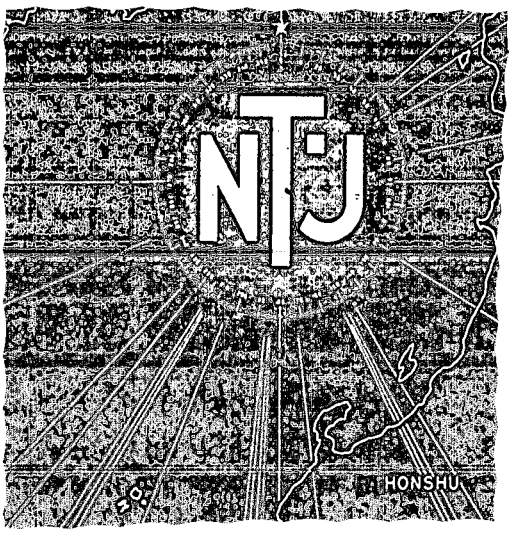
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INDEX NO. X-38(N)-4

MISCELLANEOUS TARGETS

JAPANESE FUELS AND LUBRICANTS

ARTICLE 4

PINE ROOT OIL PROGRAM

U.S. NAVAL TECHNICAL MISSION TO JAPAN

U. S. NAVAL TECHNICAL MISSION TO JAPAN
CARE OF FLEET POST OFFICE
SAN FRANCISCO, CALIFORNIA

6 February 1946

RESTRICTED

From: Chief, Naval Technical Mission to Japan.
To : Chief of Naval Operations.

Subject: Target Report - Japanese Fuels and Lubricants, Article 4 - Pine Root Oil Program.

Reference: (a)"Intelligence Targets Japan" (DNI) of 4 Sept. 1945.

1. Subject report, covering the pine root oil program in Japan outlined by Targets X-09, X-10, and X-38(N) of Fascicle X-1 of reference (a), is submitted herewith.

2. The investigation of the target and the target report were accomplished by Comdr. G. L. Neely, USNR, Lt. Comdr. C. S. Goddin, USNR, and Lieut. W. H. Millet, USNR, assisted by Ens. E. R. Dalbey, USNR, as interpreter and translator.

C. G. GRIMES
Captain, USN

JAPANESE FUELS AND LUBRICANTS
ARTICLE 4
PINE ROOT OIL PROGRAM

"INTELLIGENCE TARGETS JAPAN" (DNI) OF 4 SEPT. 1945

FASCICLE X-1, TARGETS X-09, X-10, AND X-38(N)

FEBRUARY 1946

U.S. NAVAL TECHNICAL MISSION TO JAPAN

SUMMARY

MISCELLANEOUS TARGETS

JAPANESE FUELS AND LUBRICANTS - ARTICLE 4
PINE ROOT OIL PROGRAM

The Japanese program for obtaining aviation gasoline and other liquid fuels from the dry distillation product of pine roots has been investigated. This program called for the erection of 36,000 small scale distillation units throughout Japan having an annual capacity of 2,500,000 barrels of pine root crude oil on a one to two year basis. By subsequent catalytic cracking or hydrocracking of this crude oil, it was planned to produce annually some 400,000 barrels per year of an aviation gasoline having an octane number of 90-94 with 0.15 per cent volume of lead. Full scale tests showed the performance of this fuel to be comparable to that of conventional aviation gasolines. An interesting technical development of this program was the successful catalytic conversion on a commercial scale of terpenes to aromatics. This source of fuel is not economically feasible in peace time, and it can only be considered as an emergency measure necessitated by the acute scarcity of conventional hydrocarbon-type fuels in the closing year of the war. However, the program is of significance in that it demonstrates the technical resourcefulness of the Japanese in the face of impending disaster.

TABLE OF CONTENTS

LIST OF ENCLOSURES

REFERENCES

Location of Target:

First Naval Fuel Depot, OFUNA, Kanagawa Prefecture
Third Naval Fuel Depot, TOKUYAMA
First Naval Technical Depot, YOKOSUKA
Army Fuel Research Institute, FUCHU
Imperial Fuel Research Institute, KAWAGUCHI, Saitama Prefecture
Miike Synthetic Oil Company, OMUTA, Kyushu
Toyoda Factory of Totsuka Agricultural Society, TOTSUKA, Kanagawa Prefecture
Kyushu Imperial University, FUKUOKA, Kyushu
Mitsubishi Resin-Oil Factory, SHIMABARA, Nagasaki Prefecture, Kyushu

Japanese Personnel Interviewed:

S. KOMATSU, Ph. D., Civilian Advisor to Department of Fuel Research, First Naval Fuel Depot,
(formerly Professor of Biochemistry at Kyoto Imperial University, foremost and very capable chemist of First Naval Fuel Depot).
H. FUJIMOTO, Ph. D., Engineering Commander, IJN, Head of Cracking, Dry Distillation, and Pine Root Oil Sections First Naval Fuel Depot, (research engineer of high capability).
S. SANKA, Engineering Lieutenant Commander, IJN, Head of Process Engineering Section of the First Naval Fuel Depot, (design engineer of high capability).

Research Assistants of the First Naval Fuel Depot, whose names are included in individual Japanese reports.

T. KONDO, Captain, IJN, Head of Aviation Gasoline Testing, First Naval Technical Depot, YOKOSUKA.
I. WATANABE, Engineering Rear Admiral, Japanese Navy, Superintendent of Third Naval Fuel Depot, TOKUYAMA.
N. TAKEI, Chief Chemical Engineer of Miike Synthetic Oil Company, OMUTA, Kyushu.
ISHIGARA, Business Manager of Mitsubishi Resin-Oil Factory, SHIMABARA, Kyushu.
KIROSHIMA, Ph. D., Professor of Fuel Chemistry, Kyushu Imperial University, FUKUOKA, Kyushu.

Referenced Japanese Reports

Technical Notes on Research Work, Army Fuel Research Institute, FUCHU.

INTRODUCTION

The manufacture of aviation gasoline from the product of the dry distillation of pine roots constitutes the principal and one of the most interesting emergency fuel measures adopted by the Japanese during the final period of the war. This report summarizes a mass of detailed technical information which has been obtained by the Petroleum Section of the U. S. Naval Technical Mission of Japan relative to pertinent research activity and the application of this research to the production of pine root gasoline. The history of the program is discussed, and the various methods of refining the oil are described. The properties of the fuel produced from pine root oil as well as the engine performance characteristics of this fuel are reported. Japanese research pertaining to the utilization of vegetable products of similar nature has also been investigated.

Although both the Japanese Army and Navy were engaged independently in research program on pine root oil, the emphasis of this report is on the Navy's activities since the Naval research was more complete and further advanced than that of the Army. Detailed research reports of the pine root oil research projects investigated at the First Naval Fuel Depot have been prepared in English by the Japanese technical personnel of the Depot and are included in this report as Enclosures (B)1 to (B)11, inclusive. Summaries of this research and the chemistry of pine root oil have been prepared in English by Chemical Engineering Commander H. FUJIMOTO, IJN, and Dr. S. KOMATSU, respectively. These summaries are submitted herewith as Enclosure (A). Supplementary information has been obtained by visits to various research laboratories, refineries, and rural distilling installations throughout Japan.

Since all of the research files of the First Naval Fuel Depot were burned in August 1945 by order of the Director of the Depot, it was necessary to recall the Japanese personnel to reconstruct this information from laboratory notebooks, laboratory apparatus, and pilot plant equipment. The preparation of these reports and drawings continued for a period of nearly three months, during which time each Japanese author was frequently interrogated and was assisted by the Petroleum Section of the U. S. Naval Technical Mission to Japan in the organization and translation of his reports. The material which is submitted as Enclosures (A) and (B) constitutes an integral part of this report, and although it may include minor errors in construction introduced in the translation by the Japanese authors, it does serve as an accurate indication of the quality and extent of Japanese research relative to the production of aviation gasoline from pine root oil.

THE REPORT

Part I
HISTORY OF PINE ROOT OIL PROGRAM

Prior to 1944 pine root oils were utilized for a variety of applications including their use as solvents, flotation oils, and as raw material for the synthesis of drugs and special chemicals. Up to this time they had never had seriously considered as a source of fuel. The annual production of pine root oil in Japan was essentially the same in 1944 as in 1941, - approximately 6,000 kiloliters. However, by the end of 1944, the Japanese Army and Navy as well as the Japanese civil government were devoting frantic efforts towards the proposed utilization of this material for the synthesis of high-grade aviation gasoline. More than one hundred million yen were allocated for this purpose by the Japanese government.

Early in 1944 Army laboratories approached the problem by directing their research towards the utilization of existing equipment for the refining of the crude pine root oil. However, as the refineries were increasingly damaged by bombings, it became necessary to concentrate on the fabrication of small scale equipment which could be easily constructed.

By August 1944, the Navy's research program was in full swing. Laboratory studies on the characteristics of pine root distillates were being made, and designs of simplified distillation retorts were being selected. In December 1944 a plan was proposed in which prefectural governors were to take charge of pine root oil production in their particular prefecture. Pine root stumps were to be excavated by civilian workers or home service troops and then crude distilled by the farmers. The Army and Navy were each assigned districts in which they were to provide the equipment and be largely responsible for treating the crude oil.

Several treating methods were studied and those finally selected were chosen more for ease of construction and simplicity of design than for the quality of product obtained. It was planned to erect 36,000 retorts throughout Japan. The retorts would be of standard design and would produce 360,000 kiloliters per year of pine root oil. By June and July of 1945 this production was nearly realized.

In connection with this plan the Navy published a pamphlet entitled "Pine Root Oils" which presented a complete picture of the industry and the processes which were to be applied. A Japanese translation of this interesting pamphlet was prepared for inclusion in this report and is submitted as Enclosure (C).

During 1945 pilot plant tests on hydrocracking of pine root oils were being conducted, and the construction of twenty-one small scale catalytic cracking plants was planned. At the end of the war, construction had been started on a group of simplified catalytic reforming plants, although these units never were completed.

Part II
REFINING STUDIES

The refining of pine root gasoline from pine roots takes place in several successive or alternate steps. Each of the steps is described in detail in Enclosures (A), (B), and (C). There follows a brief discussion of each of these refining procedures.

A. DRY DISTILLATION AND FRACTIONATION

A Kitagawa vertical-type retort was selected as standard for the dry distillation. The yield of pine root oil varied from 10% to 25% of the weight of pine roots charged depending on the species of pine and the age of the roots. The retort selected could treat 350 kilograms of pine roots daily and the annual production of pine root crude would amount to approximately 10 kiloliters. Details of design are shown in Enclosure (B)1, and a complete description of the raw materials, apparatus, and procedures for dry distillation is presented in Enclosure (C).

Small scale batch-type stills were designed for the fractionation of the total pine root oil (Enclosure (B)4). The product was separated as follows:

1. Fraction boiling below 185°C. (Turpentine No.1)
2. Fraction boiling from 185°C-300°C. (Turpentine No.2)
3. Residual Oil. (Subjected to further dry distillation to yield bunker fuel.)

The chemical constituents of each of these fractions are discussed in Part II of Enclosure (A) and in Enclosure (C).

A typical distillation unit, slightly different in design than that proposed by the Navy, was visited by the U. S. Naval Technical Mission to Japan in October 1945. A report on this plant, located in SHIMABARA, Kyushu, is submitted herewith as Enclosure (D). A Navy-type distillation unit in TOTSUKA was also visited. Pictures of this unit are included as Figure 6 and 7 of Enclosure (B)1.

B. CONTACT DISTILLATION

One of the procedures studied by the Army Fuel Research Institute and other laboratories under the jurisdiction of the Army Fuel Bureau, such as that of the Miike Synthetic Oil Company, OMUTA, was the so-called contact distillation method, in which a slurry of pine root crude oil and Japanese acid clay was subjected to distillation followed by subsequent rectification of the distillate. The fuel obtained was high in unsaturates and of inferior grade. No extensive application of this method was applied.

C. CATALYTIC REFORMING

The simple catalytic reforming of the pine root oil fraction boiling below 185°C was the treatment which was studied in detail at the First Naval Fuel Depot. Research on this procedure and design data for the catalytic reforming units are outlined in Enclosures (B)3 and (B)4. The reforming was effected by means of Japanese acid clay at a temperature of 350°C and atmospheric pressure. The product was too low in volatility to be of direct use as a fuel but most of the unsaturates were transformed by this treatment. The gasoline was obtained in a yield of 12% by volume of the original total pine root oil, and it was planned to blend this product with alcohol for use as an aviation fuel.

The conversion of cyclic unsaturates of the mono-terpene type to aromatics and naphthenes was effected by this treatment. A discussion of this molecular rearrangement is presented by Dr. KOMATSU in Part II of Enclosure (A). It is Dr. KOMATSU'S personal opinion that the rearrangement in the presence of acid clay is catalyzed by the presence of traces of iron abietate, although no experimental data were presented to substantiate this hypothesis.

D. CATALYTIC CRACKING

Catalytic cracking of the two pine root oil fractions was investigated at the First Naval Fuel Depot. Both a U. O. P. catalyst, prepared by the Nippon Oil

Company, and Japanese acid clay catalysts were studied. It was decided to use this process with a clay catalyst for the pine oil fraction boiling from 185°C to 300°C. An aromatic base gasoline, having an octane number of 90-92 with 0.15% added lead, was obtained in 6% yield by volume of the original total pine root oil.

Pilot plant tests on this process are discussed in Enclosure (B)5 and design details of a simplified catalytic cracking unit are shown in Enclosure (B)6. It was planned to construct twenty-one of these catalytic cracking plants, each treating from 10-30 kiloliters per day, at centrally located refineries. Two large catalytic cracking plants at the Second Naval Fuel Depot, YOKKAICHI, having a 4,000 barrel per day capacity, were also to be utilized.

E. HYDROCRACKING

Enclosure (B)7 describes autoclave and pilot plant tests relative to the hydrocracking of the pine root oil fraction boiling from 185°C to 300°C. The product was a naphthenic fuel having an octane number of 90 to 94 with 0.15% added lead and was obtained in yields of 50% of the charged pine root oil fraction. The gasoline was superior in quality and yield to that obtained in catalytic cracking, but the latter process was adopted for future construction because of simplicity of design and ease of operation.

The hydrocracking plant at the Third Naval Fuel Depot, TOKUYAMA, was utilized in applying the results of the pilot plant tests. This unit treated 1.8 kiloliters per hour of charged oil and a yield of 71% of aviation gasoline was obtained. The conditions were as follows:

Hydrogen pressure	200 kg/cm^2
Temperature	400-450°C
Space Velocity	0.5-1.0
Catalyst	$NiO-MoO_3$

A total of 600 kiloliters of pine root gasoline were prepared at TOKUYAMA by this process.

Part III
PROPERTIES OF PINE ROOT OIL PRODUCTS

The physical and chemical properties of the various pine root oils and gasolines are tabulated in Table I. Discussion of these products and more detailed information in regard to each will be found in Enclosures (A), (B), and (C).

Part IV
ENGINE TESTING OF PINE ROOT GASOLINES

Single cylinder engine tests on the untreated pine root oil fraction boiling below 185°C were conducted at the First Naval Depot and are reported in Enclosure (B)10. As would be expected the high gum content and low volatility rendered use of the fuel impracticable. When blended with ethyl alcohol, the fuel was also unsatisfactory. A blend of 80% (by volume) or alcohol with 20% of pine root oil gave test results comparable with those obtained using ethyl alcohol but the gum content of the fuel was exceptionally high.

The First Naval Technical Depot, YOKOSUKA, conducted full scale engine tests and flight tests on pine root oil gasolines which had been prepared at the First Naval Fuel Depot by catalytic cracking and by hydrocracking. Results were equivalent to those obtained with No. 1 grade aviation gasoline of 92 octane number. More detailed information on the test procedure and test results are given in Enclosure (3)11.

The blending of gasoline obtained by the catalytic reforming process with ethyl alcohol for use as a fuel for training planes was planned. The blending ratio was to be 60% (by volume) of pine root gasoline to 40% (by volume) of alcohol. No engine test data of this fuel were available.

Part V
RESEARCH ON RELATED PRODUCTS

Both the Army and Navy laboratories investigated the oils obtained from materials similar to pine roots with the purpose of including these materials in the same fuel program. The carbonization and the high pressure hydrocracking of pine resin were studied at the First Naval Fuel Depot (Enclosure (B)2 and (B)8). The Army Fuel Research Institute intended to utilize the dry distillation product of resin in the preparation of a fuel which was to be blended with alcohol. The resin was to be mixed with acid clay and dry distilled. The distillate was to be fractionated and the fraction boiling below 200°C was to be blended with alcohol. The properties of the pine resin gasoline before blending were as follows:

Specific Gravity, 15/4°C	0.82-0.83
Acid Value	0.2-0.4
Reid Vapor Pressure, kg/cm^2,40°C	0.1
Gum Content, mg/100cc	3-4
Freezing Point, °C	Below-50
Engler Distillation	
First Drop, °C	60
10%, °C	130
50%, °C	170
90%, °C	195
Octane Number, unleaded	
C. F. R., Motor Method	75

Another project which was contemplated by the Army was the steam distillation of pine needles and the subsequent catalytic reforming of the product. The dry distillation of Shirakamba bark was also investigated. This material yielded 50% distillates in a process similar to that used for pine root oil. The yield of product was higher and the volatility characteristics were superior to those of pine root oil. The extraction of limonene from orange peel was also studied as a possible fuel source.

A summary of preliminary studies relating to the utilization of such materials as the trunks, branches and needles of acerose trees and the trunks and bark of broad leaved trees has been compiled by the First Naval Fuel Depot and is included as Enclosure (C).

Part VI
ECONOMIC ASPECTS OF THE PINE ROOT OIL PROGRAM

As can be readily concluded by a consideration of the various phases of the pine root oil program, it does not constitute a sound economic measure for obtaining aviation fuel. The plan called for the use of approximately 3,780,000 metric tons of pine roots per year and could not be maintained for more than two years. The yield of total pine root oil and tar was planned as 360,000 metric tons per year. This corresponds to slightly more than 400,000 barrels per year of aviation gasoline to be obtained by utilizing both the catalytic reforming and catalytic cracking processes.

The cost of obtaining the crude pine root oil is exceptionally high, although refining costs are nominal. It was estimated that the cost of 1 "to" (18 liters) of total pine root oil was 25.9 yen in November 1944. This represents a cost of 226 yen per barrel of crude oil. Most of this cost may be attributed to the expense of digging, transporting, and hand chopping the pine roots.

The Japanese technical personnel, who have studied and originated some of the procedures discussed in this report, are of the opinion that pine root oil can be utilized in Japan's post war economy as a source of such materials as menthol and camphor to be used for export. For this purpose, the Department of Commerce and Industry of Japan desires to stabilize the annual production of pine root oil at 10,000 kiloliters so that the pine tree resources may be preserved.

Table I
PROPERTIES OF PINE ROOT OILS

Description	Total Pine Light Fraction	Root Oil Heavy Fraction	Pine Root Oil Gasoline			
	-185°C	185-300°C	Contact Distillation	Catalytic Reforming	Catalytic Cracking	Hydrocracking
Specific Gravity	0.86	0.95	0.85	0.85	0.762	0.727
ASTM Distillation						
I.B.P. °C	110	165	70	70	44	36
10% °C	140	184		135	77	64
50% °C	170	234	160	165	105	101
97% °C	190	331(90%)	206	200	128.5	144
E. P.	250	332	210	280	172	175
Composition,%						
Unsaturates	89.2				11	0.5
Aromatics	6.8			60	32	12.9
Naphthenes	4.0			40	20.9	21.9
Paraffines					36.1	64.7
Octane Number						
Plain	70		80	85	75	71
Leaded(0.15%)	75		84	90	90	93