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RESEARCH ACTIVITIES OF THE
IMPERIAL FUEL RESEARCH INSTITUTE
KAWAGUCHI

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SUMMARY

This report records and summarizes technical data on the Imperial Fuel Research Institute, at Kawaguchi, Saitama Prefecture, Japan, by the Petroleum Section of NavTechJap during the period 16-28 January 1946.

The Institute is an agency of the Japanese Department of Commerce and Industry, engaged almost totally in research on the subject of utilization of coal substances native to the Japanese home islands. Its field of activity embraces studies on the production of both liquid and solid fuels from coals. Specifically, low temperature carbonization, coal hydrogenation and the Fischer-Tropsch Synthesis have been the principal media of approach to the production of such fuels.

The Institute has operated a six-retort carbonization assay plant since 1921. This installation has a throughput capacity of 6 tons per day. Non-cooking bituminous coals of Japan have been thoroughly studied in this plant. At present, interest is centered upon the carbonization of brown coals found principally in Central Honshu.

The Fischer-Tropsch plant was operated from early in 1938 to 1940, this work being discontinued when the MITSUI interests began full scale plant operation at MIKE. The Institute's plant never attained more than 30% of design capacity, which was 200kg of total crude oil product per day. Failure was partly due to poor catalysts, and partly to faulty and cumbersome converter design.

The coal hydrogenation plant was operated between 1936 and 1940, being abandoned when the FUSHUN and AGOCHI commercial coal hydrogenation plants began operations. The Institute's plant had a throughput of 3 to 5 tons of paste per day, representing 1.5 to 2.5 tons of coal input. It was operated only as a liquid phase unit, hence produced mainly heavy oil. When Japanese high-volatile low rank bituminous coals were processed in this plant at 200 atmospheres pressure, 80% or more liquefaction was obtained.

Although this plant apparently operated at rated capacity and with relatively little difficulty, no outstanding process or equipment developments were attained as a result of its operation. This was partly due to over-simplification in its design and partly to failure to incorporate the vapor phase step.

From 1944 until the end of the war, the Institute was under the administrative supervision of the newly formed Department of War Production, which abolished the Department of Commerce and Industry. At the end of hostilities, prewar status was regained. The war program of the Institute directly concerned with military activities comprised seven projects, none of which were reduced to commercial practice.

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I. JAPANESE PERSONNEL INTERVIEWED

The following personnel of the Imperial Fuel Research Institute were interrogated and supplied verbal information or submitted written documents from which this report was compiled.

Mr. Y. BAN	Director of the Institute.
Mr. T. SHIMURA	Assistant Director and Supervisor of the Fischer-Tropsch Group.
Mr. M. KUROKAWA	Supervisor of the Coal Hydrogenation Group.
Mr. N. SAITO	Supervisor of the Carbonization Group.

II. LOCATION AND GENERAL DESCRIPTION

The Imperial Fuel Research Institute (Nenryo-Kenkyusho), of the Japanese Department of Commerce and Industry, is located in the town of KAWAGUCHI, Saitama Prefecture, Honshu. KAWAGUCHI is situated just north of TOKYO, from which it is separated by the ARAKAWA River. The Institute covers an area of 483,624 square feet, all of which is quite level, hence completely utilizable for building construction. In this area are a total of more than forty buildings, of which about fifteen are totally devoted to laboratory and pilot plant operations and administration. Total building floor space, that is, laboratories plus shops, warehouses, service buildings, etc., amounts to more than 180,000 square feet. Pilot plants and some laboratories are housed in sheet iron structures; the administration building and some laboratories are concrete. Figure 1(E) is a layout plan of the grounds and buildings.

III. HISTORICAL

The Institute was founded in 1921, and at that time was under the jurisdiction of the Department of Commerce and Forestry. About 1923, this Department was reorganized and divided to form the Departments of Commerce and Industry, and Agriculture and Forestry. The Institute became part of the Department of Commerce and Industry, and so remained until 1944, when the Department was placed under military administrative supervision and given the name Department of War Production, as nearly as the new title could be translated. At the end of the war military supervision ceased, and the pre-war name and status were regained.

IV. GENERAL DESCRIPTION OF ACTIVITIESA. Normal Activities

At the time of its organization, the field of investigation assigned to the Institute was to study and promote processes for the economical utilization of coal substances native to the Japanese home islands. These comprise mainly low rank and low grade bituminous coals, brown coals and lignite. Their chemical nature is, therefore, such that they are not attractive to most industrial consumers. Research activities were later extended to include utilization of natural gas and petroleum.

Coal studies most intensively pursued have related to complete gasification, low temperature carbonization, and combustion in industrial furnaces. Before the war, the Institute's technologists were active in studying the direct gasification of coal in one step. They claim credit for developing processes now used in commercial plants serving TOKYO, YOKOHAMA, and KOBE. A continuous program of coal carbonization has been carried on, aiming toward the production of an acceptable substitute for charcoal for domestic heating and cooking. Institute technologists believe that general acceptance of low temperature semi-coke or "coalite" from brown coal would practically revolutionize the Japanese home-fuel

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situation. In the rural districts and towns of moderate size (i.e., less than 50,000 population) in central and southern Japan, wood and wood charcoal are universally used for both heating and cooking; also a large proportion of the people in the larger cities use charcoal exclusively. In TOKYO, for example, it is estimated that about 40 per cent of the homes use no fuel other than charcoal. Successful application of low temperature carbonization to this market would also make available large quantities of low temperature tar, which is particularly valuable in Japan as a petroleum substitute.

Because of Japan's poor position with respect to natural petroleum, the Institute has conducted research on synthetic liquid fuel production from coal by the Bergius and Fischer-Tropsch process since 1931 and 1933, respectively. However, no industrial process or equipment applications have evolved from this work, and experimentation has dwindled since 1940. Present personnel state that the Japanese government has withdrawn support of these projects because of the pressing need for work in more direct utilization of Japanese coals during the reconstruction period.

Studies on the utilization of Japanese raw materials other than coal, gas, and petroleum have been very limited. The Institute is in no way connected with the metallurgical or mineral industries. A small amount of work is done on agricultural wastes (such as straw and chaff), and forestry products, particularly in problems dealing with obtaining liquid fuels from such sources.

A continuous program of coal assay is carried on for the benefit of both public interests and the industries. Similarly, coal sampling, coal dressing, studies of coke properties, coal combustion tests, tar treatment and the like are continuous problems. No fundamental research is done which is not directly connected with fuel problems.

B. WAR ACTIVITIES

Present personnel state that throughout the war the major part of the Institute's work continued to be along the lines of fuel production for civilian or normal industrial use. A few specific war projects were assigned when the military group was in control (1944-1945), the general subject titles of which are the following:

1. Hydrogenation of Daido Coal.
2. Low-Temperature Carbonization of Coaly Shale.
3. Carbonization of Pine Wood.
4. Preparation of Aviation Gasoline from Pine Root Oil.
5. Catalytic Cracking of Low-Temperature Tar.
6. Synthesis of Triptane.
7. Explosive Combustion of Coal Dust.

Results of this work are summarized in Section IX.

C. PROPOSED FUTURE ACTIVITIES

As an outcome of the war, Japan has lost her most valuable coal fields of former days, that is, in Korea, Manchuria, and China. She has also lost a fairly important source of oil in the poor quality shale deposits of FUSHUN. These circumstances are cited as evidence of the necessity for concentrating to a greater degree than in the past on extending the fields of utilization of native Japanese coals and related materials.

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V. PRESENT PERSONNEL ORGANIZATION

Total personnel now numbers 121, which is considered the normal complement. The employees are classified as follows:

Director	Mr. Y. BAN
Assistant Director	Mr. T. SHIMURA
Mechanical Engineers	2
Physicists	1
Research Chemists	18
Assistants	25
Clerks	3
Laborers and Operators	70

The laborer classification includes skilled as well as unskilled workers.

During the time when the Institute was under the control of the Army and Navy, personnel was increased to 166, classified as follows:

Director	Mr. Y. BAN
Assistant Director	Mr. SHIMURA
Research Chemists & Engineers	25
Assistants	38
Clerks	3
Laborers	98

The annual budget for a normal year is 500,000 yen, of which 200,000 yen is paid out in salaries. The average wage of the professional man is 3,000 yen per year, the assistant 2,000 yen, and the laborer 5 yen per day. Salaries have recently been increased by 10%, this increase being granted to offset present currency inflation. The Institute is financed entirely by government funds, administered by the Department of Commerce and Industry.

VI. COAL HYDROGENATION PROCESSESA. Laboratory Work

The first experiments on direct coal hydrogenation began in 1931. The principal object of this work was to study in a comprehensive manner the hydrogenation of Japanese coals, the results of such experimentation to be applied to development of a commercially-feasible synthetic liquid-fuel process. The statistical technical information obtained on the behavior of the various coals was to be made available for general distribution to those interested in development of industrial hydrogenation. It was considered the duty of the government to provide such information for the industry. This work was carried on as a continuous project until about the last two years of the war, and has involved a very large number of tests using high pressure autoclaves with charging capacities from $\frac{1}{2}$ liter to about 5 liters.

More specifically, the purpose of these investigations may be outlined thus:

1. To study a wide variety of Japanese coals with respect to their suitability for liquefaction by direct hydrogenation. Over 100 coals have been assayed.
2. To study the effect of catalysts on the liquefaction reactions.
3. To study the kinetics of depolymerization-hydrogenation reactions.

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4. To study hydrogenation of the complex separable constituents of coal.
5. To study vapor phase hydrogenation of products obtained by primary liquefaction or dry distillation of coal.

Results of this work have been prepared for distribution in the form of technical papers and bulletins, some have also been published in technical journals. Copies of all these bulletins (written in Japanese) were obtained, but not translated except for their titles, which are as follows:

1. The Hydrogenation of Phenols, Part I.
2. Studies on the Liquefaction of Coal, Part II. (Test Results of Japanese Coals)
3. The Hydrogenation of Low-Temperature Tar.
4. The Hydrogenation of Heavy Oils.
5. The Hydrogenation of Phenols, Part II. (On the Preparation of Hydrocarbons from Low-Temperature Tar Acids).
6. Studies on the Liquefaction of Coal, Part III.
7. The Preparation of Benzol from High-Temperature Tar by Hydrogenation.
8. Studies on the Liquefaction of Coal, Part V. (Action of Catalysts on the Coal Paste)

(ATIS and ND numbers for these publications will be found in Appendix I of this report).

The material contained in these bulletins is of general technical and scientific interest, and so has bearing on the broad subject of coal hydrogenation, but cannot be said to be the basis for any specific process, or improvements applicable to any specific process.

According to the present personnel, the most important general conclusions obtained from this work are:

1. Practically all coals of the Japanese islands were found suitable for hydrogenation. In general, Hokkaido and Sakhalin coals are more easily liquefied than those of the central and southern regions of Japan.
2. The action of catalysts on coal hydrogenation is two fold:
 - a. Acceleration of the reactions of depolymerization-hydrogenation. Catalysts effective for this purpose are oxides and sulfides of tin, lead, and nickel.
 - b. Acceleration of hydrocracking reactions. Catalysts suitable for this purpose are oxides and sulfides of molybdenum, zinc, iron, and copper. Halides capable of acting as a source of hydrogen halide at elevated temperatures are also extremely effective hydrocracking catalysts.
3. Mixtures of hydrocracking catalysts (such as MoO_3) with alumina gel or with halides (such as NH_4Cl) were more effective than either used alone. The alumina gel is said to catalyze decomposition of asphaltic bodies. Depolymerization catalysts (such as SnO or PbO), when mixed with halides (such as NH_4Cl or CHI_3), were likewise more effective than either component alone.

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B. Semi-Plant Work

In 1934, the Institute began the operation of a semi-pilot-plant scale liquid-phase coal hydrogenation plant, capable of treating 3 to 5 tons of paste per day. Usual practice was to use paste in which the ratio of coal to vehicle was 1:1, hence the amount of coal treated was 1.5 to 2.5 tons per day. The product was heavy oil which, in a complete hydrogenation plant, would have been split into two parts, one going to the vapor phase step, and the other (after separation of solid matter), to vehicle storage for paste making.

The entire hydrogenation system except the hydrogen compressor was designed and built by the Kobe Iron Works. The operating staff consisted of 1 research chemist (group leader), 3 assistants, 5 operators, 2 analysts, 1 producer gas operator, and 1 hydrogen plant operator.

As a matter of record, this is Japan's second hydrogenation plant, with respect to age. The oldest is the one built at TOKUYAMA and later moved to OFUNA.

The object of experimentation with this plant was to compare plant and laboratory results on the hydrogenating characteristics of a few representative Japanese coals which gave good liquefaction yields in laboratory tests, and also to determine whether Japanese-engineered and Japanese-built plant equipment could be made to serve in this particular high pressure field.

It was planned, if satisfactory performance was obtained, to continue expanding the scale of operation to a demonstration plant of commercial size.

This plant was operated for a total of about 5000 hours between 1934 and 1940. Its operation was discontinued when full scale plants began running at FUSHUN and AGOCHI. There seemed to be no immediate need for further pilot plant experimentation by the government.

The flow diagram of the plant is shown in Figure 2(E). Process hydrogen was manufactured by electrolysis of water in a small Knowles generator having a capacity of 16 m³ per hour. Low pressure hydrogen storage consisted of a gasholder system of 115,000 cu. ft. total capacity. A ballast vessel of 110 liters free volume was placed between the compressor and the hydrogenation system. Producer gas made in a Karpely generator was used for all process heat not furnished by electrical resistance.

Paste making was a batch operation, enough being made at one time for one day's throughput (3 tons). Coal preparation equipment consisted of a jaw crusher and a pan mill. There was no drying equipment. A screw-type conveyor transported the pulverized coal (minus 200 mesh) to the paste mixers, which were elevated so that their bottom outlets were about 6 feet from the floor. Paste mixing and storage equipment consisted of 2 open top cylindrical vessels with conical bottoms, fabricated from steel plate. They were heated by internal steam coils and provided with vertically mounted motor-driven propeller type stirrers. The paste injector consisted of two single-acting hydraulically operated reciprocating piston type units in parallel, with variable speed stroke. Paste feed to the injector was by gravity.

The hydrogenation system consisted of an indirect gas-fired tubular paste preheater, two converters in series, electrically heated hydrogen preheater, a separator-cooling system and a gas scrubbing unit. Materials of construction and significant dimensions of this equipment will be found on

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Figure 2(E). The converters and paste preheater were enclosed in brick-work housings, through which pass the hot gases from the combustion chamber in order to maintain proper operating temperature. The entire hydrogenation system was housed inside the plant building. Protection was provided for the valve operators in the form of a 9 inch thick re-enforced concrete wall (with small glass windows) which separated the pressure vessels from the control room. Valve wheels were fixed to this wall in the operating room, and were connected to the valve stems by means of a shaft and gear arrangement.

Heavy product discharge was manual and intermittent. The heavy oil was separated from solid matter as completely as possible by gravity in the first high pressure separator, the remainder of the oil being removed by a distillation-carbonization operation. Light product carried through the separators by the exit gases was recovered in the oil scrubbing system, employing a creosote oil fraction (boiling range 230-300°C) as the absorbing agent. This unit operated at atmospheric pressure. The light oil was recovered by distillation.

There was very little instrumentation in the plant. All flow controls and product discharge controls were manually operated. There were no mechanical safety devices on moving machinery except for open-type safety valves on the hydrogen compressors. Likewise, the only enclosed motor was on the hydrogen compressor.

The converters and preheater were designed to operate at 200 atmospheres. The remainder of the plant featured extra-heavy-duty equipment, particularly valves, piping, and the paste injector. Such over-design resulted in reasonably infrequent replacement of the parts receiving most wear. However, it was admitted that heavy oil discharge valves suffered severe erosion, requiring change of the working parts about every two weeks. These valves were fairly wide-angle needle type, with stellite needle tip and stainless steel seats. There had been no failure of preheater tubes, but these were changed from time to time because of rusting due to long periods of idleness.

Other features, which simplified operation and thus added to reliability of performance, were incorporated into the plant. The most important of these, from the engineering standpoint were:

1. Omission of heat-exchange from the paste injection system.
2. Direct condensation of vapors from converter gases, without heat exchange.
3. Direct preheating of process hydrogen.
4. Omission of the step of recirculating part of the hydrogen to process after partial stripping of the exit gases under pressure.
5. Use of electrolytic hydrogen for process.

The only operating difficulty admitted by the staff, aside from the inadequacy of instrumentation, was the heavy oil discharge valve failure previously mentioned.

Experimentation was confined to hydrogenation of three Japanese coals which laboratory tests had indicated to be most suitable for liquefaction, and consisted in:

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1. One month running on Iriyama coal from central HONSHU, a coal lying in rank between lignite and sub-bituminous.
2. Two months on Harutori, a low-rank high-volatile bituminous coal from HOKKAIDO.
3. Two months on Kawakami, a low-rank high-volatile bituminous coal from SAKHALIN.

This work led to the following general conclusions:

1. The three coals tested were readily hydrogenated in the plant at 200 atmospheres pressure.
2. Coals containing as much as 10% ash presented a serious valve problem in discharging heavy product.
3. Zinc chloride was found to be a superior catalyst for hydrogenating low-rank coals. There was no mention of corrosion of HCl.
4. Japanese pressure equipment and machinery showed no particular weakness in performance at 200 atmospheres.
5. No specific improvements of value to the coal hydrogenation industry were developed, either with respect to process or equipment.

The only publication on the operation of this plant is a short paper in the "Journal of the Society of Chemical Industry, Japan", Volume 44 No.10, dated 1941, entitled "On the Hydrogenation of Coal by Semi-Technical Scale Plant".

The data of Table I(E) were submitted as a typical record of operating results.

VII. FISCHER-TROPSCH PROCESS

A. Laboratory Work

Laboratory experiments on the synthesis of hydrocarbons by the Fischer-Tropsch process were begun in 1933. The purpose of the work was to provide Japan with a source of liquid hydrocarbon fuels from raw materials available within the limits of Japan proper. It was the goal of the Institute to develop and promote the full scale exploitation of a complete system of synthetic petroleum manufacture by this method, with particular emphasis placed on catalyst development, converter design, and possibly gas generation.

The key man in the organization was Dr. S. TSUTSUMI, who remained at the Institute from 1933 to 1939. According to present personnel, he personally carried out all the more important work concerning catalyst preparation, catalyst testing, and evaluation of results. When he left the organization in 1939, he took with him all notes and other records concerning his work, leaving nothing to guide his successors in continuation of the program. While at the Institute, he prepared one bulletin describing his work up to 1934 or 1935. This publication has not been translated, except for its title, which is "The Synthesis of Gasoline (Part I)". (The ATIS and KD number of this document will be found in Appendix I of this report).

From the results discussed in this publication, and from the statements of present personnel, the substitution of nickel for cobalt was the object of

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much of TSUTSUMI's research. The reason for this was the high cost of cobalt. Good yields were obtained from nickel catalysts over short periods of time, i. e., well above 100 grams per cubic meter of gas input, but the catalysts could not be maintained in an active condition for longer than about a month. Even during this short period they required two high-temperature activations. This performance is not suited to a commercial process.

TSUTSUMI also conducted extensive experimentation on the promoter effect of catalytic materials such as oxides of uranium, thorium and uranium on nickel and cobalt catalysts. His results led to the conclusion that thorium and uranium are most effective for this purpose. Miscellaneous research included studies on the effect of changing the ratio of H₂ to CO in the synthesis gas, the effect of changing the linear gas velocity through the catalyst, the effectiveness of various bulking agents, and studies on the effect of changing the amount of bulking agent. He did no work on iron catalysts.

These experiments were performed at atmospheric pressure, using an electrically-heated single tube converter 20mm inside diameter and 80cm long. There is no record of experiments at intermediate pressures, although some work of this kind was done.

B. Semi-Plant Work

Experiments began on a semi-plant scale in 1936. As a matter of record this was the year in which the MITSUI interests purchased the rights to exploit the Ruhrchemie Fischer-Tropsch process in Japan. It is claimed that the Japanese license was limited to operation at atmospheric pressure, hence the Institute concentrated upon developing a process, operating at pressures above atmospheric. Specifically, 5 atmospheres gauge pressure was arbitrarily chosen for experimentation.

The first converter was a failure, consisting merely of a tube 30cm in diameter and about 2 meters long, without provision for removing the heat generated by the highly exothermic hydrocarbon synthesis. In 1938, operation began on a plant which had been provided with a converter of more appropriate design with respect to heat-control characteristics. Temperature control was effected by circulating water through a system of narrow tubes in parallel, the catalyst being packed in the interstices between the tubes. Design production capacity was 200 kg total crude oil product per day, to be obtained from a throughput of 100 normal cubic meters of synthesis gas per hour with an H₂:CO ratio of 2:1. The catalysts were composed mainly of nickel. Actual composition of the catalyst reported most effective was (in parts by weight) Ni : Co : Cr : Kieselguhr as 40:10:10. 5:150. The catalyst charge was said to be one cubic meter bulk volume but actual calculation indicated 3/4 cubic meter to be more nearly correct. Oil production never exceeded 30% of design capacity, according to the records offered as representative of best operating practice. Throughput was likewise about 30% of the design figure.

Performance of catalysts was unsatisfactory with regard to their useful life, although while active, yields as high as 118cc (78 gm) of pentane and heavier hydrocarbons per cubic meter of synthesis gas were obtained. The longest total period of useful activity observed was about a week, this being obtained only by reactivation with hydrogen on the third and fifth days. This type of catalyst had been maintained in an active condition in laboratory tests for about a month. The explanation given for the shorter life in the plant was sulfur poisoning. Spent catalysts were not reworked.

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This plant was operated intermittently from early in 1938 until near the end of 1940. During this time, no changes were made in the plant equipment. Operations ceased when the Mitsui plant at MIIKE began running. This action was explained as partly due to belief that the Miike plant was practically certain of success, and partly because it was realized that modification of the Institute's converter design to give better temperature control would introduce considerable additional expense in its fabrication, and partly because of the unsatisfactory life of the catalysts.

During the time the plant was active, it was estimated that 12 runs of from 1 to 2 weeks duration were made per year for a period of nearly 3 years. The plan followed was to make one run per month.

There was no work at the Institute on refining of the crude oil product or on engine performance of Fischer-Tropsch fractions from any source.

A flow sheet of the plant is shown in Plate I(E). The converter was of the single-pass downflow type, designed to operate at 10 atmospheres gauge pressure, but actually operated at 5 atmospheres. It consisted essentially of a steel cylindrical shell 116cm in diameter and 150cm long, inside which were placed, longitudinally, 860 small steel tubes described as $\frac{1}{2}$ inch "gas pipe". The shell was provided with one integral and one removable head, the latter being at the top. The tubes were connected in parallel by means of an intricate welded header system at top and bottom with pipe connections passing to the outside through removable packing-gland connections in the heads. The entire pipe assembly could be removed and again set in place for the operations of charging and discharging catalyst.

Hot water, at substantially the temperature of the synthesis (180-200°C), was mechanically circulated through the tubes and an external closed condenser system, for controlling catalyst temperature. The catalyst, in pellet form, was loosely but uniformly packed in the interstices between the tubes, which were spaced 35mm between centers. This design is essentially the reverse of the American Downs converter.

The product recovery system comprised:

1. An air-cooled pressure receiver, immediately following the converter, in which water and the heavier hydrocarbon products were collected.
2. A water-cooled shell and tube-type pressure condenser to collect part of the lighter reaction products.
3. A batch-type packed tower pressure scrubber employing a creosote oil fraction boiling in the range 230-300°C for the final stripping of light products from the gases.
4. A continuous steam distillation tower and condensing unit, operating at atmospheric pressure, for stripping light product oil from wash oil.

Exact records of catalyst preparation was not available. From memory, present personnel recalled that all catalysts used in the plant consisted principally of nickel, and the one giving the best results had the approximate weight composition Ni:Co:Cr:Kieselguhr as 40:10:7.5:50. In preparing this catalyst, nitrates of nickel, cobalt, and chromium were dissolved in hot distilled water in the proper ratio to give a final product of the desired composition, the proper amount of kieselguhr added, then, with agitation of the solution, the metals were precipitated by addition of a

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hot solution of potassium carbonate slightly in excess of the stoichiometric requirement. The precipitate was washed with hot distilled water until no alkalinity was detectable with phenolphthalein indicator. The slurry was then filtered, dried, and the product pelleted. Pellet size was 7mm in diameter and 2.5mm thick at the edge. Pellet faces were convex. Temperatures, time schedules, and solution concentrations during these operations could not be recalled.

Catalysts were all prepared in the laboratory, using glass and stoneware apparatus for the precipitation reactions. Approximately a month was consumed in making a catalyst, requiring the services of 8 persons, working 9 hours daily.

In conditioning for the synthesis, the catalyst was reduced at atmospheric pressure in the converter, with hydrogen. Bayonet-type electric heaters, inserted in sheaths extending from top to bottom inside the converter, supplied the heat required to attain the reduction temperature of 350°C. The reduction schedule was described as follows: Hydrogen was passed over the catalyst while, at the same time, the temperature was increased from atmospheric to 350°C, over a 20-hour period. After 10 hours at 350°C, the temperature was decreased, over an 8-hour period, to 180-190°C. Synthesis gas was then substituted for H₂, and proper operating pressure and flow rate attained at once to start the synthesis.

The method for preparing gas for the synthesis consisted in blending blue water gas with electrolytic hydrogen. The blue gas generator was a small unit which was operated intermittently, gas being prepared and stored in a 30,000 cu. ft. gasholder. Similarly, H₂ was stored in a 10,000 cu. ft. holder. The two were mixed, as required, in another 10,000 cu. ft. holder. There was no storage of compressed gas except an accumulator, about 20 inches I. D. and 42 inches long, immediately following the compressor. All the plant operation was carried out with a synthesis gas of the approximate composition H₂:CO as 2:1.

Because of the sensitivity of the synthesis catalyst to sulfur poisoning, an elaborate but unconventional system for sulfur removal was adopted. Following the blue gas generator was a conventional dry iron-oxide box (to remove all but traces of H₂S), from which the gas passed directly to the water gas holder. Organic sulfur removal occurred after blending and compression for the synthesis. For this step, the gas was passed through a system of five cylindrical steel tubes 30cm in diameter and 2m high, filled with random-packed rectangular pieces of copper gauze (10cm x 20cm, about 10 mesh) coated with a mixture of 90% Ni and 10% Cu. The amount of Ni-Cu mixture in each vessel was about 15 kg. This mixture was prepared by precipitation from the nitrates with sodium carbonate, and was pasted on the gauze while still wet. It was conditioned for use by reduction in the desulfurizers with hydrogen at 350°C. The temperature of operation for desulfurization was 200-250°C.

The five units containing the desulfurizing agent were arranged so that only four were used at any given time, one comprising the first stage and the other three, the second. The fifth was a first-stage spare. A brickwork housing enclosed the units, except for about 6-8 inches at the top and bottom. The housing also served as a combustion furnace chamber, in which stoker coal was burned to supply heat to maintain the required operating temperature. Once fouled, the Ni-Cu mixture could not be regenerated.

The amount of sulfur in the gas entering this unit could not be ascertained, but it was stated that with an active, fresh charge, the sulfur content, after passing through this system, was about 0.02 grain/m³.

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The synthesis plant was without instrumentation to control pressure, gas flow, or recovery operations. Temperature control in the converter was essentially automatic through the action of the condenser in the water circulation system.

The data of Table II(E) was submitted as operating results of one of the best runs.

VIII. LOW-TEMPERATURE CARBONIZATION

Ever since its organization in 1921, the Institute has carried on a continuous program of coal-carbonization studies. The tar has always been recognized as the most important product of the process, but tar production alone cannot carry the expense. It was, therefore, necessary to give attention to the production of a semi-coke end product which would have a ready sale. The coke comprises 65% or more of the weight of the coal charged.

The most attractive potential market for the coke was as a substitute for charcoal as a domestic heating and cooking fuel. Although its characteristics are excellent for burning in domestic furnaces, it was not found applicable to the braziers used in Japanese homes, because of high ignition temperature. It is not usable for metallurgical purposes, and too expensive for boiler use, therefore, a dependable market was never found. Rather recently, some progress has been reported in blending it with coal for coke making. During the war, it was planned to use it to some extent for making water gas and hydrogen as raw materials for synthetic liquid fuel processes.

Carbonization studies at the Institute were carried out both on test-tube scale and in an assay plant having a throughput capacity of about 6 tons per day. A flow sheet of the plant is shown in Figure 2(E) and a drawing of the retorts in Figure 3(E). The retort system, which was designed at the Institute, comprised 6 vertical chambers of rectangular cross section, the carbonizing sections of which are constructed of fireclay tile, externally heated by combustion of producer gas or gas from low-temperature carbonization. Carbonizing temperature is 500°C, the skin temperature on the outer wall of the retort being 700°C.

The retorts are connected in parallel with the recovery system for gases and distilled products, which comprises a water cooler, a Pelouze tar extractor, a Hurdle ammonia scrubber, a gas meter, and gasholder. In this manner, if all 6 retorts are operated simultaneously, total coke and gas production are averaged as though 6 separate tests had been made. Coal is intermittently fed by hand, and coke is carried away continuously by a conveyor.

Three bulletins relating to the carbonization work, have been prepared for distribution, the titles being as follows:

1. "Japanese Coals as Raw Materials for Low-Temperature Carbonization" (out of print).
2. "Report of the Experiment on the Low-Temperature Carbonization of Coal".
3. "The Experiment of the Continuous System for the Low-Temperature Carbonization of Coal" (ATIS and ND numbers of the last two of these publications are given in Appendix I of this report).

The carbonization group is now emphasizing the utilization of brown coal. The full extent to which this may develop cannot yet be estimated, but it is strongly contended that use must be found for Japan's brown coal, the reserve of which is estimated at 500,000,000 metric tons. The deposits are located principally in northeastern and central HONSHU. This substance resembles

ENCLOSURE (E)

American lignite more than it does the brown coal of Germany. It has a low calorific value, and because of its structure and composition weathers rather severely.

At present, tar from brown coal carbonization is visualized as a petroleum substitute, and the carbonization residue as a substitute for charcoal. It has already been found possible to make a "coalite" with reasonably satisfactory ignition characteristics, at about half the price of charcoal. However, the product so far obtained emits a slight but objectionable odor on combustion.

The data of Table III(E) were submitted as representative yields of tar and gas by low-temperature carbonization of typical Japanese coals.

IX. SUMMARY OF REPORTS DESCRIBING WAR ACTIVITIES

The Institute was under military control from 1944 until the end of the war, and hence was subject to complete reorganization of personnel and activities. However, personnel was increased by only 45 persons (classified as 5 research chemists, 13 assistants and 28 laborers), and hostilities ended to soon for complete conversion of the program to specific war work.

Seven reports were submitted to NavTechJap as covering the complete war program of the Institute. All except one of the investigations described in these reports had its beginning in 1944. The exception is the hydrogenation of Daido coal, which was studied in about 1941. The reports are in Japanese, but with each was submitted an English summary. Following is a digest of the summaries, amplified in some instances by interrogation of Institute personnel. (ATIS and ND numbers will be found in Appendix II of this report).

A. "Hydrogenation of Tatung (Daido) Coal", by M. KUROKAWA and S. ANDO.

The purpose of this investigation was to determine the suitability of this coal as the raw material for establishing a coal hydrogenation plant in China. The Institute's 3-ton-per-day pilot plant was used for the liquid-phase hydrogenation and a 5-liter high-pressure batch-type rotating autoclave for the vapor phase.

The experiment comprised three parts:

1. Coal was hydrogenated in the liquid phase to heavy oil.
2. Heavy oil from (1) was batch-hydrogenated as the first cycle of the vapor stage.
3. Oil heavier than gasoline from (2) was batch-hydrogenated as the second cycle of the vapor stage.

Operating conditions in the liquid phase were 200 atmospheres pressure, a 410°C operating temperature, with a catalyst consisting of SnCl_2 , equivalent to 1%, by weight, of the paste. Conditions in the vapor phase were 240 atmospheres pressure, a 460°C operating temperature, 1 hour of contact, and a catalyst consisting of 10 parts MoO_3 to 1 part of sulfur, added in an amount equal to 10%, by weight, of the oil charge.

The overall results are given in Table IV(E).

It was concluded that the maximum gasoline production from this coal would be about 3% of the weight of the coal, with a total liquefaction of 60% of the coal.

ENCLOSURE (B)

B. "The Low-Temperature Carbonization of Coaly Shale" by N. SAITO

The object of this project was to build and operate a low-temperature carbonization plant to treat 20 tons, per day, of coaly shale, such as is found in the western part of HOKKAIDO and in parts of HONSHU and KYUSHU. The product distilled from the shale was to serve as a petroleum substitute for production of aviation gasoline. The plant was built and preliminary operation began August 10, 1945, but was stopped with the cessation of hostilities.

C. "The Carbonization of Pine Wood", by N. SAITO

The purpose of this problem was to study the low-temperature carbonization of pine wood, as a source of tar for conversion to gasoline. A tar yield of 10%, by weight, of the wood charge was obtained in laboratory experiments. However, when the experiment was repeated in the Institute's 6-retort pilot plant, the yield was only 4.38%. The work was not continued long enough to determine whether the laboratory yield could be achieved in plant practice.

D. "The Preparation of Aviation Gasoline from Japanese Pine Root Oil",
by T. AMEMIYA

When the gasoline fraction from pine-root oil was subjected to catalytic vapor phase treatment at 400-500°C, using as catalysts such substances as Japanese acid clay, activated acid clay, Kanuma earth and yellow earth, gumming tendency was corrected and a fuel with high octane value was obtained. However, the vapor pressure of the product was low, so it was usable only as blending stock.

Treatment of both the gasoline fraction and the higher boiling components of pine root oil at higher temperatures resulted not only in the production of lower-boiling constituents, but also in excessive decomposition. It was concluded that there is no better process for aviation gasoline production from pine root oil than to treat in a continuous system at 500°C, removing the lighter fractions as they are formed, although their instantaneous concentration is small.

E. "The Catalytic Cracking of Low-Temperature Tar", by M. KUROKAWA,
N. ASAOKA, and K. SUGAWARA.

A fraction from low-temperature tar, boiling in the range 200-300°C, was solvent-extracted with phenol containing 20% water. The raffinate was used as a charging stock for catalytic cracking, using pelleted commercial activated clay as the catalyst.

The content of gasoline in the cracked distillate was found to increase with increasing temperature, but the yield on charging stock decreased above 550°C.

F. "The Synthesis of Triptane", by M. KATSUNO

Chemically pure trimethyl butane was prepared in the laboratory to investigate its properties. It was synthesized in the following manner:

Pinacoline and methylmagnesium bromide were subjected to the Grignard reaction, and the alcohol thus produced dehydrated to 2, 3 + 3 methylbutene -1, which was hydrogenated to 2, 2, 3-trimethylbutane.

ENCLOSURE (E)

As the industrial method of preparation, alkylation of isobutane and propylene was proposed. Only a few preliminary experiments were carried out, using phosphoric acid and $AlCl_3$ as catalysts. The problem was discontinued before the products could be examined.

G. "The Explosive Combustion of Coal Dust in Air", by T. INATOME

The ultimate object of this investigation was the development of explosives whose ingredients are powdered coal and air. Three series of preliminary experiments were performed in the laboratory.

1. Inside a 19mm I.D. silica tube was compressed a hollow cylindrical plug of coal dust. The hole through the plug was 6mm in diameter, the length of the plug, 55mm. The silica tube, containing the coal dust was suddenly introduced into a tube furnace heated to 800-1000°C, and oxygen was passed through the hole in the plug. Carbonization, combustion of volatile matter, and combustion of fixed carbon occurred in the order mentioned, and at a rate which was slow and irregular, because of the slow rate of heating of the pellet.
2. A tube containing the hollow plug described above was placed in a cold zone (below 100°C) outside the tube furnace, but coaxial with it and near enough to the hot zone to be strongly affected by the radiation from it. Oxygen was passed through the hole in the plug, as before. In this experiment, radiation from the hot zone caused ignition of the plug at the end nearest the furnace, and combustion proceeded until the plug was consumed, but the reaction rate was slow.
3. If oxygen and coal dust were mixed in the stoichiometric proportions for complete combustion before passing into the tube furnace, the rate of reaction was of the order of the speed of an explosion.

ENCLOSURE (E)

Table I(E)
HYDROGENATION OF HARUTORI COAL
IN THE SEMI-PLANT

Type of Coal: High volatile bituminous.
Proximate Analysis: (%)

Moisture	5.44
Ash	13.3
Volatile Matter	42.56
Fixed Carbon	38.7

Ultimate Analysis (Dry Basis): (%)

Ash	14.07
C	62.59
H	4.87
O	17.23
S	0.42
N	1.07

Heating Value 6250 cal/gm

Weight ratio coal: vehicle	1:1
Catalyst	Nickel Oxide
Rate of paste input	157 kg/hr
Hydrogen input	146 m ³ /hr (N.T.P.)
Mean reaction temperature	410°C
Mean reaction pressure	200 atmospheres
Duration of run	about 2 months
Degree of coal liquefaction	86.2%
¹ Heavy oil production	788.3 kg/metric ton paste (60.08% dis- tilling up to 300°C)
² Light Oil production	39.9 kg/metric ton paste
³ Volatile oil production	24.6 kg/metric ton paste
⁴ Residue	81.5 kg/metric ton paste
Gas	27.8 kg/metric ton paste
Loss	48.0 kg/metric ton paste
H ₂ consumed	10.1 kg/metric ton paste

Note

- (1) Heavy oil : product boiling above 200°C
 (2) Light oil : product boiling up to 200°C, condensed
 by air and water cooling in recovery
 equipment preceding oil scrubbing
 (3) Volatile oil : low boiling product by oil scrubbing of
 exit gases
 (4) Residue : unliquified carbonaceous material and
 ash

ENCLOSURE (E)

Table II(E)
RESULTS OF TEST RUN OF
FISCHER-TROPSCH SEMI-PLANT

Catalyst	Ni:Co:Cr:Kieselguhr, 40:10:7.5:150	
Working pressure	5 atm. gauge	
Temperature	195°C	
Vol. inlet gas	931 m ³	
Duration of run	30 hrs	
Average rate of gas flow	31 m ³ /hr	
Oil yield from condensers	61.83 lit. (sp. gr. 0.7583 (20°C) Oil-fines 5%(vol.)	
Oil yield from scrubber	28.04 lit. (sp. gr. 0.7248 (20 C) Oil-fines 2%(vol.)	
Total oil yield	89.87 lit.	
Total yield/m ³ inlet gas	117.6cc (inlet gas corrected for components other than H ₂ and CO)	
Total water	161.12 lit.	
Vol. exit gas	298 m ³	
Contraction	68%	
Gas Analysis		

	Inlet Gas	Exit Gas
CO ₂	2.670%	12.0%
O ₂		
CO	27.4	10.0
H ₂	54.6	19.5
CH ₄	0.7	12.3
N ₂	14.7	46.2

ENCLOSURE (E)

Table III (E)
YIELDS OF CARBONIZATION PRO-
DUCTS FROM JAPANESE COALS

Name of Coal	Type of Coal	Proximate Analysis						
		H ₂ O	Ash	Vol. Matter	Fixed C	Heating Value (Cal/gm)	Tar Yield*	
Horonai	Non-coking bituminous	3.41	10.94	40.77	45.33	7,035	15.38	
Okinoyama	Sub-bituminous	10.79	14.03	49.93	25.95	5,622	7.78	
Kidomo	Brown Coal	27.67	14.71	34.70	22.92	3,667	4.00	
Name of Coal	Tar Analysis (wt %)					Sp.Gr.	Heating Value (Cal/gm)	
	Org. Acid	Phenols	Bases	Neutral Oil	Ether Insoluble			
Horonai	2.53	18.18	3.33	75.39	0.57	0.9656	9,249	
Okinoyama	10.92	19.49	1.85	66.93	0.81	0.9668	9,398	
Kidomo	11.46	20.85	0.34	53.23	14.12	0.9811	8,730	
Name of Coal	Gas Yield (m ³ /Metric Ton)	Analysis of Gas						
		CO ₂	Illuminants	O ₂	CO	H ₂	CH ₄	H ₂ O
Horonai	700	6.1	3.7	1.1	8.9	21.0	48.6	10.4
Okinoyama	1,070	21.3	3.8	0.8	10.2	21.2	38.6	4.1
Kidomo	726	3.3	1.2	1.2	15.8	11.2	24.3	2.5

* On basis of coal charged (wt %)

ENCLOSURE (E)

Table IV(E)
HYDROGENATION OF DAIDO COAL

	Input (kg)		Product (kg)	
1st Stage	Coal	1,000	Heavy Oil	550
	H ₂	22.4	Light Oil	90.6
			Light Hydrocarbons	18.6
1st Cycle 2nd Stage	Heavy Oil	550	Middle Oil	112.3
	H ₂	31.4	Refined Gasoline	297
			Light Hydrocarbons	93.2
2nd Cycle 2nd Stage	Middle Oil	112.3	Middle Oil	23
	H ₂	6.4	Refined Gasoline	54.8
			Light Hydrocarbons	18.4

ENCLOSURE (E)

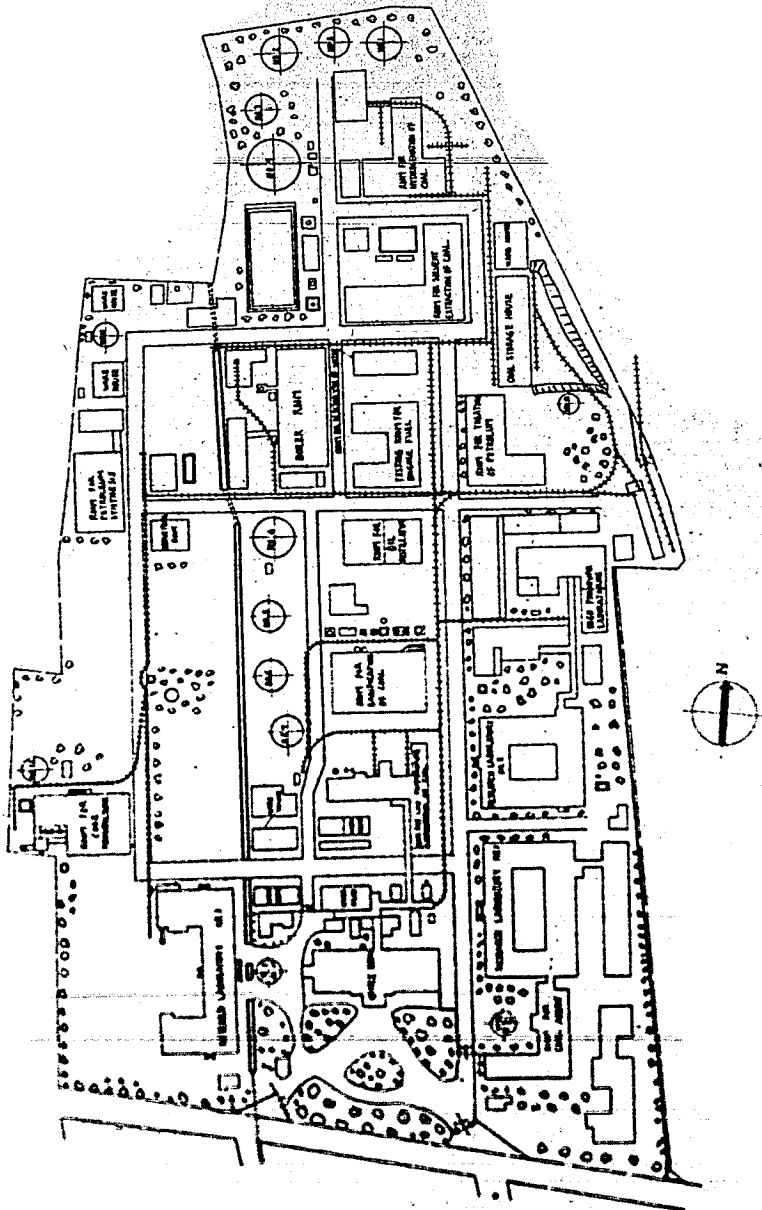


Figure 1(E)
 THE IMPERIAL FUEL RESEARCH INSTITUTE
 KAWAGUCHI, Saitama, Pref., Honshu, Japan

ENCLOSURE (B)

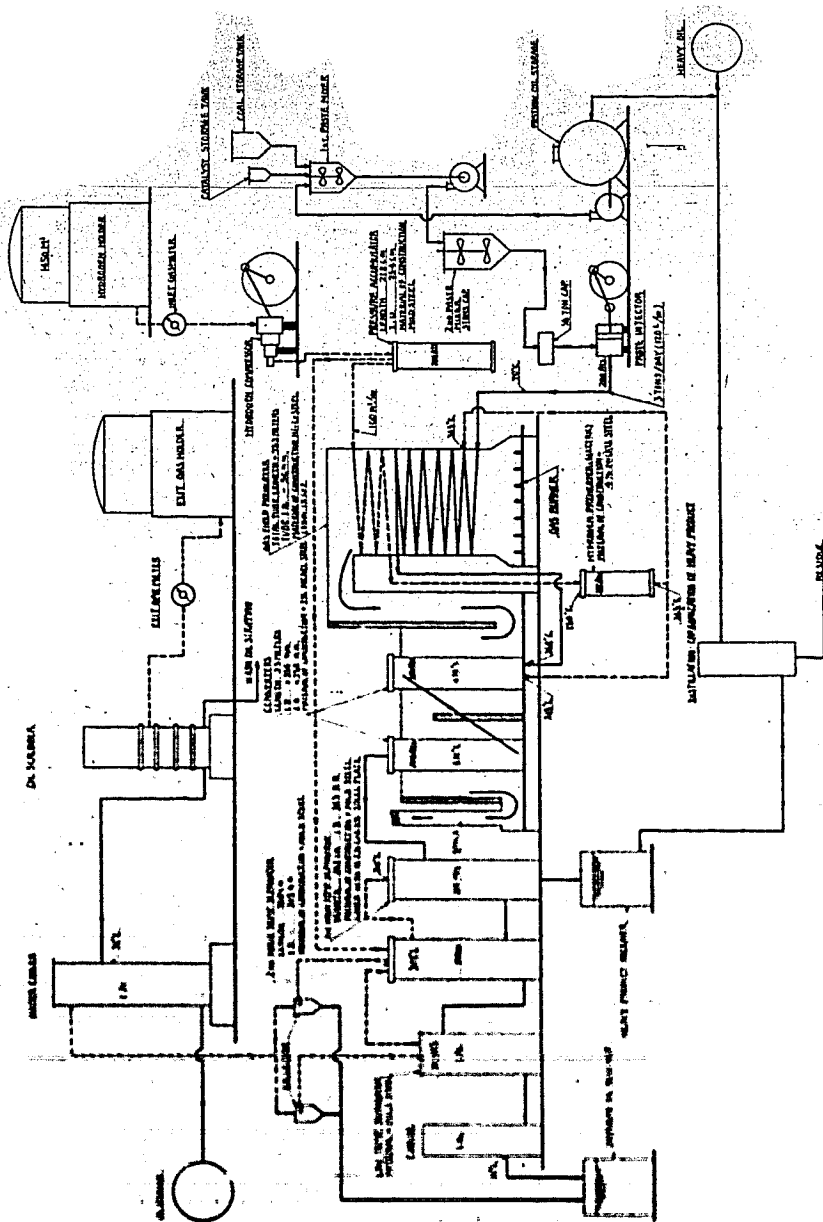


Figure 2(5)
FLOW SHEET OF IMPERIAL FUEL RESEARCH INSTITUTE
PILOT PLANT FOR HYDROGENATION OF COAL
KAWACUCHI, Settsu, Japan

ENCLOSURE (E)

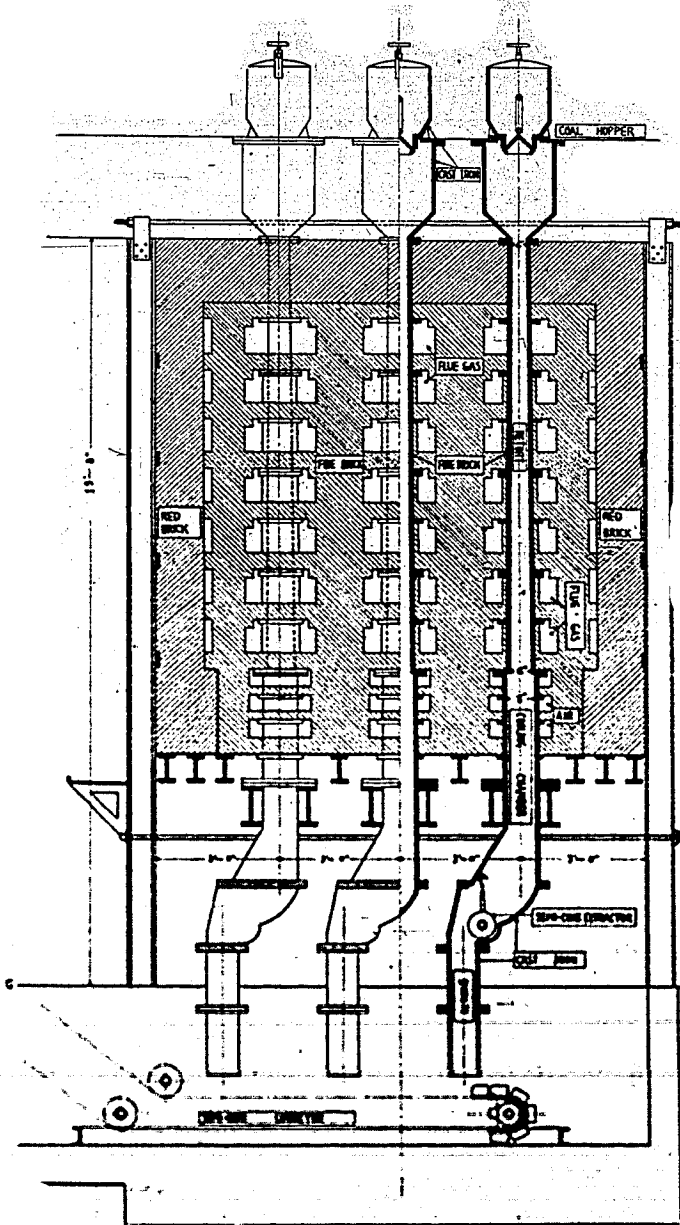


Figure 3(E)
 ELEVATION
 OF LOW
 TEMPERATURE
 CARBONIZATION
 RETORT SYSTEM
 Imperial Fuel
 Research Institute,
 KAWAGUCHI,
 Saitama Pref.,
 Japan

RETORT BENCH

12 x 13 x 19

NO. OF RETORTS

Six

OUTPUT

Six Tons Per Day

DIMENSIONS OF RETORTS

Top- 4 x 29"

Bottom- 6 x 33"

Height- 11

WORKING TEMPERATURE

500°C Inside Retort

700°C Outside Retort

FUEL

Product. Gas. or Low

Temp. Carbonization Gas

ENCLOSURE (E)

APPENDIX I
JAPANESE DOCUMENTS

Bulletins of the Imperial Fuel Research Institute
Related to Synthetic Fuel Investigation, Forwarded
Through ATIS to the Washington Document Center

<u>NavTechJap Document No.</u>	<u>ATIS No.</u>	<u>Title and Author</u>
ND26-0023.1	4594	Studies on the Liquefaction of Coal (Part V) (Action of Catalysts on the Coal Paste) by M. KUROKAWA.
ND26-0023.2	4594	The Preparation of Benzol from High Temperature Tar by Hydrogenation, by T. NIIMURA, H. NOMURA and N. TAKASE.
ND26-0023.3	4594	Studies on the Liquefaction of Coal (Part IV) (The Preparation of Hydrogen by the Water Gas Reaction) by M. KUROKAWA and K. TAKENAKA.
ND26-0023.4	4594	Studies on the Liquefaction of Coal (Part III) (Test Results by Various Catalysts) by M. KUROKAWA, K. HIROTA, K. FUJIWARA and S. ASACKA.
ND26-0023.5	4594	The Hydrogenation of Phenols (Part II) (On the Preparation of Hydrocarbons from Low-Temperature Tar Acids) by A. ANDO.
ND26-0023.6	4594	The Hydrogenation of Heavy Oils, by S. ANDO.
ND26-0023.7	4594	The Hydrogenation of Low-Temperature Tar, by S. ANDO.
ND26-0023.8	4594	Studies on the Liquefaction of Coal (Part II) (Test Results of Japanese Coals) by S. UCHIDA.
ND26-0023.9	4594	Hydrogenation of Phenols (Part I) by S. ANDO.
ND26-0023.10	4594	Studies on the Liquefaction of Coal (Part I) (Recovery of Hydrogen from Waste Gases) by K. TAKENAKA.
ND26-0024.1	4595	The Experiment on the Tully Gas Manufacture, by Y. BAN, R. KOMURA and N. SAITO.
ND26-0024.2	4595	The Synthesis of Gasoline (Part I) by S. TSUTSUMI.
ND26-0024.3	4595	The Preparation of Water Gas from Methane, by S. TSUTSUMI.
ND26-0025.1	4596	The Experiment on the Low-Temperature Carbonization of Coal, by Y. BAN, N. SAITO and S. ISHIKAWA.
ND26-0025.2	4596	The Experiment on the Continuous System of the Low-Temperature Carbonization of Coal, by Y. BAN, N. SAITO and R. KOMURA.

ENCLOSURE (F)

APPENDIX II
JAPANESE DOCUMENTS

Reports on War Activities of the Imperial Fuel
Research Institute Forwarded Through ATIS to the
Washington Document Center

<u>NavTechJap Document No.</u>	<u>ATIS No.</u>	<u>Title and Author</u>
ND26-0023.11	4594	The Catalytic Cracking of Low Temperature Tar. By K. SUGARAWA.
ND26-0023.12	4594	The Hydrogenation of Tatung (Daido) Coal. By M. KUROKAWA and S. ANDO.
ND26-0025.3	4596	The Low-Temperature Carbonization of Coaly Shale. By N. SAITO.
ND26-0025.4	4596	The Carbonization of Pine Wood. By N. SAITO.
ND26-0020	4591	The Preparation of Aviation Gasoline from Japanese Pine Root Oil. By T. AMEMIYA.
ND26-0021	4592	The Explosive Combustion of Coal Dust Fuel. By T. INATOME.
ND26-0022	4593	The Synthesis of Triptane. By M. KATSUNO.

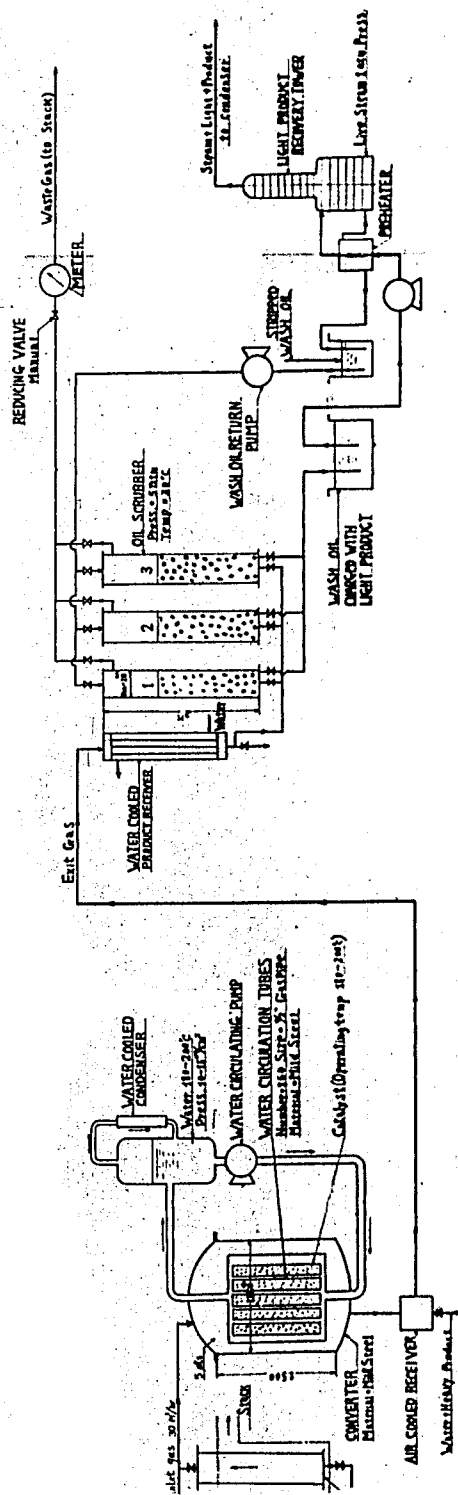
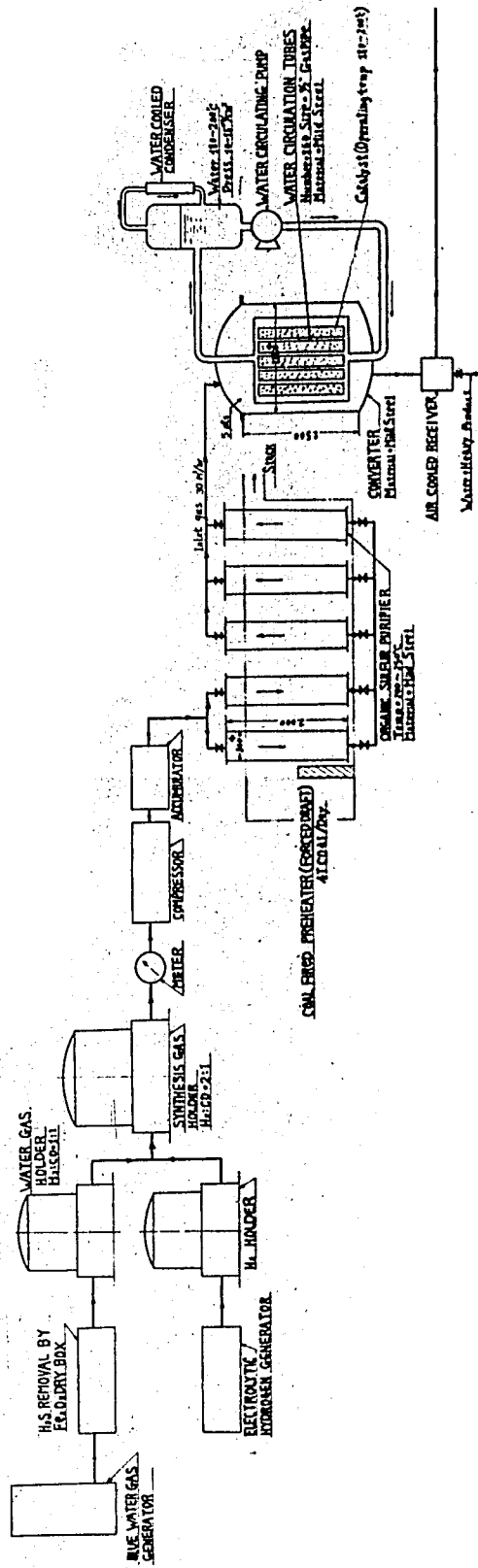


PLATE 3 (E)
FLOW DIAGRAM OF FISCHER
TROPSCH PILOT PLANT
IMPERIAL FUEL RESEARCH INSTITUTE
SAITAMA - KAWAGUCHI, JAPAN

RESTRICTED

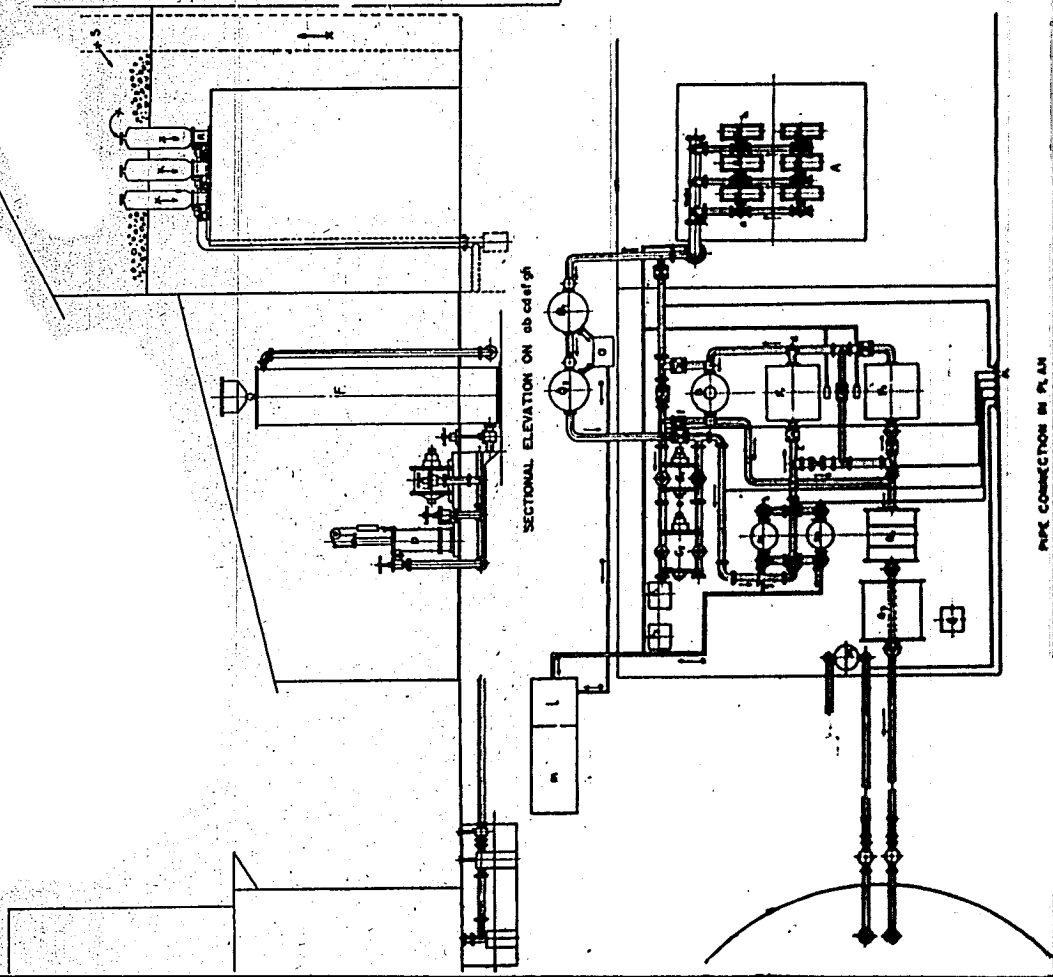


A	FURNACE
B	OIL SCRUBBER
C	GAS EXHAUSTER
D	COKE TAN EXTRACTOR
E	MOTOR
F	COKE SCRUBBER
G	COKE GAS METER
H	COKE TANK
I	GAUGE ROOM
J	COKE TANK
K	COKE TANK
L	COKE TANK
M	COKE TANK
N	COKE TANK
O	COKE TANK
P	COKE TANK
Q	COKE TANK
R	COKE TANK
S	ELEVATOR



PIPE CONNECTION AT RETDRI
SCALE 1"=1'-0"

NOTE
 --- Gas
 --- Tar
 --- Cool
 X



SECTIONAL ELEVATION ON ab cd ef gh

PIPE CONNECTION IN PLAN

PLATE II (C)
 FLOW SHEET OF LOW-TEMPERATURE
 CARBONIZATION PLANT
 IMPERIAL FUEL RESEARCH INSTITUTE
 KAWAGUCHI-SAITAMA, JAPAN