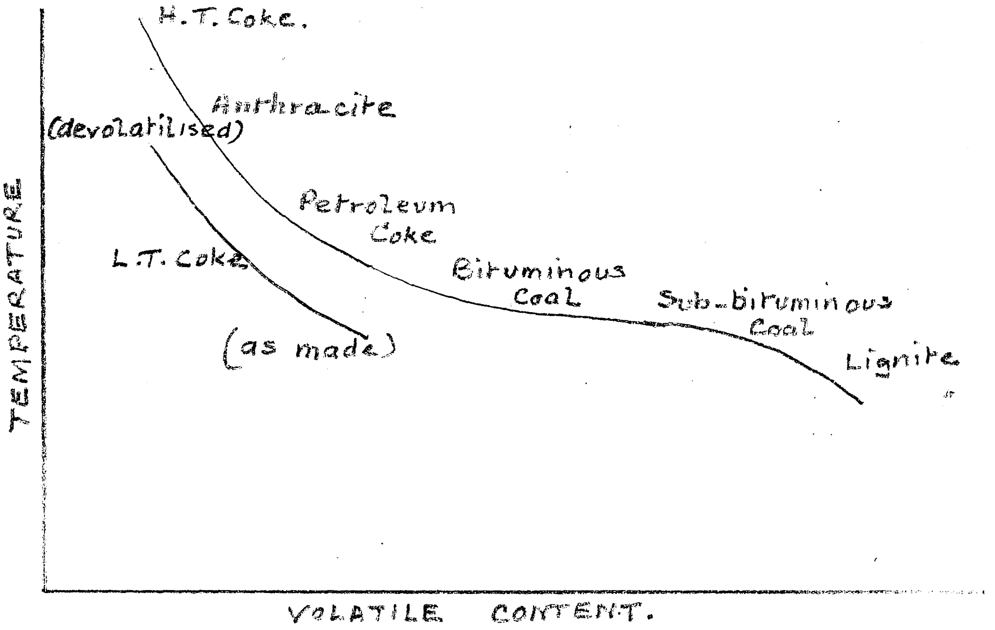

FIG 14 CORRELATION BETWEEN REACTIVITY AND VOLATILES (ORNING)

The reactivity of coke to a mixture of hydrogen and carbon dioxide or of steam and hydrogen has been investigated by passing those gas mixtures through a heated tube containing the coke sample. Under these conditions, a water gas "shift" was discernable at 650°C, though no gasification of the coke could be detected below 900°C. When the tube was packed with broken quartz instead of coke, no shift was effected at temperatures below 900°C. These phenomena could be explained by a catalytic action by constituents of the coke ash. The explanation preferred at Carnegie, however, is that oxidation of carbon, whether by oxygen, steam or carbon dioxide, is effected in two stages, viz. the attachment of anatom of oxygen to one of carbon and the subsequent detachment of a molecule of carbon monoxide. It is considered to be the second step which determines the rate of gasification of carbon.

This theory of oxidation is being tested in a fluidised gasification apparatus, with a coke bed 1" in diameter and 4" deep. The rate of reaction has been determined in two series of tests, using oxygen at different concentrations and temperatures in one case and steam in the other. The highest temperature at which useful information could be obtained from the oxygen reaction was found to be 700°C. At higher temperatures, temperatures could not be stabilised satisfactorily and fuel was consumed at a rate equivalent to exhaustion in a few minutes only. The lowest temperature at which the reaction rate with steam was fast enough for accurate measurement was found to be 900°C. Had it been possible to bridge or even partially close the gap between 700°C and 900°C, a searching test might have been applied to the theory just postulated for the mechanism of oxidation. That theory would be substantially confirmed if the reaction rate to oxygen at rising temperatures and the reaction rate to steam at falling temperatures were found to vary as the same function of the temperature.

To shed further light on the mechanism of combustion, work is being done on the determination of surface areas of coke by absorption of mercury under very high pressures.

Coal is bing partially oxidised on a pilot plant scale for the production of organic acids. A slurry of pulverised coal in caustic soda solution is pumped into an autoclave, in which it is heated to 170°C with a steam coil. Oxygen is then blown into the slurry, resulting in a rise in temperature which is limited to 270°C by passing cold water through the steam coil and by interrupting the oxygen supply intermittently. The temperature is maintained at 270°C until the reaction is complete. Of the carbon present in the coal, some 40% is oxidised to carbon dioxide and the rest to a mixture of acetic, oxalic and substituted benzene carboxylic acid up to the penta-carboxylic.

The reacted mixture is filtered to remove coal ash and some sodium carbonate. The filtered liquid is neutralised with sulphuric acid and the organic acids are extracted with M.E.K. The bulk of the water and M.E.K. is removed by distillation and the acids are finally dried in a film dryer. Octyl and butyl esters have been prepared, both deeply coloured. The butyl ester can be decolourised only by discarding about 60% of the product. This difficulty militates against the disposal of the butyl ester in what would otherwise be an attractive market, viz. the plastics industry.

The Battelle Institute has carried out combustion studies to which reference has been made elsewhere in this report. Early work on a pulverised fuel burner for gas turbine locomotives was assigned to the Institute; investigations are proceeding on the combustion of char from the fluid bed retort operated by Pittsburgh Coal.

In conjunction with the Norfolk and Western railroad, a study is being made of the emission and suppression of cinders from locomotives. It is believed that 10 to 30% of the fuel supplied to a locomotive may be lost in this way. The work in progress aims at the collection of those cinders and their return to the fire-box. A grit arrester has been designed for installation in locomotive smokeboxes and tests are proceeding on the device.

Model locomotives are being used for experiments on the effect of windage on the supply of air to the firebox and on the resistance to the flow of gases through the boiler.

Domestic heating appliances for central heating, cooking ranges and open fires, and for consuming coke, coal or anthracite, are being tested at a large number of institutions including Battelle, the Illinois State

Geological Survey Laboratories, the Anthracite Institute, the Fuel Research Station and the laboratories of the British Coal Utilisation Research Association. The design of modern equipment is the result of applying much hard work and ingenuity, in an effort to develop furnaces with a considerable degree of automatic operation.

The boiler availability work being carried out by B.C.U.R.A. comprises a study of sulphate deposits on the furnace side of boiler tubes and of the conditions leading to their formation. A method has been developed for the separate estimation of sulphur dioxide and sulphur trioxide in furnace gases. The sample is bubbled through isopropanol to absorb the trioxide and inhibit oxidation of the dioxide, the latter being subsequently estimated by reaction with iodine. An apparatus has been developed, also, for measuring the dew point of furnace gases. Condensation is detected by the closing of the gap of an electric circuit at the tip of a glass thimble. The temperature is determined simultaneously by a thermo-couple. The dew points so found are compatible with determinations of sulphur/dioxide and trioxide made as briefly described above.

It has been discovered that sulphate deposits may be inhibited by a trace of zinc oxide smoke. The cost of the zinc, however, is just a little too high to permit of practical application of this discovery.

GAS TURBINES[+]

It is generally conceded that while British manufacturers may still hold lead in the design and construction of aircrafy turbines, Switzerland is well in advance of other countries in the development of large machines for use on land and sea. Much work is now being done in the United States in the direction of applying temperatures considerably higher than those yet adopted by Swiss manufacturers. The experience of the next two or three years will be most interesting, in that it will probably establish whether efficiencies will in excess of 30% may be achieved over long periods, orwhether, in the more conservative view of the Old Worls, the materials of construction economically available limit gas turbine efficiencies in commercial applications to between 20% and 30% according to the use made of expensive heat exchurges etc.

BROWN BOVERI.

The Company has now manufactured, or is manufacturing for early delivery, gas turbines to a total capacity of 90,000 K.W. In this total are included 16 Howdry units, Eight similar turbines, manufactured under license by Allis Chalmers are not included. Partly because of the volume of work in progress but mainly because of a desire to gain more operating experience and to take stock of recent developments, Brown Boveri is temporarily refusing further orders. The largest machine yet completed by the Company is one of 13,000 K.W. capacity for peak load service at Beznau power station, Switzerland. A 27,000 K.W. turbine of the same make is to be installed in that station at the end of this year. Amongst other interesting applications of Brown Boveri gas turbines is one by the Anglo Iranian Oil Company. Three 4,000 K.W. units are being tested under field conditions with a view to the adoption of this prime mover for pumping oil through desert pipe lines. A 2,000 H.P. locomotive has been in operation for the Swiss railways since late 1941. On one occasion, the turbine was burnt out as the result of a failure of safety devices, permitting temperatures to rise to 850°C. Additional controls are now provided and a recurrence of that accident is regarded as unlikely. The smallest unit ever manufactured by Brown Boveri as an independent power unit is one of 1,000 K.W. capacity. Units of that size areregarded as hopelessly uneconomical and are unlikely to be repeated.

Brown Boveri turbines use the open cycle. In all applications except that of a locomotive for British Railways, heavy fuel oil is used and the combustion chamber is lined with refractory material. It is the Company's considered opinion that if distillate fuels are used in gas turbines to avoid troubles in combustion, the turbine loses its advantage over the heavier and more expensive diesel engine. Only by consuming a less expensive fuel can the turbine nullify the advantage which the diesel engine possesses in respect of efficiency. The maximum normal operating temperature in Brown Boveri gas turbines is 600°C and the metal used for parts under load at that temperature is 18:8 alloy. For short time service, this alloy is inferior to those used in the construction of gas turbines for military planes, for which an operating life of a few hundred hours may be accepted as satisfactory. If gas turbines are to justify themselves in land service, however, a useful life to be measured in thousands of hours is necessary. For such service, Brown Boveri has found 18:8 to be the best alloy now available, and 600°C the highest temperature applicable continuously with safety.

In order to obtain from a turbine operating at 600°C an efficienty somewhat higher than is normally realisable at that temperature, Brown Boveri is developing a device known as the "compreg" ++

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(108) See Woods M.W. and Wisdom J.L. Industrial Applications of the Gas Turbine. J. Inst. Eng. Aust. Vol.21 No. 4-5 P.67.

Karthauser F.B., Battcock W.V., and Ross F.F., The Coal-Fired Gas Turbine J. Inst. Fuel Vol.22 No.125 P.56

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(109) Meyer, Recent Developments in Gas Turbines. Mech. Eng. April, 1947. P.273.

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The unit consists of a steel rotor incorporating a number of cylindrical cells, each of which communicates in turn with air and hot gas passages. The combustion chamber temperature is maintained at 800°C, but in passing through the comprex, the gas is expanded and cooled to 600°C before admission to the turbine. The comprex itself is kept cool by the air which is compressed and heated in the process. This device has been found to function satisfactorily in principle but mechanical troubles have been experienced.

In the Velox boiler developed by Brown Boveri, power generation can be combined with steam raising by burning the fuel under pressure and passing the boiler flue gases through a gas turbine. A Velox boiler and 4,000 K.W. gas turbine installed at Brown Boveri's Baden works are fired in this way. In commercial practice, Velox boilers have been fired with oil and natural gas only. Brown Boveri has done work on feeding and burning pulverised coal under pressure, but apparently not with encouraging results. The Company is now experimenting with pre-gasification as an alternative means to the same end.

<u>OTHER SWISS MANUFACTURERS</u>.

Escher Wyss, Sulzer and Örlikon also build gas turbines, differing amongst themselves and from Brown Boveri in design. Escher Wyss uses a closed cycle. Örlikon has recently adopted a centrifugal compressor claimed to be as efficient as an axial machine. Sulzer's original idea was to carry out the combustion in a diesel engine, generating just enough power in the engine for the compression of the air. The products of combustion would be released at a moderate pressure and suitable temperature to a gas turbine, the whole output of which would be useful power. The idea has been abandoned and Sulzer uses, instead, a semi-closed cycle and re-circulation of air. The Company has installed a 7,000 K.W. unit in a Swiss power-station and is adding a 10,000 K.W. unit late this year. With this exception, it is understood that, no Swiss manufacturer other than Brown Boveri has sold more than than one gas turbine.

DEVELOPMENTS in ENGLAND.

Because they can be brought on load so quickly - in 15 minutes from cold - and because they have no standby losses, gas turbines are well suited to peak load operation in power houses. It appears to be these characteristics of turbines that arouse most interest in England. Electricity authorities are providing an opportunity for testing gas turbines in this service by placing orders for three 10,000 K.W. units. Metropolitan Vickers is to instal one at Trafford station shortly and Parsons is to manufacture one for Dunston. John Brown is making one, to Sulzer designs under licence it is understood, for Dundee. Two turbine locomotives are on order for British Railways. All these British gas turbines are to burn distillate fuel oils in metal combustion chambers of a type designed originally for aircraft jet engines.

It has been reported that gas turbines are being developed in England for the propulsion of motor vehicles, a field of application which on first thought, seems quite unattractive.

DEVELOPMENTS in U.S.A.

There appears to be little interest in gas turbines for power station work in U.S.A., the favoured fields for projected application being locomotives and to a smaller extent ships. The Navy first aroused interest in ship propulsion with this prime mover by ordering a turbine from Allis Chalmers for practical investigation and experience. The Elliott Company is completing, or has recently completed, two 3000 hp. turbines for the Navy, and one other unit for a merchant ship. The data reported herein relate to the Naval turbines.

The arrangement of the components of the power plant is as follows :-

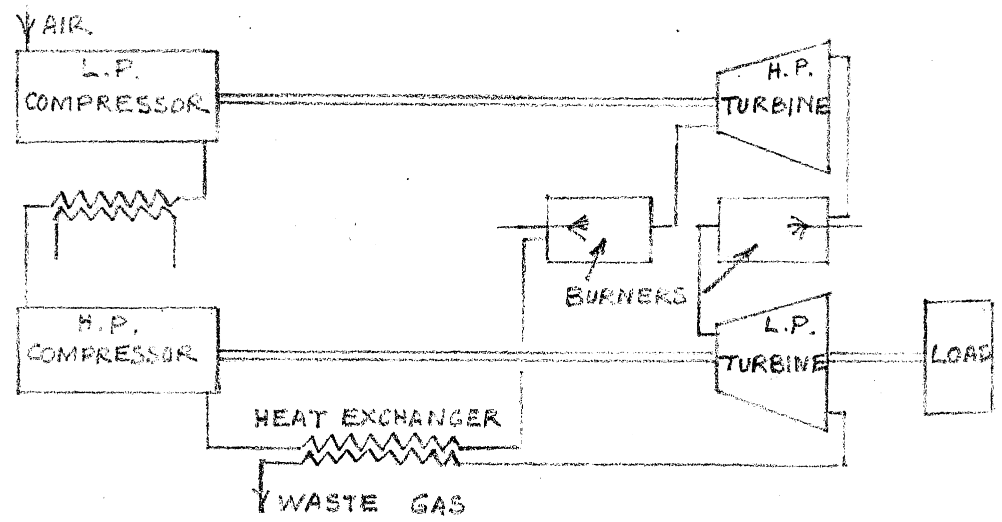

FIG 15. MARINE GAS TURBINE ELLIOTT CO.

Lysholm turbines are to be used for air compression, for reasons of convenience rather than efficiency. The Lysholm compressor is very stable and reliable, and it has been adopted in this instance so that the troubles likely to arise in the gas turbine and in the turbo-compressor may be encountered and solved separately. The turbines are ten stage machines with cast wheels and forged blades. The drive to the propeller shaft is electrical and reversing is effected by switch gear.

The temperature in the combustion chambers is to be 1450°F. The fuel to be burned is a distillate oil and there is one air-cooled, metal combustion chamber for each stage. The steel exposed to the high temperature zone is to contain 20% each of chromium, nickel and cobalt and 4% each of columbium, titanium and molybdenum. The outer shell of the combustion chambers, being exposed to less severe temperature conditions, contains 19% of chromium 9% of nickel and 1% of titanium. The pressure in the first stage is 3 atmospheres and in the second 7½. The anticipated efficiency at the shaft is 32-34%.

The Allis Chalmers turbine supplied to the Navy was to have been operated at 1250°F. for a time and thereafter at the full designed temperature of 1450°F. It has been stated that operation at temperatures above 1300°F has not been found practicable.

The move away from steam in American railroad practice has reached a stage at which there is no new construction of steam locomotives. The diesel electric locomotive operates with a lower fuel cost; its axle loading is lower so that wear and tear of the track is reduced; maintenance charges are lower and it spends a greater proportion of its time in useful service (95% as compared with 50%); finally, its operation is much smoother, particularly when starting - an advantage which appeals greatly to the passenger. Strange as it may seem at first sight, the diesel electric locomotive cannot be criticised on the score of oil conservation. For from one ton of coal fired to a steam locomotive it would be possible, otherwise, to obtain about two barrels of oil. Those two barrels in a diesel-electric locomotive would give at least twice as many ton miles as the ton of coal in the steamer. The diesel-electric locomotive does have some disadvantages, however, minor perhaps but genuine. Its capital cost is high and its length considerable. In a standard sixteen unit train calling for about 6000 h.p. the first three units are for traction.

Interest in the gas turbine locomotive has been inspired partly by a desire to compress the power unit into a space more nearly comparable with that occupied by a steam locomotive. This consideration applies equally to coal-fired and oil fired turbines. The other reason is the anxiety of coal interests to recover the market which has already been lost to a considerable extent to oil. Through their research organisation, Bituminous Coal Research Inc., they have spent much money in improving the performance of coal-fired locomotives generally. In co-operation with several railway companies, they have established and financed the Locomotive Development Committee to undertake the development of a coal-fired gas turbine locomotive.

There is one gas turbine locomotive in service in U.S.A. at the present time. The turbine etc. were made by Allis-Chalmers, originally the licensee of Brown Boveri in U.S.A., but now manufacturing to its own designs. The electrical equipment is by General Electric and the chassis by American Locomotive Co. The locomotive is the property of Union Pacific. It burns distillate fuel oil in eight metal, air-cooled, combustion chambers developed from the combustors used in aircraft engines. At the time of the last advice (June, 1949) it had been operated for 4,000 miles in freight service on routes adjacent to the manufacturers' works. It was believed to have given general satisfaction, but no details of performance were then available, particularly in respect of temperature effects. The design temperature was 1300°F. Information on the experience gained with this locomotive is due to be published by the A.I. Mech. E.

The Elliott Company is building an oil-fired gas turbine for a Santa Fé locomotive. The cycle is a simple one with a single stage of compression to 4½ atmospheres and a heat exchanger. The combustion temperature is to be 1300°F and the single, metal burner is made of steel containing 19% of chromium, 9% of nickel and 1% of titanium. The efficiency is expected to be 24% at the turbine shaft and 19% at the rails. The compressor is a two-stage, centrifugal machine, the ratio of compression per stage being greater than the 1½ preferred for stability but less than the recommended maximum of 3, which can be attained with some sacrifice in steadiness of performance.

The Locomotive Development Committee has two gas turbine power plants on order, one of 3,750 hp. by Elliott and presumably similar to the Santa Fe turbine, the other of 4,200 h.p, by Allis Chalmers. In the latter plant, air is compressed to 75 lb. pressure by a 21 stage axial machine. The hot gases are expanded in a 5 stage turbine. These locomotives were expected to be completed in the middle of 1950, but delays in solving combustion problems now make that target seem unlikely of attainment. This development is further reported in the section on combustion.

Westinghouse is understood to be building a gas turbine locomotive for an unidentified railroad. Chesapeake and Ohio has given some consideration to purchasing a locomotive powered by De Laval, but it is understood that no decision has been made.

It is the considered opinion of the Elliott Company that until gas turbines are standardised for mass production methods of construction - a development which is many years away - manufacturers are unlikely to become interested in gas turbines of so small a capacity as 1,000 K.W.

GAS PURIFICATION.

The removal of sulphur compounds from gases is being actively studied with many objects in view. In towns' gas a further lowering of sulphur content would reduce deterioration and maintenance of gas appliances. The oxide purification process in common use needs much improvement. For the synthesis of oil and methane, gas must be purified to extremely low concentrations of sulphur compounds. In conformity with other interests, the work being carried out in England is directed towards improving oxide purification of towns' gas and preparing water gas for methane synthesis. In America, the main problem being investigated is the purification of oil synthesis gas.

GAS RESEARCH BOARD:

In spite of claims made for various liquid purification processes over the last twenty years, oxide boxes are still used almost universally in the gas industry. Plant of this type costs almost as much as the retorts in which the gas is made, so there is much to be gained by improving the performance of oxide purifiers. The Board is currently working on the extraction of sulphur from charged oxide, using toluene as solvent. Two or three plants are believed to be operating in Germany, but using carbon bisulphide instead of toluene. The advantages to be gained by solvent extraction are -

1. The recovery of sulphur in a convenient form for utilisation.
2. Less loss of iron oxide,
3. The amelioration of the unpleasant conditions under which boxes are cleaned.

The purification of towns'gas in fluidised oxide is being studied in a bench scale apparatus. The purifier is about 2½" in diameter by about 30" high, with a short disengaging section above. The iron oxide is graded between 85 and 200 mesh, and forms a bed about 9" deep under static conditions. At a space velocity of 1000 - which contrasts with 5 in an oxide box - the expansion is about 30%.

The gravity of the suspension is rather less than unity and the back pressure about 12" W.G. Steam is added to the gas to keep the oxide properly hydrated. Sulphising and oxidation were originally carried out in separate fluid beds, so as to avoid dilution of the gas with nitrogen. The complications were found to outweigh the advantages of separate reaction vessels, however, so both operations are now carried out in the same bed.

A second fluid bed of the same diameter as the first, but somewhat shallower, is used in series with the other, to permit a high degree of purification while still maintaining a high concentration of sulphur in the spent oxide. It is intended that the clean-up column shall be super-imposed upon the main column, and that a continuous flow of oxide shall be established counter current to the gas. It is expected that 90% of the sulphur removal will be effected in the lower column and that the sulphur concentration in the discharged oxide will be 50%. It is further intended that the sulphur shall be extracted from the spent oxide by a solvent.

It has been found that elemental sulphur does not segregate in the fluid bed, nor does it become mechanically entrained in the purified gas. There is no significant change in the particle size distribution of the oxide in continuous service. Apparently the reduction in size by attrition is accompanied by a compensating crystal growth due to chemical reactions.

The advantages of fluid bed purification in conjunction with solvent extraction are reduced capital expenditure, the mechanical handling of foul oxide in a closed circuit, the recovery of elemental sulphur and the recovery of iron oxide in a usable form. The main disadvantage of the process is the pressure drop across the purifier. It is not expected that a reasonable excess of moisture in the gas will cause the oxide to agglomerate and impair fluidisation, but trouble of this nature may be experienced in commercial practice due to the tar fog commonly present in foul gas.

Work is being done on the decomposition of organic sulphur in the foul gas, in the hope of avoiding two stages of removal of hydrogen sulphide. To provide the steam needed for the catalytic reaction, the gas is cooled to 70°C only, which is just short of its dew point. At that temperature the condensation of light oils is incomplete, so the catalyst becomes fouled with adsorbed gums. The catalyst found to be most effective consists of chromium and copper on activated carbon. So far, it has not been found possible to revivify the catalyst, because gums can be removed only at temperatures which result in deterioration or loss of the active carbon. A test extending over a period of 100 days was made at a works equipped with an electrostatic precipitator. Results were as follows :-

	Average content of sulphur compounds, as grains of sulphur per 100 cubic feet of gas.	
	Raw gas.	Treated gas.
Hydrogen sulphide,	600	not quoted
Thiophene,	6	6
Other organic sulphur compounds	35	8.6

Thiophene does not respond to the process. The technique could be applied only at works where electrostatic precipitators are installed and there are few such gas works in England at present.

The same catalyst has been used in the Board's work on the conversion of organic sulphur in the gas to be used for the synthesis of methane. In this service, the gas is water gas free from tar fog etc. and it is indicated that the life of the catalyst is indefinitely long. The hydrogen sulphide formed from the organic sulphur compounds is removed by iron oxide followed by unreduced nickel. The sulphur content is 0.1 to 0.2 grain per hundred cubic feet at the outlet of the oxide boxes and only 0.001 grain after final purification. Because of the small amount of sulphur to be absorbed by the nickel, its life is satisfactorily long.

AMERICAN GAS ASSOCIATION:

The Association has sponsored work at the Institute of Gas Technology, which is aimed at the reduction of the total sulphur content of gas to one grain per 100 cubic feet. The method being investigated consists in scrubbing the gas with an oil solution of amines. Present indications are that the method is effective but expensive, costing about two cents per 1,000 cubic feet. As part of this programme, work is in hand for the development of suitable analytical methods for estimating mercaptans, carbon disulphide, carbonyl sulphide and thiophene separately.

BUREAU of MINES:

At the Morgantown laboratories of the Bureau of Mines, work is being done on the estimation and removal of sulphur compounds in gases.[+] A method has been developed for the estimation of organic sulphur using methylene blue as reagent. The test is so sensitive that triple distilled water must be used if consistent results are to be obtained. A study has been made of the removal of organic sulphur by passing the heated gas over a Girdler catalyst consisting of iron and chromium. The organic sulphur other than thiophene is thereby converted to hydrogen sulphide even when that constituent is present in relatively high concentrations, such as 200 grains per 100 cub. ft. The reduction in organic sulphur content, excluding thiophene, effected under such conditions may be from 30 or 60 to only two grains per 100 cub. ft. There is reason to believe that the two grains is an irreducible minimum for gases containing carbon monoxide and that the residual sulphur is present in the form of carbonyl sulphide. When carbon monoxide is absent, the

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[+] Sands A. E., Wainwright H. H. and Schmidt, L. L. Purification of Synthesis Gas Produced from Pulverised Coal. Ind. Eng. Chem. Vol.40, P.607, April, 1948.

gas may be purified in the same way to an organic content well below two grains per 100 cub. ft.

A small pilot plant is used for investigating the continuous removal of sulphur compounds from gas by solution in mono.-, di-, or tri- ethanolamine.

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SYNTHESIS of OIL and METHANE.

There is no longer any significant interest in the original form of the Fischer Tropsch process, using a cobalt catalyst in a fixed bed. Two German plants are understood to be in operation for the production, mainly, of synthetic fatty acids and other chemical products. In England the process appears to be now regarded as of purely academic interest, and of possible use only in the study of the mechanism of certain features of the synthesis reaction. All work in America is concentrated upon modifications of the process using an iron catalyst and improved means of temperature control in the reactor. Of those improved techniques, the fluid bed process developed by Standard Oil, Texaco, Hydrocarbon Research and others has made most progress. The Bureau of Mines has deliberately avoided the fluidised process because it is being adequately developed by private enterprise. The Bureau has chosen instead to investigate various methods of using oil as a cooling agent in the reaction zone.

Iron catalysts and the oil cooling technique were German inventions, but were not adopted in any German commercial plant. The failure to apply these processes may be attributed to Hitler's action in "freezing" further developments, for reasons of policy, in the early years of the war. In so far as the first large scale use of a fluid bed in a chemical process was in Winkler's water gas generator, some measure of credit must be given to Germany for later revolutionary applications in U.S.A. However, it seems unlikely that the Germans would themselves have applied fluidisation to oil synthesis, even if research and development had been permitted to proceed normally. A reading of the patent literature indicates that Winkler was interested rather in the Duft Schmidt variation of the Fischer Tropsch process, using a circulating oil stream for temperature control.

The activity of cobalt catalysts is dependent upon the cobalt being dispersed in particles of molecular fineness on the kieselguhr carrier. Because of this heterogeneity, the fluidised technique cannot be applied to the cobalt catalyst. The attrition inevitable in a fluid bed separates the cobalt from the kieselguhr and a substantial loss of activity results.

There has been a tendency for the improvements effected by fluidising the catalyst bed to be overemphasised. In German practice, the capital and operating costs for synthesis reactors were about 20% of the total in each case. The savings made possible by fluidisation or oil cooling must, therefore, be within that limit. Improvements of probably equal magnitude have resulted from adopting the iron catalyst, which gives a product of greatly superior value, and from a considerable reduction in the cost of the synthesis gas. Up to the present time, the lower cost of the gas in American or American sponsored projects is due to cheaper fuel being available and not to the application of some gasification technique not previously known. It seems likely however, that out of the efforts now being concentrated on the gasification of coal in U.S.A., some process will be developed, making substantial economies possible in the production of synthesis gas.

COMMERCIAL PROJECTS:

About four years ago, a decision was made by a number of companies with natural gas interests in Southern Texas to instal a plant at Brownsville for the synthesis of oil from that gas. The Texas Company had the largest individual holding (about 38%) in the enterprise and half the capital was advanced by the Reconstruction Finance Corporation. Plant design was the responsibility of Hydrocarbon Research Inc., who, in conjunction with Texas had developed the processes to be used. Those processes were the partial oxidation of methane to synthesis gas using oxygen, and fluid bed synthesis using an iron catalyst. The capacity of the plant was 7,500 barrels of liquid product per day. The unit was originally planned to be completed late in 1947. It is not yet ready, but is reliably reported to be nearing completion and due for commissioning not later than February, 1950.

It would appear that delays in installation have not been due mainly to shortages of materials, but rather to the unexpectedly long time needed for pilot plant studies. It is believed that difficulties were experienced in the control of carbon formation on the catalyst. That carbon, though finely divided, has the agglomerating characteristics of lamp black and interferes with fluidisation. It was found that the trouble could be suppressed by maintaining a high ratio of hydrogen to carbon monoxide in the reactor, with higher pressures and temperatures to suit. These adjustments probably entailed, in turn, changes in the proportions of the liquid products. Due to rising costs during the period of delay, but probably in some measure also to changes in design, the capital cost of the plant has increased from the original estimate of sixteen million dollars to the thirty two now considered necessary for completi

In the early days, claims were made for "five cent oxygen" and "five cent gasoline". Little significance attaches to the price for oxygen, for it excludes the two major items of cost, viz. -

1. Power or steam (assumed to be available at no charge because of the waste heat steam generated in the reactors).
2. Capital charges (on $3,000,000 for 2,000 tons of oxygen per day)

The cost of the petrol needs revision today, even if only for the increased capital expenditure, but there is little doubt that the plant will earn substantial profits. It is understood that a long-term contract was negotiated for natural gas at three cents per thousand cubic feet.

The oxygen plant is being installed in two units of 1,000 tons daily capacity per unit. The turbo compressors manufactured by Clark Bros. have been said to be the largest machines of their kind yet constructed. The double fractionating column is 12 feet in diameter and, because of the dimensions of the plant, has three external reboiler-condensers instead of the usual internal unit. The high pressure section is being made of stainless steel because copper is not readily available. The lower column is of aluminium, 2" thick. A.L.C.O.A. and Foster Wheeler, the manufacturer, have collaborated in the development of a new welding technique for this job. The actual welding is done while the aluminium plate is heated in a furnace improvised to fit the job.

It is understood that combustion of the natural gas is to be carried out in two chambers, and that six synthesis reactors are to be installed 14 feet in diameter by about 40 feet high. The fluid catalyst bed will be about 15 feet deep. Boilers will be built into the vessels, with tubes immersed in the catalyst. The quantity of synthesis gas required per day is 250 million cubic feet of which one seventh will be supplied by circulating tail gas. To keep down the nitrogen content of the synthesis gas, tail gas amounting to 5% of the total quantity will be withdrawn and used as fuel. Catalyst will be withdrawn for revivification at the rate of 1% per day.

Of the carbon monoxide supplied in the synthesis gas, about 90% is converted, re-appearing as carbon dioxide (15%) liquid products (80%) and hydrocarbons below C_3 (5%). Of the liquid product, about 12% consists of oxygenated compounds, 13% of diesel and fuel oil and 75% of petrol. About 5% of the petrol is obtained by polymerisation of propylene and butylene. The petrol is of high quality, having octane numbers of 80 by the motor method and nearly 90 by the research method, in each case without the use of lead. The diesel oil is of poor quality, its cetane number being low in the forties. The oxygenated compounds are a complex mixture of alcohols, aldehydes, fatty acids and ketones, with ethyl alcohol, acetaldehyde, acetone and acetic acid the most important constituents.

Shortly after the Brownsville project was announced, the Stanolind Oil and Gas Company decided to erect a similar to make use of surplus gas from the Hugoton field in Kansas. The capital cost of the project was to be over eighty million dollars. Stanolind was to have processed the oxygenated products from both plants and to have sold refined products to the U.S. Chemical Co. Equipment was to have been installed also for the catalytic dehydration of oxygenated compounds, presumably those which would have been produced in excess of the market demand for them in their original form.

It was announced twelve months ago that Stanolind had decided to go no further with the project, and it was understood that cancellation of orders already placed had involved the Company in losses amounting to about eight million dollars. There are probably several contributing reasons for the decision, viz. the high capital cost of the plant, the improved value of natural gas for other purposes and, perhaps, a desire to have the process proved on a commercial scale at Brownsville before further expenditure is incurred by Stanolind. The Hugoton gas contains 18% of nitrogen. Its value, therefore, may have been appreciated by recent work on the removal of nitrogen from gases intended for transmission by pipe line for long distances.

It is understood that Stanolind is continuing its work at Tulsa on the refining and disposal of oxygenated products of the synthesis process. It may well be, therefore, that the project has been deferred rather than completely abandoned.

A project has now been under consideration for two or three years, for the installation in South Africa of a synthesis plant of the same capacity as the two previously mentioned. The Company has committed itself to the extent of retaining Hydrocarbon Research to prepare the necessary plans and specifications. It is understood, too, that a license agreement for the patented processes involved has been negotiated with Texaco. Further action is dependent upon suitable arrangements being made for the provision of capital, of which about eighty million dollars is believed to be needed.

The raw material for the process is a coal of 28% ash content valued at £1. per ton. It may be the coal which is mined with the oil shale at Ermelo and which is produced in considerable excess of market requirements. Hydrocarbon Research has recommended for this project the Lurgi high pressure gasification process with certain modifications suggested by H.R.I. to improve its efficiency. The South African project is superficially very attractive because of the cheap coal available and because of the high price of petrol and other petroleum products in the interior of the country where the plant would be situated. Production costs under South African conditions are quoted by H.R.I. as 14 cents per U.S. gallon.

RESEARCH WORK in PROGRESS.

The oil companies most active in the investigation of the synthesis process are believed to be Standard Oil, Texaco and Hydrocarbon Research. It is understood that H.R.I does work on contract for two other companies, between them, however, it seems that there is no interchange of information on current work. It has been reported that the M. W. Kellogg Company and Cities Service Oil Company are working in collaboration, and that a fourth group has been formed by Gulf and Koppers. Little is known of individual activities. Patents of value and relevant to the synthesis process have been pooled with Texaco acting as the co-ordinating authority. This arrangement applies only in respect of licenses issued for operations in U.S.A. and does not include patents on gasification of coal by fluid bed or other processes.

The particular technique favoured in the oil industry is the fluidised synthesis method and the scale of the pilot plant operations conducted by various organisations to acquire information for the design of commercial units is about ten barrels of oil per day. Work on catalysts, gas mixtures, operating conditions etc. is performed on a much smaller scale. It is often convenient in bench scale units to operate with low gas velocities, in which case mechanical stirrers are used to maintain fluidised conditions. Because of abnormal heat losses from small plants and the abnormal effect of wall conduction, Dowtherm jackets are commonly applied.

The Bureau of Mines[+] is investigating, or has investigated three means of cooling synthesis reactors with oil. The evaporative method in which an oil boiling at the reaction temperature is supplied to the converter, has been abandoned. The techniques still under investigation are :-

1. The circulation of oil through a fixed catalyst bed and an external cooler, i.e. the Duff Schmidt process.
2. The circulation of a slurry of oil and catalyst through the reactor and an external cooler.

The reaction space in each of the two Duft Schmidt units at the Bruceton station is 3" in diameter by about 8 feet high. It has a volume of 0.4 cub. ft. and takes a charge of 72-75 lb of catalyst. The catalyst is a sintered iron product as used for ammonia synthesis and is screened between 4 and 6 mesh. Gas of low hydrogen: carbon monoxide ratio can be used, a point in favour of the technique because the water gas shift operation becomes unnecessary. Equimolecular proportions are commonly used with a recycle rate of 1:1. The pressure on the unit is 330 lbs and the reactor temperature 248°C and 263°C at the bottom and top respectively. The spread in temperature across the catalyst bed is determined by the rate of circulation of "cooling" oil, which in so small a plant actually needs heating. Under the conditions stated, about 12% of the carbon monoxide is converted to hydrocarbon gases. The yield of higher hydrocarbons is about 150 grams per cubic metre, 50% comprising petrol of 65 octane number and excess gasol, 35% being wax and the remainder diesel oil. There is no significant production of oxygenated compounds.

[+] See U.S. Bureau of Mines R.I.4456. Synthetic Liquid Fuels. 1948 Annual Report of the Secretary of the Interior, Part 2. Oil from Coal.
U.S. Bureau of Mines Tech. Paper 709 Storch H.H. et al. Synthetic Liquid Fuels from Hydrogenation of carbon monoxide.

At the time of the last information, the longest period of service obtained from a charge of catalyst was three months. Recharging had become necessary because of agglomeration, not loss of activity. It was believed that agglomeration could be prevented and that much longer catalyst life was possible. The current cost of ammonia catalyst in small quantities is 30 cents per lb. It could be produced, in large amounts certainly for 15 to 20 cents per lb. and possibly at a still lower price.

A plant is being installed, and is probably now in service, for studying the Duft Schmidt process on a scale of 1½ barrels of product per day.

Less progress has been made with the slurry circulation process, but results have been encouraging. The catalyst used is the same but it is pulverised to pass a 250 mesh sieve. The medium in which it is suspended and circulated is mainly wax, by which is meant any synthetic oil product boiling at a temperature above 250°C. To recover used catalyst from the suspending medium it is filtered while hot through a porous brass thimble, which retains particles coarser than five microns. In the slurry process, the mean temperature may be kept below 250°C and the spread limited to 2°C. Under these conditions, the conversion to methane is restricted to 4-6%.

The different mechanical conditions in the fluidised and oil cooled catalyst beds entail changes in physical conditions and these in turn result in differences in chemical reactions. Wax cannot be permitted to form in a fluid bed, or it will ultimately prevent fluidisation. Temperatures must therefore be raised above the optimum for yields of liquid hydrocarbons. Consequently, methane and oxygenated compounds are formed in greatly increased amounts. (It should be noted, in this connection, that the methane production herein discussed for the fluid bed operation is not the methane lost in the purge gas. Three times that quantity of methane is circulated in the plant, consuming oxygen in the process.) To prevent carbon being deposited on the catalyst at those higher temperatures, pressure and hydrogen concentration must be increased as already mentioned. Therefore, losses of free and combined hydrogen in the purge from a fluid bed reactor are higher than they would be in comparable circumstances from an oil cooled reactor. It follows, also, that liquid yields should be slightly lower in theory, oxygen consumption somewhat higher and waste heat evolution also higher in the fluid process.

The synthetic petrol from the fluidised catalyst requires treatment for the decomposition of traces of oil-soluble, oxygenated compounds which would otherwise impair its stability in storage. Under the conditions applied for the dehydration of those compounds, some isoforming of olefines also is effected. That is to say, double bonds in the terminal position are transferred to an inner position with an accompanying improvement in octane number. To some extent - admittedly a minor one - the high octane rating of synthetic petrol from a fluid bed operation is a result of this isoforming. The 65 octane petrol from the Duft Schmidt process has not been subjected to catalytic dehydration and incidental isoforming. Whether the treatment is needed or not for the elimination of oxygenated compounds, it could be applied for the improvement of octane rating. The advantage of the fluid bed process in respect of petrol quality is not to be measured, therefore, in terms of octane numbers herein quoted, viz 80 and 65. The difference under comparable conditions would be much less.

However for the production of synthetic oil in limited quantities, the fluid process has an undoubted advantage in respect of the quality and value of products. If operations were to be instituted on a considerable scale, however, the oxygenated fraction of the product would become an embarrassment. Surplus wax from the oil-cooling process could always, on the contrary, be profitably cracked to petrol. In these circumstances there might be little margin between the two techniques in point of product value.

It is not possible, at the moment, to assess the relative merits of the two processes in respect of specific output per reactor, because the precise meaning of the space velocities quoted is not known. It appears however, that production from equipment of comparable dimensions would be of the same order of magnitude. The advantage might rest with the fluid technique but it would not be considerable.

Regardless of what may ultimately prove to be the better process, there is no alternative to the fluidised method for early commercial adoption. Only little work has yet been done on the 1½ barrel per day unit at Bruceton, and some time must elapse still before results are forthcoming from the 80 barrel per day plant being installed by the Bureau of Mines at Louisiana, Mo.

<u>The Fuel Research Station</u> has a small fluidised synthesis plant with a reactor 1" in diameter and 9 feet high to the bottom of the disengaging zone which is 3" in diameter and 1 foot high. In current operations, the hydrogen: carbon monoxide ratio of the gas used is 2:1 and two volumes of tail gas are recirculated for each volume of fresh gas. The pressure is 25 atmospheres and the reactor temperature 340°C. The linear velocity through the reaction zone is 0.38 to 0.4 feet per second, equivalent to a space velocity of 1400 on the fresh feed calculated at atmospheric temperature and pressure. Linear and space velocities of 0.6 feet per second and 2,000 have been tried and found too high for best results. The total liquid product comprises about 60 parts of petrol and surplus gasol, 30 of heavier hydrocarbons and 10 of alcohols etc. The gasol is completely unsaturated, whereas the corresponding product from a cobalt catalyst is only 60% unsaturated.

Some interesting work has been done on a cheap catalyst prepared from common mill scale by activation with potassium carbonate. Early results were very encouraging, but it was found ultimately that the rather copious deposition of carbon precluded further use of the catalyst. Ammonia catalyst is now being used once more.

For visual observation of the mechanics of fluidisation and the behaviour of catalysts in a fluid bed, a transparent replica of the operating model has been made, with a height, however, of only four feet. One service to which this model has been put is the examination of used catalysts for segregation, particularly of carbon.

Preliminary work is proceeding on the slurry version of the synthesis process using an ammonia catalyst suspended in wax. It is indicated provisionally that very little wax is made in the process, the wax present in the circuit after several days operation not differing materially in quantity or quality from that originally used as the vehicle for the catalyst.

Work done at the Station and elsewhere has indicated that methane is not merely a diluent in synthesis gas but is capable of taking part in the reaction to form higher hydrocarbons. Information on the subject is somewhat contradictory and inconclusive, however. It is regarded at the Station as a likely explanation that the apparent activity of methane is obtained only with a fresh catalyst. Operating results over long periods at Essener Steinkohle failed to show any such activity. In planning commercial operations, it would be wise to regard methane in synthesis gas as a diluent in respect of conversion and a depressant in respect of reaction rate.

METHANE SYNTHESIS.

The latest information on this subject is that reported by the Gas Research Board* in England. The Board has set itself the task of studying methane synthesis as a means of enriching water gas throughout the winter peak in the British gas industry. The preliminary purification of the gas is considered elsewhere in this report.

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* Communication G.R.B.20, An Investigation into the Catalytic Synthesis of Methane for Towns Gas Manufacture.

Work being done by the Board on the water gas process itself, so as to adapt it more efficiently to methane synthesis is also reported elsewhere.

The gas used in the investigation was purified to a sulphur content of about 0.001 grain per 100 cubic feet. The synthesis catalyst was nickel activated with alumina in the proportions of 20:80. Used in this proportion, the nickel is so finely dispersed as not to form a crystal lattice. Because carbon deposition normally occurs by penetration of the lattice, the catalyst so prepared is not inactivated by carbon. It may be heated to 800°C without any sintering. In the four month's operation at a pressure of 20 atmospheres, and with a 3:1 ratio of hydrogen to carbon monoxide, no activity was lost and the absorption of sulphur was only 0.1%. The experiment was stopped at the end of four months, because that is the duration of the winter peak load in England, and because it was sufficient to establish that production need not be interrupted for that time to permit of catalyst being reactivated.

A plant is now being installed by the Board at Poole gas works for the synthesis of methane from blue gas at atmospheric pressure. The reactor is about 12" in diameter and its effective height about 18". The catalyst is contained in an annular space 3" thick, through which the gas is to pass radially with a low pressure drop. The space velocity is to be 2000 per hour and the throughput of the plant 1,000 cubic feet per hour. Even for a large commercial plant, therefore, a methane synthesis reactor would be a very compact unit. Temperature control at Poole is to be by recirculation of partially cooled gas. The inlet temperature to the reactor is to be 300°C and the outlet temperature 500°C. It is expected that a methane concentration of 70% will be achieved.

Opinion regarding the process in the Board is to the effect that it is technically sound and practicable but commercially inapplicable under English conditions because of the heat dissipated in the reaction.

HYDROGENATION of COAL.

Except for the work being done by the Bureau of Mines, interest in the commercial hydrogenation of coal has almost completely disappeared. Imperial Chemical Industries Ltd. continues to hydrogenate tar in its Billingham plant, but regards coal hydrogenation under existing conditions as hopelessly uneconomic.

+ Until quite recently, the Bureau's investigations+ on coal hydrogenation had been carried out in small autoclaves at the Bruceton station. Several small continuous plants have now been installed, with pressure vessels about 12 feet long by 3 inches in diameter. The completion of this installation is so recent that no operating experience has yet been disclosed. The larger scale demonstration plant at Louisiana, Mo. has, also, been completed only this year. At the time of the last advice, the product distillation plant had been commissioned on crude oil, and the vapour phase, hydrogenation plant had also been put into operation on light oil.

From its close engineering study of German and British practice, the Bureau has concluded that the efficiency of the process may be improved considerably by the use of self-heating preheaters and elaborate heat exchangers, by the recovery of power in the let-down of high pressure products, by flashing residues in highly superheated steam and by other means. It has been estimated that in this way the efficiencies realised in European practice, viz little over 30%, could be raised to 50% or more. Unfortunately, except in the recovery of oil from residues, these ideas are not to be put to practical test in the Louisiana plant. The high capital cost of equipment has necessitated considerable curtailment of expenditure on the plant. It is believed that the efficiency of conversion will not greatly exceed 20%.

+ U.S. Bureau of Mines RI.4456 Synthetic Liquid Fuels, 1948 Annual Report of the Secretary of the Interior, Part 2. Oil from Coal.

At Bruceton, some very preliminary work is in progress on a variation of the hydrogenation process which is regarded provisionally as the most attractive method yet devised. The suggestion ~~is that coal shall be partially~~ is that coal shall be partially hydrogenated in the absence of a vehicle and in a fluidised condition. It is believed that the intimacy and uniformity of contact so obtained will result in an even partial hydrogenation of the coal. In a fixed bed, under such conditions, part of the coal would be completely hydrogenated and therest merely coked, leading to insuperable operating difficulties. When coal is completely hydrogenated suspended in a vehicle, for each ton of coal converted to petrol with an efficiency of about 80% another ton is consumed in supplying heat, power, steam and hydrogen for the process. The present aim is, in effect, to hydrogenate both tons partially for a petrol yield of 40% and to use the residues for power generation etc. It is expected that a sufficient degree of hydrogenation will be effected at 500 lb pressure and at a temperature lower than that normally applied. Capital expenditure, hydrogen consumption and asphalt production should be reduced and reaction rates increased. A substantial reduction in costs should, therefore, be feasible without any sacrifice in yield. The development is still in the test tube stage and there are obviously mechanical difficulties to be overcome.

At the Carnegie Institute, hydrogenation+ is being studied for the information which it may shed on the constitution of coal. Hydrogenation is effected under the mildest possible conditions leading to substantial conversion to soluble products. Temperatures applied are about 375°C, pressures 200 atmospheres and periods of treatment about 72 hours. Copper chromite is used as catalyst because of its effect on carbon-oxygen combinations. The product is analysed by chromatographic methods.

+ Fuel, Vol.27 pp 96-99. (July - August 1948).

COAL DRYING.

Current American practice in the drying of coal has been reviewed and reported by the Battelle Institute.[+] In the course of visits to various plants, usually with other primary objectives, coal drying plants embodying three different and interesting techniques were seen.

FLASH DRYING:

As previously mentioned in connection with the low temperature carbonisation of coal by the Disco process, the new plant at Imperial is equipped with flash dryers for the wet coal. To dry the washed screenings from 14% to 4% of moisture, three units have been installed with a capacity of twenty tons of coal per hour each. The coal is fed into vertical shafts about three feet in diameter, meeting hot products of combustion at a temperature of 1,000°F or 1,100°F. The coal is carried upwards at a velocity of 5,200 to 5,600 feet per minute to a cyclone about 30 ft. above the coal inlet. The time of contact between the coal and the hot gas is about ½ second. The coal is separated in the cyclone and delivered onto conveyors leading to the carbonising plant. The hot gas and steam evaporated from the coal are discharged to atmosphere at about 350°F. The hot gas is provided by combustion of coal in a furnace and admixture of excess air for the proper adjustment of temperature.

At the Golden station of the Bureau of Mines, sub-bituminous coal is being flash dried in a pilot plant using a slightly different technique from that described above. The plant is illustrated in figure 16. Natural gas is burned in a combustion chamber and the temperature at the outlet is adjusted and maintained at 1,600°F by circulating the mixture of used gas and steam evaporated from the coal. Coal crushed by a hammer mill to a maximum particle size of 1/8" or 1/16", is carried by a

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[+] Lyons O. R. and Richardson A.C. "Thermal Drying of Coal". A.I.M.M.E. Technical Publication 2399. Class F. Coal Technology, August, 1948.

stream of inert gas into the bottom of the vertical pipe through which the hot gases leave the combustion chamber. Those gases heat the coal to 700°F and carry it upwards to the top of the pipe and then downwards into a separating hopper. The bulk of the coal isdisengaged from the gas stream merely by reduction of velocity and change of direction. Most of the fine coal, still entrained in the gas, is separated in a cyclone and returned to the hopper. The gas, with a little fine dust suspended in it, is withdrawn from the cyclone at a temperature of 300°F. by a fan which returns most of it to the burner as previously mentioned. The remaining gas is discharged to a holder through an electrostatic precipitator. The inert gas for carrying coal into the plant is obtained by cooling part of the products of combustion generated in the furnace.

The coal as charged has a moisture content of 18%. The gas requires only 1.2 seconds to pass from the combustion chamber to the disengaging hopper. The temperature of the gas and coal entering the hopper is about 400°F. The dried coal is withdrawn from the hopper in particles substantially of the same size as those of the raw coal.

It is understood that a plant of this type is to be installed to dry 200 tons per day of Montana coal.

DRYING in a FLUID BED.

At the Golden Station, preliminary work has been done also on the drying of fluidised coal. The arrangement of the plant is indicated in figure 17. About 500 cubic feet of gas and 1,000 of air, both preheated, are burned each hour in a combustion chamber. Products of combustion at 1800°F are admitted to the bottom of the fluid bed, which is 6" in diameter and about 6 feet high, containing 15 lbs. of pulverised coal. The coal is injected, also to the bottom of the bed, by 300 cubic feet of inert gas per hour. The temperature resulting from the mixing of coal, inert gas and products of combustion is 600°F. The mean period of retention of the coal in the fluid bed is 200 seconds, the rate of coal feed being, therefore, about

250 lb. per hour. The dried coal dust and gas pass through a vertical standpipe into a cyclone separator. The gas is discharged through an electrostatic precipitator to a gas holder, while the dried coal is withdrawn from the system through an exchanger which cools the coal and preheats the gas and air supplied to the combustion chamber.

FLEISSNER PROCESS:

Little further work has been done on the Fleissner plant at the Bureau of Mines station, Grand Forks, N.D., since progress was last reported in the technical literature. The process consists in heating the coal in an autoclave by admitting saturated steam at 400 lb. pressure and subsequently releasing the steam. The drying effect may be increased by later applying a vacuum to the autoclave or by blowing air through it. North Dakota lignite containing 35% of moisture as mined is dried to about 13% moisture content, in which condition it is in equilibrium with the atmosphere. The drying is attributed to :-

(a) The sqeezing of one third of the moisture out of the coal by pressure alone.

(b) The heating of the coal to 450°F.

(c) The rupture of fine pores in the coal when the pressure is suddenly released, allowing moisture to drain out of the coal.

There is a considerable reduction in the size of large particles of coal when dried in this way, but only little reduction in the size of small particles.

It is understood that charging and discharging pouches are being fitted to the Grand Forks plant, so that it may be operated semi-continuously. So far as is officially known, Fleissner dryers have not been successfully charged and discharged continuously.

DRYING LIGNITE in the MILL:

At the Otter Tail power station, Crookston, Minn., a Riley boiler rated at 75000 lb of steam per hour is being fired with pulverised lignite, flash dried in the mill. It is understood that this method of drying was first applied in Raymond mills grinding Texas lignite and that interest in the technique lapsed entirely when the East Texas oil field was discovered. The idea had now been revived and is being further developed by the Riley and Otter Tail Companies in co-operation.

Drying is effected by admitting to the Attrita pulveriser air which is normally preheated to 650°F. The air temperature has been as high as 800°F, however, under conditions of high load. The dried coal and primary air reach the burner at 130°F. Combined primary and secondary air from the forced draft fan pass through a preheater in the usual position behind the boiler, acquiring a temperature of 400°F by exchange with flue gases. The secondary air proceeds directly to the burners, but the primary air is further heated to 650°F, as stated, in another preheater located above and to the rear of the boiler. The air so heated, amounting to about 1 lb per lb of fuel and representing about 25% of the total air supply, passes to the mill. Controls are provided so that the hot flue gases, proceeding in turn through the primary air heater, the combined air heater and the induced draft fan to the chimney, can be drawn from either the beginning or the end of the last pass in the boiler. It appeared to be normal practice for the boiler gases to be completely diverted from the last pass. The temperature of the waste gas entering the chimney is high, viz. 500°C.

The power consumption of the mill is stated to be 10 K.W. hours per ton of lignite; a normal consumption for so hard a coal is 20 units per ton. It would appear that flash drying in this manner makes the lignite more easy to crush, perhaps as the result of a development of steam pressure within the coal particle.

No trouble had been experienced in the brief period of these operations (three weeks only) from fires or explosions in the mill or in pipes leading to the burners. This technique of drying and firing lignite is still regarded as in the experimental stage; further experience and adjustment - particularly in respect of heat losses in the flue gas - are desired before an assessment of the method can be made.

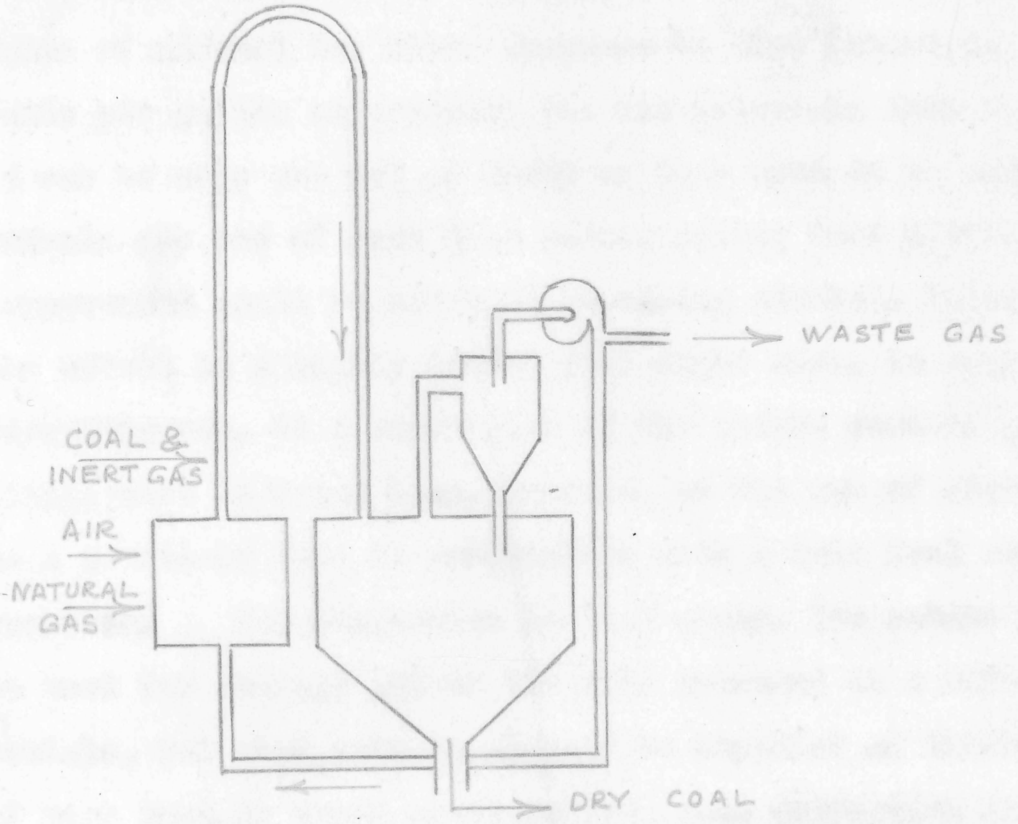

FIG 16. FLASH DRYING OF COAL
BUREAU OF MINES, GOLDEN COLO.

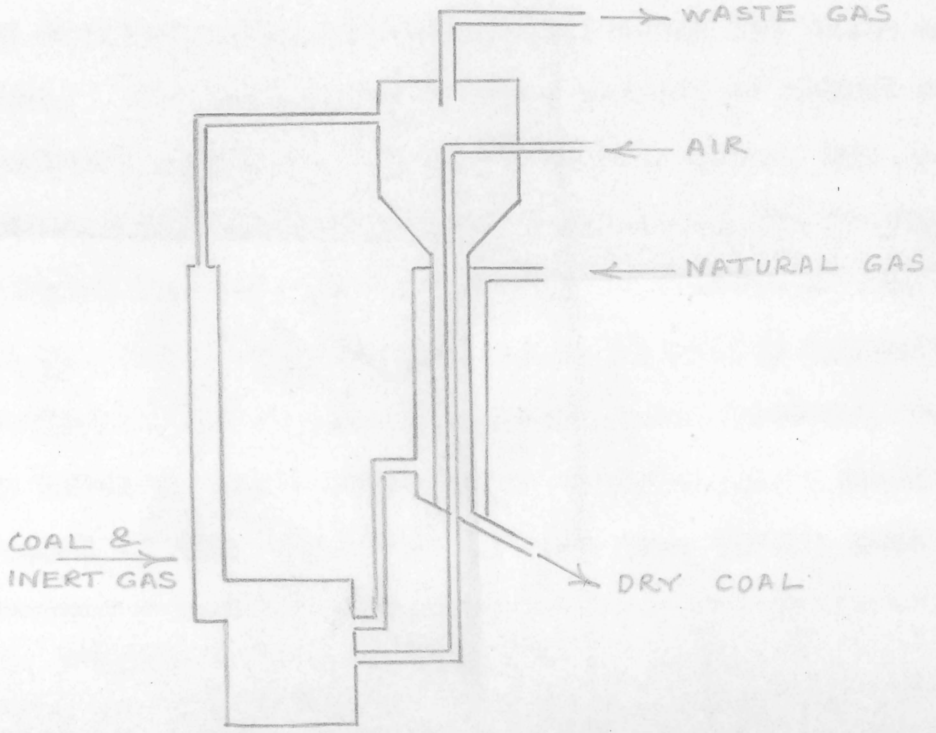

FIG 17. DRYING OF FLUIDISED COAL
BUREAU OF MINES, GOLDEN COLO.

MISCELLANEOUS.

ALCOHOL etc. from FARM WASTES.

There has never been any considerable interest in the use of power alcohol as a motor fuel in the United States, and today there is practically none. The value of alcohol for other purposes is high (about 35 cents per gallon at present) the raw materials from which it may be made are not as cheap as they used to be and petrols are now of such high octane rating that little improvement could be effected by adding alcohol. Indeed, the effect on a highly leaded fuel might even, in some circumstances, be a depression of the octane number. A little work is being done, however, on the use of alcohols as a secondary fuel in conjunction with a twin fuel carburettor. The suggestion is that cheap, low octane petrol be used for perhaps 90% of the fuel consumed in a motor vehicle, but that aqueous alcohol be supplied at times of high load or rapid acceleration, when detonation is apt to occur. Carburettors are available commercially for automatically carrying out the desired functions, the control being usually on the manifold vacuum. It is claimed that in this way, even modern engines will perform satisfactorily on a 60 octane petrol as the main fuel. At one garage in Columbus, Ohio, a carburettor and auxiliary fuel are being sold under the trade name Vitol. The fuel is an aqueous mixture of methyl and isopropyl alcohols. Work on similar lines, but using aqueous ethyl alcohol, is in progress at the Northern Regional Research Station of the Department of Agriculture.[+] The oil industry is understood to be mildly interested in the development. It is inconceivable, however, that the industry would contemplate establishing a market for alcohol in this way. It is much more likely that if

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[+] U.S. Bureau of Mines R.I. 4458.
Synthetic Liquid Fuels. 1948 Annual Report of the Secretary of the Interior. Part 3. Liquid Fuels from Agricultural Residues.

the practice threatened to spread, a secondary fuel, competitive with alcohol in performance and cost, will be prepared from petroleum and put on the market.

The main interest of the North Regional Station is the utilisation of farm wastes such as corn cobs, oat hulls, flax shives, bagasse etc. It is realised that chemical industry offers a market for many products which may be prepared from these raw materials, at a price much higher than that of alcohol. The Quaker Oat Company is currently producing furfural, worth 9½ cents per pound, from oat hulls in large quantities. The problem being studied at the Station is, therefore, the separate utilisation of pentosans and cellulose in corn cobs, under conditions chosen for optimum conversion in each case. In this way yields are obtained much higher than those realised in any single stage process applying compromise conditions of temperature and acid concentration.

The crushed corn cobs are first contacted with sulphuric acid of 5% concentration at 100°C for a period approaching one and a half hours. By this treatment the pentosans are hydrolysed substantially completely while only 5% of the cellulose is attacked. The liquor is neutralised with lime, filtered, diluted from 15% to 5% of sugar content and fermented to butanol, acetone and ethanol. About 30% of the xylose in the syrup may be crystallised by concentration, and furfural also may be recovered from it by acid treatment.

The pentosan-spent material is dried to a moisture content/to 3% of 50% sulphuric acid. The dried residue is disintegrated and further treated with sulphuric acid of 85% concentration. A pressure of 200 lb is applied to force the acid into the capillaries of the material. The pressure is released, water is added and the pulp is cooked for ten to thirty minutes with steam at 35 lb pressure. The pulp is then cooled and filtered to remove lignin. The filtrate is neutralised and filtered leaving a syrup ready for fermentation or other treatment. About 50% of dextrose may be recovered if desired; otherwise the hydrolysate may be fermented to alcohol or to butanol-acetone - ethanol. Both pentosan and cellulose syrups may be used for growing fodder yeast by inoculation with torula.

The current recovery of hexoses is about 80%. The great improvement upon the yield claimed for the Scholler process (55 to 60%) is attributed to the two stage hydrolysis, first with cool, strong acid in the impregnator, then with hot, weak acid in the cooker. The yield is improved, too, by restricting the sugar content of the liquor to 10%.

For the distillation of fermented liquors, the station has an azeotropic unit with a capacity of 500 gallons of alcohol per day. The entraining agent is ether and the azeotropic column is operated at a pressure of 135 lb. The beer still and rectifier are under vacuum and are heated by steam raised in the condenser of the azeotropic column. Steam consumption is said to be 23 lb per gallon, or little more than a third of that experienced in the more usual benzole dehydration plant.

Information was given to the effect that the wood hydrolysis plant, erected at Springfield, Ore. in the early days of the war, was operated for three weeks only. Owing to the low priority for materials which was accorded to the project, pipe lines were installed in ordinary mild steel. It would now cost $500,000 to replace those lines in stainless steel and there is no prospect of the work being done.

FLUO SOLIDS TECHNIQUE:

The Dorr Company has applied the fluidisation principle to chemical and metallurigical processes under the name of the Fluo-Solids[+] technique. A commercial plant supplied to British Columbia for the roasting of gold concentrates has been in operation for nearly three years. The oxidised concentrates are now cyanided on the spot and only bullion is shipped from the plant. The previous practice was to ship a 5 oz. concentrate to Seattle for roasting, at considerably greater expense.

+ (p.144)

In conjunction with the New England Lime Company, a triple-bed, fluidised lime burner has been developed. A small plant has been built and operated at the lime works, and a 100 ton per day unit is now being installed together with a fluid bed plant for preliminary drying and classification. For the drying process, limestone containing 4% of moisture is crushed to pass a 4 mesh screen. Products of combustion from a Dutch oven enter the dryer at 600°C through a grid which supports the fluid bed. A temperature of 150°F is maintained in the charge which is 18" thick. Dust carried out of the fluid bed is separated in a disengaging zone, a cyclone and multiclones in series. The fines separated in the cyclone and multiclones are not charged to the burner, but are sold instead as agricultural lime, so avoiding the subsequent loss of the same material in a more valuable form. The charge to the burner is the bottom discharge from the dryer.

In the three stage burner, the lime is first preheated in exchange with waste gases, then burned and finally cooled in exchange with the air required for combustion. All three operations are performed in fluid beds. The oil fuel is injected straight into the intermediate bed. Fuel consumption has been reduced to two thirds by adopting the fluo-solids technique. Because fuel is expensive at the works, this saving is a big consideration. The use of pulverised coal instead of oil has been contemplated and some work is planned on the subject. It will be necessary for the coal to save a considerable amount in fuel costs, if it is to replace oil, for it has the disadvantage of reducing the quality of the product by forming calcium silicate to the extent of the silica contained in the ash.

A project is being developed for the roasting of a Bolivian tin ore in a fluid bed. The ore contains two ounces of tin per ton as cassiterite, and sufficient pyrite to provide the heat needed for the process. The tin is volatilised and subsequently recovered, by cooling, in the form of the sulphide.

INDUSTRIAL CARBONS from COAL.

The Powell Duffryn Company is producing at Hayes, Kent, a wide range of industrial carbon products from coal. The coal, which may be chosen from many types, is pulverised and briquetted without a binder at a pressure of 10 tons per square inch. The briquettes are carbonised and then heated in an electric muffle furnace formed of a carbon cylinder. The rate and final temperature of heating are determined by the coal used and the product desired. For low ash carbons, the heating is continued to 2,700°C to reduce the ash by volatilisation to as little as 0.1%. The carbons made vary in hardness up to 8½ on Moh's scale; that is to say, the hardest of them scratch glass and topaz. Densities range up to 1.8. The product may be prepared in moulded, extruded and fabricated shapes. The tensile strength of the best carbon is 10 tons per square inch.

STORAGE of COAL:

At the Golden station of the Bureau of Mines, a technique has been developed for the safe storage of subbituminous and other coals subject to autogenous heating in stock piles. The procedure consists in laying down the coal in layers two to four feet thick, preferably

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(143) Kite, R.P. and Roberts, E.J. Fluidization in Non-Catalytic Operations Chemical Engineering, Dec.1947.

the former, and compacting each layer by driving a bulldozer across it before adding the next increment. This method has proved satisfactory in the storage of Colorado sub-bituminous coal at sugar mills, where coal is stocked for periods up to ten months awaiting the resumption of seasonal operations. Lignite is now being stored by this method at the Garrison dam (Missouri River, North Dakota) where several million tons won in the course of excavation work are to be held for some years, until a steam station is installed to consume it.

GRINDING of COAL:

Foster Wheeler has recently developed a new type of pulveriser, called in the organisation a "ring" mill. It comprises three horizontal rollers equidistant from one another with two above and one below. Each roller is shaped in the form of two conical frustra joined at their smaller ends. A number of rings encircle the rollers, picking up the coal just as lubricating oil is picked up in an oil ring bearing. The crushing is done between the rings and rollers. The performance of the machine is somewhat similar to that of a ball mill, but it has the advantage that power consumption is approximately proportional to the load, instead of being substantially independent of it.

The first commercial pulveriser of this type has been operating for about four years at Rochester N.Y. The mill has just been released for general manufacture and sale in U.S.A. only.

The Fuel Research Board and B.C.U.R.A. are collaborating on studies of power consumption in pulverisers. From a questionaire issued to electricity station operators it has been learned with some surprise that commercial consumption is substantially independent of coal hardness as determined by standard grindability tests. This anomaly is believed to be due to the load being determined by the rate of removal of coal from the grinding zone. Experiments are planned to test this theory.

For the estimation of the surface area of pulverised coal, B.C.U.R.A. has developed a device for counting the number of particles in a weighed sample. The pulverised coal trickles in a very thin stream from a jet through a narrow beam of light. Each particle intercepts the light falling upon a photo-electric cell. The impulses are registered on an instrument which counts at rates up to 100 per second. A typical sample weighing 15 milligrams and containing perhaps 100,000 particles can be counted in half and hour.