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RECLAMATION OF CONTAMINATED KINNEY AND DIFFUSION PUMP OILS

AND

REMOVAL OF URANIUM THEREFROM

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RECLAMATION OF CONTAMINATED KIMNEY AND DIFFUSION PUMP OILSANDREMOVAL OF URANIUM THEREFROMABSTRACT

Test distillations were made with Kimney and diffusion pump oils under vacuum in order to determine accurately the temperature range over which satisfactory distillate may be collected. Also recommendations are made as to the most advisable type of heating equipment necessary to produce the required heat and maintain the desired temperature. In addition, several important refinements in the distillation equipment were checked for operational characteristics and for their efficacy in aiding the production of uranium-free distillates.

RECLAMATION OF CONTAMINATED KINNEY AND DIFFUSION PUMP OILS

AND

REMOVAL OF URANIUM THEREFROM

INTRODUCTION

Additional experimentation was initiated on the recovery from and reclamation of vacuum pump oils, supplementing the information already given (Reports C-3.320.2 and C-3.320.3). Particular attention was given to specific temperatures of vaporization of the oils and the temperatures that would have to be maintained in the fractionating column of a distillation system. Certain modifications and improvements over previous experimental equipment were incorporated and their performance checked during the distillation work reported herein.

The oils under test were Kinney and diffusion pump oils, taken from the oil systems in the beta buildings and constituted representative samples of used oil with respect to water content and uranium contamination.

Specific modification of the equipment included a new reflux column of a baffle type and a heated preoutgassing unit with a chamber injection inlet for charging the used oil into the equipment. These items are described in detail in the subsequent text.

EXPERIMENTAL

Moisture Content of Used Oils

During these distillations an attempt was made to overcome the difficulties which are encountered due to excessive amounts of moisture present in these oils as well as to facilitate the handling of used oils of a more routine nature with respect to water content. Removal of moisture from the oil, made excessively long outgassing periods necessary before distillation could begin at the high vacuum necessary for producing the distillate of the required quality insofar as low vapor pressure was concerned. Without special equipment, an average water content of 75 to 300 ppm is sufficient to disrupt flash distillations at greatly reduced pressure. Distillation is even more difficult, of course, in instances where emulsions or free water are present as has not infrequently been the case.

Outgassing Oil Charges

In the course of earlier experiments, it had been the practice to charge the used oil in small amounts, two to three liters, into a feeding flask of five liters capacity at atmospheric pressure. After this amount had been admitted, the valve to the vacuum pump (roughing) was opened slowly in order to begin the evacuation and outgassing without the violent foaming that would be caused by starting this operation too rapidly. As a result, considerable time was consumed, usually in excess of one hour before the oil could be outgassed to a point where it would be at all practical to drain it into the distillation flask.

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No heat was applied during this period and consequently the distillation was stopped whenever new charge was being admitted, unless the heat supplied to the still was substantially increased or the rate of admission reduced appreciably. An additional drawback to this preparation procedure was the inability to achieve a completely satisfactory degree of outgassing without the application of heat. This resulted in some loss of pressure in the still when new charge was being admitted, thus making it necessary to further throttle the rate of input to the still.

A unit was designed which, it was believed, would facilitate the urgently needed faster and more thorough preliminary handling of the oil to be distilled. The principal features of the unit were an overhead injection chamber and a heating unit. The oil is admitted through a small orifice into a chamber already completely evacuated. (See Plate I) The oil foams into this chamber where, almost instantly, the oil is largely outgassed after which it drains down into a receiving reservoir immediately below. The receiver is externally heated (Glascol mantle) to raise the temperature of the oil to a point approaching the vaporization temperature of the lighter oil fractions. Since such a temperature is well above that of the boiling point of water, especially at these reduced pressures, the vapor contribution of that origin is thoroughly and rapidly removed. Once the oil is outgassed, the oil is displaced with helium or nitrogen and allowed to flow into the distillation flask. Since the temperature of the oil at the time of admission is so near that of vaporization and because the oil is already thoroughly outgassed, distillation begins at once with only negligible heat and pressure losses.

PLATE I DISTILLATION EQUIPMENT

A OIL INLET	I DISTILLATION FLASK
B EXPANSION CHAMBER	J FRACTIONATING COLUMN
C TO ROUGHING PUMP	K CONDENSER
D HELIUM INLET	L TO ROUGHING & FINISHING PUMPS
E STANDARD PYREX PIPE FLANGES	M MCLEOD GAGE CONNECTION
F OUTGASSING & PREHEATING FLASK	N RECEIVING FLASK
G CHARGE INLET	O DISTILLATE DISCHARGE
H RESIDUE DISCHARGE	

Reflux Column Revisions

The experimental distillations of Kinney and diffusion pump oils, previous to this time, had been accomplished in stills equipped with long reflux columns of the unpacked type. Although the distillates produced with this type of column were relatively free of uranium, indicative of satisfactory fractionation, the column was somewhat sensitive to temperature control and was restricted to a limited capacity of oil vapor flow. In order to reduce or eliminate these undesirable characteristics and to effect a possible reduction in overall length of this item, a baffle type column was designed.

Reviewing considerable experience with the operation of long, unpacked columns, it was believed that a baffle section could be incorporated successfully into the design. An arrangement was desired that would effectively prevent entrainment due to vertical splashing of the unvaporized or condensed material, and yet not be of such complex nature as to cause actual flooding of the column. Flooding invariably occurs if the column is packed with rings or some similar material. In addition, because of the choking effect of a closely packed column, the pressure differential between the distillation flask and the vacuum pumps, would be of intolerable magnitude for the preparation of quality diffusion pump oil.

With the above facts in mind, a column was designed with the following general features. A 12-inch section of a 4-foot length of glass pipe, 1-inch in diameter, was enlarged to a diameter of two inches.

The wall of this enlarged section, which section being eight inches from the lower extremity of the column, were indented by collapsing the glass wall to the center of the tube. (See Plate I) Another indentation was made at 90 degrees rotation and at a point slightly higher than the first indentation. When four such indentations had been made in the enlarged section, the column was completely obstructed to undeflected axial flow. The actual throat diameter had not been constricted at any point to a degree where the cross sectional area was less than that of the 1-inch-diameter portions of the column. A column, fabricated as described, was equipped with external heating elements, insulated and installed.

The column was used for distillations of both Kinney and diffusion pump oil. The performance was excellent, since not only did the baffle section prevent vertical splashing within the column but it also prevented flooding of the narrow section of the column when the rate of vapor flow exceeded what would have otherwise been greater than a critical magnitude. The excess condensate was held within the enlarged baffle section until it drained or was revaporized. The distillates were analyzed for uranium and were found to be acceptably free. The uranium aspect of these distillations is discussed in more detail in a subsequent paragraph.

The type of baffle section as described, is believed to be sufficiently effective to make possible a substantial reduction in the length of the entire column. Such a reduction would be extremely desirable since it is expedient that the oil be kept in the vapor state

for only as little time as possible if the original characteristics of the oil are to be preserved without cracking. A photograph of the equipment used for this work is shown in Plate II on the following page.

Instrumentation for Temperature Determination

Thermocouples were located at six points throughout the distillation equipment in order to facilitate the determination of temperatures at all critical points of interest, pertinent to the distillation of these oils. The still was an assembly of 1-inch pyrex pipe, similar to previous models used for oil distillation, as illustrated in Plate I and II. The outgassing and preheating flask, distillation and receiving flask are each of five liters capacity. The first two flasks are heated by spherical mantles (Glascol). The location of thermocouples of the equipment, as described above, is indicated in the following paragraph.

Thermocouple no. 1 was replaced between the mantle and wall of the distillation flask at the underside. Thermocouple no. 2 was placed in a thermowell extending down into the liquid phase within the distillation flask (residual charge temperature). Thermocouple no. 3 was placed in a thermowell extending only into the upper or vapor phase portion of the distillation flask. Thermocouple no. 4 was located within the upper portion of the reflux column. Thermocouple no. 5 was duplicated on points five and six on the Micromax, recording the temperature in the distillation elbow at the distillation condensation point (vapor condensation temperature).

The temperatures at the various thermocouples were recorded continuously by a 6-point Leeds and Northrup Micromax.

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Plate II

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Diffusion Pump Oil Distillation Temperatures

The distillation was begun after installation of the equipment was complete and the system pumped down to 0.5 microns pressure. The first distillations were made with diffusion pump oil and four separate runs were completed. For the purpose of obtaining further information with regard to temperature and pressure, the conditions of each run were altered over a varied range of probable operating conditions as regards the heat required to distill the oils in question. The amount of charge per run was three liters of used oil. All oil charges had been subjected to outgassing in the new injection unit and were preheated before admission to the distillation flask.

Run no. 1 was distilled at 0.7 to 6.0 microns pressure and at the maximum rate permissible with the distillation flask and reflux column being used. Distillate was received at temperatures ranging from 100° to 285° C. The temperature developed in the residual charge (boiler) during the period that distillate was being collected was 195° to 315° C. None of these temperatures are corrected for thermocouple lag, which would tend to make the initial figure of the temperature range for the vapor condensation temperature slightly lower than is actually the case. Experience and observation have shown that thermocouple lag is negligible in the case of all other temperatures. The distillation was terminated after effecting approximately 90 percent distillation in 35 minutes. (See Figure 1.)

Run no. 2, a charge of 3 liters in addition to the residues remaining after run no. 1, was distilled at 0.6 to 1.2 microns pressure

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and at a minimum rate in an effort to determine accurately the lowest possible distillation temperatures that would be practical for routine work. Distillate was received at temperatures ranging from 60° to 250° C. The temperature developed in the residual charge during the period that distillate was being received was 201° to 282° C. A practical distillation rate was not achieved until the boiler temperature had reached 250° C or better. The distillation was terminated after collecting distillate for a period of four hours and six minutes. (See Figure 2.)

Runs no. 3 and 4 were two consecutive distillations, the latter being charged into the still with as little loss of time as possible after run no. 3 had been terminated. Both the distillations were conducted at an average pressure of 25 microns and at an average rate as compared with runs no. 1 and 2. Distillate was received during run no. 3 at temperatures ranging from 122° to 238° C. The temperature developed in the residual charge during the period that distillate was being received was 202° to 261° C. After distilling for one hour and thirty minutes the distillation flask was recharged with pretreated oil and the distillation resumed at a slightly increased rate. Distillate was received at temperatures ranging from 192° to 254° C. Note the higher initial temperature due to partially overcome thermocouple lag effected by the rapidly resumed distillation from preheated oil. The temperature developed in the residual charge during the period that distillate was being received was 207° to 274° C. The total time consumed from the moment that the first drop of distillate was received during run no. 3 until run no. 4 was terminated was 3 hours and 18 minutes. (See Figure 3.)

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Graphs have been prepared from the data recorded on the Micromax charts and appear in figures 1 to 4. Since the temperatures recorded by points 2, 5, and 6 proved to be the ones providing the most important information, only those points appear in the temperature graphs. It will suffice to say that the temperature at point no. 3 was consistently a few degrees lower than the temperature at point no. 2, during the period of actual distillation. The thermocouple at point no. 4 absorbed some heat from the column heating elements but indicated an average temperature midway between point no. 2 and 6. The temperature at point no. 1 usually exceeded the residual charge temperature (point no. 2) by at least 100 degrees, depending upon how rapidly the temperature increases were being accelerated.

For the purpose of comparing temperature differences for the low (0.7 - 6.0 microns) and high (25 microns) pressure distillations carried out, the temperature data reported above and shown in the pertinent figures, is presented in table form, plus the average temperature calculations, in Table I.

Table IDIFFUSION PUMP OIL - TEMPERATURE MEASUREMENTS

Run No.	Vapor Phase		Liquid Phase		Pressure (microns)
	Initial	Final	Initial	Final	
<u>Recorded Temperatures in Degrees Centigrade</u>					
1	100	285	195	315	0.7 - 6.0
2	60	250	201	282	0.6 - 1.2
3	122	232	202	261	25
4	192	254	207	274	25
<u>Average Temperatures in Degrees Centigrade</u>					
1-2	80	268	198	299	10
3-4	162	246	205	268	high

The table indicates that the average initial temperature of both liquid and vapor phases were lower at the lower pressures. These figures are not an accurate indication of the relation between temperature and pressure because of insufficient data and because the experiments that are reported were not distilled at uniform rates. A sufficiently accurate determination of the basic temperatures involved, however, is attained.

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Kinney Pump Oil Distillation Temperatures

Following the completion of four distillations of diffusion pump oil, three additional distillations (run no. 5, 6, and 7) were made with Kinney pump oil. The same equipment was used, including the preheating-outgassing unit, and the volume of each charge was again three liters.

Run no. 5 was distilled at 0.8 microns pressure and at the minimum possible rate in an effort to determine the minimum permissible distillation temperature. Distillate was received at temperatures ranging from 95° to 238° C. The temperature developed in the residual charge during the time that distillate was being received was 185° to 274° C. The run was terminated after collecting distillate for 2 hours and 36 minutes.

Run no. 6 was admitted to the still from the charger immediately upon termination of the preceding run. This distillation was done at an increased rate and at 25 to 30 microns pressure. Distillate was received at temperatures ranging from 153° to 280° C. The temperature developed in the residual charge during the period that distillate was being received was 165° to 330° C.

Run no. 7 was also admitted to the still immediately upon termination of the previous run. This run was distilled at maximum rate and at an average pressure of 35 microns. Distillate was received at temperatures ranging from 220° to 285° C. The temperature developed in the residual charge during the time that distillate was being received was 225° to 365° C. An apparently satisfactory distillation rate was attained during this run which was completed in 1 hour and 30 minutes. Temperature data for run no. 5, 6, and 7, appears in Table II.

Table IIKINNEY PUMP OIL - TEMPERATURE MEASUREMENTS

Run No.	Recorded Temperatures in Degrees Centigrade				Pressure (microns)
	Vapor Phase		Liquid Phase		
	<u>Initial</u>	<u>Final</u>	<u>Initial</u>	<u>Final</u>	
5	95	238	185	274	0.8
6	153	280	165	330	25 - 30
7	220	285	225	366	35

A more comprehensive picture of the temperature variation during the distillations may be gained by an inspection of the graph (in Figure 4) which has been prepared from the micromax chart recordings.

Uranium Distribution

The original used Kinney and diffusion pump oils, as well as the distillates produced during the experimentation, were analyzed for the uranium they contained. The analytical results, as tabulated below, show that uranium contamination is at least reduced by the distillation operation. It will be noted, however, that the concentration of uranium in the oil is initially of such low order of magnitude that positive analyses in this range are difficult to obtain. The trend shown by the analyses is, nevertheless, in the desired direction. It is reasonable, therefore, to assume that the efficacy of uranium isolation in the distillation residua has not been impaired by the design revisions undergoing performance tests during these experiments.

Table IIIURANIUM CONTAMINATION

(mg. Uranium/liter oil)

<u>Sample No.</u>	<u>Diffusion Pump Oil</u>		<u>Kinney Pump Oil</u>	
	<u>Crude</u>	<u>Distillate</u>	<u>Crude</u>	<u>Distillate</u>
A	0.55	1.9	4.0	1.6
B	0.69	1.9	1.8	0.7
C	0.5	2.9	1.9	1.6
D	—	—	1.2	—
Average	0.6	0.2	2.2	1.5

SUMMARY

The temperature data acquired from the distillations discussed herein, provides comprehensive information in regard to the heat requirements necessary for vacuum distillation of Kimmey and diffusion pump oils. The tables and graphs included show the range of temperatures developed in the residual charge during the progress of distillation under a variety of probable operating conditions in the equipment described. It becomes apparent that equipment designed to accommodate distillations of the nature described will not be adequately served by heat from high pressure steam (175 p.s.i.) which was one of the principle points of conjecture at the time the experimentation was undertaken. Although the heat for these experiments was supplied by Glascol mantles, electrical heat supplied from heavier duty elements would seem to be the most practical for large scale operations. One precaution must be taken, however, that the heaters are not of the bayonet or some immersion type, since exposure of the heating elements when the liquid level in the boiler is extremely low would cause severe overheating in the nonconductive atmosphere of an evacuated chamber. External heating elements, preferable attached to the underside of a unit, would be satisfactory.

The two major changes in the distillation equipment used, namely the baffle-type reflux column and the injection-expansion feature of the feeding unit have proven themselves to be of invaluable aid in these reclamation distillations. It becomes possible to prepare for vacuum distillation oils of high water and dissolved gas content within a gratifyingly short period of preliminary process and pumpdown time. The

distillation proper benefits from the improved column design by increased rate of production and improved quality of distillate.

The oil distillates are essentially freed of any uranium contamination they originally contained, since such uranium remains in the still-bottoms and is recovered therefrom by subsequent solvent extraction.

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