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MONITORING FOR URANIUM ACCUMULATIONS IN DIFFUSION PLANT EQUIPMENT

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OAK RIDGE GASEOUS DIFFUSION PLANT

OAK RIDGE, TENNESSEE

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MONITORING FOR URANIUM ACCUMULATIONS IN DIFFUSION PLANT EQUIPMENT*

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INTRODUCTION

The enrichment of uranium in a gaseous diffusion cascade is a continuous, repetitive process which requires that multiton quantities of uranium hexafluoride (UF_6) be pumped through process equipment of varying physical size. Figure 1 is a photograph of the largest equipment employed.[1] One of the potential operating problems associated with the process is the inadvertent accumulation in equipment of solid uranium compounds. The events leading to such accumulations are summarized below in the presentation of the techniques employed at the Oak Ridge Gaseous Diffusion Plant for locating uranium depositions using gamma radiation detection equipment and estimating their mass by neutron counting.

CHARACTERISTICS OF UF_6

Brief mention of some of the characteristics of the uranium hexafluoride process gas will be helpful in the following discussion. It is the only known uranium compound suitable for the gaseous diffusion process. Figure 2 illustrates the pressure-temperature relationship for UF_6 and shows that it is a solid at room temperature and atmospheric pressure. Consequently, a diffusion cascade must be operated at temperatures and pressures necessary to maintain gas phase UF_6. Chemically, the compound is stable; however, it is a vigorous fluorinating agent and is very reactive with water to form uranyl fluoride (UO_2F_2) and hydrogen fluoride (HF).

MECHANISMS FOR ACCUMULATION

The reactivity of UF_6 with water provides the most common mechanism for accumulation of nonvolatile uranium compounds in plant equipment. An isotope separation cascade operates at subatmospheric pressure and inleakage of humid ambient air through, for example, a small crack in a valve or expansion joint bellows, will result in the formation of uranyl fluoride. The result of a leaking valve bellows and seat is shown in figure 3. Typically, the uranyl fluoride remains as a solid deposit in the area of inleakage while the hydrogen fluoride reaction product is swept away as a gas phase cascade contaminant.

The second most common cause of deposition is UF_6 condensation resulting from high pressures or low temperatures due to operational changes or heat supply failures. Figure 4 illustrates the appearance of solid UF_6 inside a 5-in.-diam pipe with plastic windows. If not rapidly detected and controlled, significant uranium masses may be accumulated by condensation; however, recovery without equipment removal is readily accomplished by careful restoration of heat to sublime the UF_6 to the gas phase.

Finally, the mechanical failure of compressors may, in rare instances, result in temperatures due to friction which are sufficient to induce a reaction between UF_6 and metal surfaces to produce solid uranium tetrafluoride (UF_4). Temperatures in the range of 500°F to 550°F are required for this reaction.

NUCLEAR CRITICALITY SAFETY CONSIDERATIONS

From the standpoint of nuclear criticality safety and/or optimum production, it is necessary that any significant uranium accumulation be rapidly located and recovered. The criticality considerations of an accumulation depend upon the ^{235}U enrichment, mass, equipment geometry, and moderation. The ^{235}U enrichment can usually be determined or conservatively inferred from known operating parameters and history, and moderation can be controlled by appropriate procedures such as evacuating the equipment and purging with an inert gas. Presently the Oak Ridge Gaseous Diffusion Plant operates at enrichment levels of less than 5% ^{235}U and nuclear criticality safety is assured providing the accumulation remains unmoderated.[2,3] When uranium recovery requires equipment removal, moderation control is maintained by closing all openings in equipment containing an accumulation immediately upon disconnection from the cascade system. An inert gas bleed is applied and the equipment is carefully transported to a special dry room area in a uranium recovery facility for dismantling and recovery using established, nuclearly safe procedures.

INDICATIONS OF ACCUMULATIONS

Instrumentation and procedures essential to the efficient operation of a gaseous diffusion cascade normally will provide sufficient information that judgment of a possible accumulation can readily be made; such indications are considered sufficient to warrent spot check radiation surveys. Daily material input-output balances and periodic uranium accountability inventories will also provide indications of excessive material retention in the cascade system. These indicators have practical limits and the slow buildup of a significant uranium accumulation due, for example, to a small sustained atmospheric air inleakage may not be readily detectable. The need for gamma radiation detection equipment usable in a plant environment and sufficiently sensitive to the low intensity gamma fields produced by a ^{235}U enriched uranium deposition was early recognized. Appropriate equipment was developed and a program instituted to periodically survey the entire cascade system.

IONIZATION CHAMBER DETECTION EQUIPMENT

The basic ionization chamber instrument used for this service is shown in figure 5. It was developed about 1950 and consists of a 1/16-in.-thick wall stainless steel probe, 2 1/2 in. diam x 19 in. overall length, with a sensitive volume of 32 cubic inches.[4] The probe is cable connected to the vacuum tube circuitry of a battery powered, highly sensitive alpha particle survey meter developed earlier.[5] Initially, argon at 400 psig was employed as the probe's ionization media; xenon was used in a later modification.

In comparison tests between the argon and early xenon filled probes, a thin layer of highly enriched uranium oxide was used to provide a source of 0.185 MeV ^{235}U gamma radiation and radium, shielded by 1/4-in. lead, to simulate the higher energy gamma spectrum from ^{238}U daughter products. The argon instrument discriminated in favor of the lower energy gamma radiation by a factor of about 2 1/2 while the xenon probe discriminated by a factor of about 30. Heightened sensitivity at the lower gamma energies is advantageous since only the soft ^{235}U gamma field will be present at the site of a recent accumulation. Unfortunately, with the limited range of the ratemeter used, the xenon probe's sensitivity to ^{235}U gamma radiation was so great, 0.014 mr/hr on the least sensitive scale, that the instrument was essentially unusable in the process environment. Following changes in the probe input resistance and xenon pressure, the sensitivity was decreased by a factor of 5 to provide an instrument usable in the survey program.

SCINTILLATION CRYSTAL DETECTION EQUIPMENT

The argon and xenon probes have been in use for many years and have performed reasonably satisfactorily. Recently, however, a commercially available scintillation probe and ratemeter, shown in figure 6, have come under evaluation as a replacement for the ionization chamber instruments due to their age and increased maintenance requirements. Several advantages of the newer instrument include solid state circuitry and reduced size and weight. Although evaluation tests indicated the scintillation instrument to be four to five times more sensitive on a counts per minute basis than the present xenon-filled ionization probe, it has a greater ratemeter range, 40 times that of the xenon probe's ratemeter, and the increased sensitivity is usable for monitoring process equipment. Operational experience with the instrument is limited as yet; however, it appears that the time required to survey a given area will be greatly decreased.

In the process environment, gamma sensitive instruments give only qualitative indications of the presence of any accumulation. No reliable estimate of the uranium mass may be made from the measurement of gamma radiation intensities since readings are seriously affected by shielding effects of process equipment and self-absorption by the deposit. Further, a solid UF_6 condensation which has been present for some time prior to in-place recovery by sublimation back to the gas phase will leave behind ^{238}U daughter products, in the form of nonvolatile fluorides. These daughters may be detected as an accumulation even though no uranium is present. Thus, there was a need for an instrument to confirm the presence of an accumulation and determine its mass. Such an instrument was developed about 1962 based on the fact that diffusion plant accumulations are typically uranium-fluorine compounds.

NEUTRON DETECTION EQUIPMENT

A small neutron flux will be present at the site of a uranium-fluorine compound deposition due to the $^{19}_{9}F\ (\alpha,n)^{22}_{11}Na$ reaction where alpha

particles are supplied by decaying uranium, and the resultant neutron energy is in the 4- to 6-MeV range. The number of neutrons per second emanating from an accumulation bear a quantitative relationship to the uranium mass present; thus, the presence of an accumulation may not only be confirmed but an estimate of the uranium mass can also be made using neutron counting techniques.

Two neutron counting devices have been developed for plant service. Figure 7 illustrates the first, developed about 1962. The probe has a 2 in. diam x 12 in. sensitive volume filled with BF_3 enriched to 96% in the $^{10}_{5}B$ isotope at 40 cm Hg pressure surrounded by 1 3/4-in. paraffin to provide neutron moderation. The 22-lb probe is cable connected to a 14-lb case containing the scaler and electronic circuitry. The instrument is sensitive to a neutron flux as low as 0.005 neutron per square centimeter per second.[6]

Figure 8 shows the estimated masses obtained using the neutron counter for actual uranium accumulations.[6] Note that the upper limit of the estimated mass present is consistently greater than that actually recovered over a wide range of masses and ^{235}U enrichments, a conservative factor from a nuclear safety standpoint. Further, in one instance, no accumulation was present, as confirmed by neutron counting, the gamma activity detected being from uranium daughter products.

Revisions based on field use resulted in a completely redesigned neutron counter when a more sensitive detector filled with $^{3}_{2}He$ became available in 1965. The newer model shown in figure 9 weighs 25 lb, divided equally between the probe and the indicator unit. Other improvements over the original $^{10}_{5}BF_3$ counter include a ratemeter, built-in battery charger, and improved temperature tolerance.[7]

No published data on the $^{3}_{2}He$ neutron counter with actual uranium accumulations are available; however, a comparison of the response of the $^{10}_{5}BF_3$ and $^{3}_{2}He$ units to a 1 x 10^5 neutrons per second Pu-Be source shows the $^{3}_{2}He$ unit to be more sensitive by a factor of 2/3.[7]

THE ROUTINE SURVEY PROGRAM

The routine survey program at the Oak Ridge Gaseous Diffusion Plant requires that over 1500 process locations be checked twice annually. Computer printed listings of the locations to be surveyed per shift are provided to supervisory personnel for scheduling purposes at survey time. The results of the present and nine previous surveys are computer listed by date and location, along with the computed deviation from the trend indicated by each location so that suspect areas may be readily identified and investigated.

FUTURE PROBLEMS

Future problems in surveying a diffusion cascade for uranium accumulations center around ^{232}U. With the apparent proliferation of nuclear power, a time will come when uranium hexafluoride produced using uranium recovered from spent reactor fuel will be a significant fraction of the feed to gaseous diffusion plants.[8] Current Federal Regulations limit the permissible level of ^{232}U contamination in UF_6 delivered to AEC diffusion plants for enrichment under the Toll Enrichment Program to 0.110 part ^{232}U per million parts ^{235}U.[9]

The introduction of any significant quantity of ^{232}U and the growth of its radioactive daughters in plant equipment will render present gamma survey methods ineffective as well as hinder neutron counting due to the increased neutron background. The solution appears to lie in gamma spectrometry methods which can be used to detect the 0.185 MeV principal ^{235}U gamma peak in the presence of the 0.24 MeV gamma peak characteristic of ^{232}U daughter products. Equipment for this purpose has been developed and evaluated[8]; however, field experience is limited at present and the equipment is unwieldy compared to the existing equipment.

CONCLUSION

In closing, it is noted that observations of operating anomalies have, in many cases, led to the rapid discovery of uranium accumulations; however, the routine survey program, utilizing the instruments described, continues to be a proven method for locating slowly occurring accumulations. In anticipation of events yet to come, appropriate equipment has been developed to permit the program to fulfill its mission in the future.

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9. *Federal Register*, Vol. 32, No. 230, p. 16291, November 29, 1967.

PHOTO NO. ORO-70-400

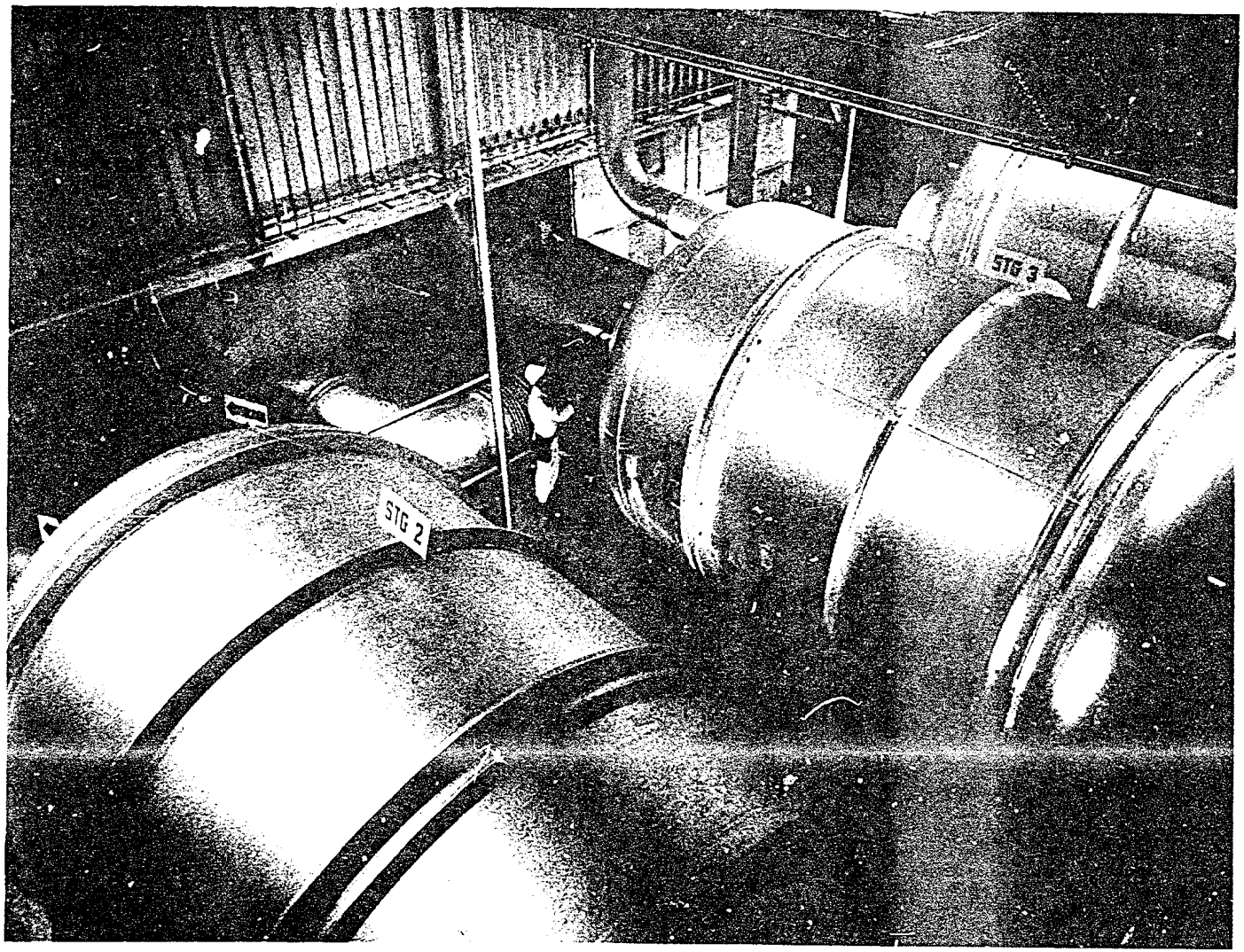

Figure 1

CLOSE VIEW OF COMPRESSOR AND DIFFUSERS

PHOTO NO. PH-69-1219

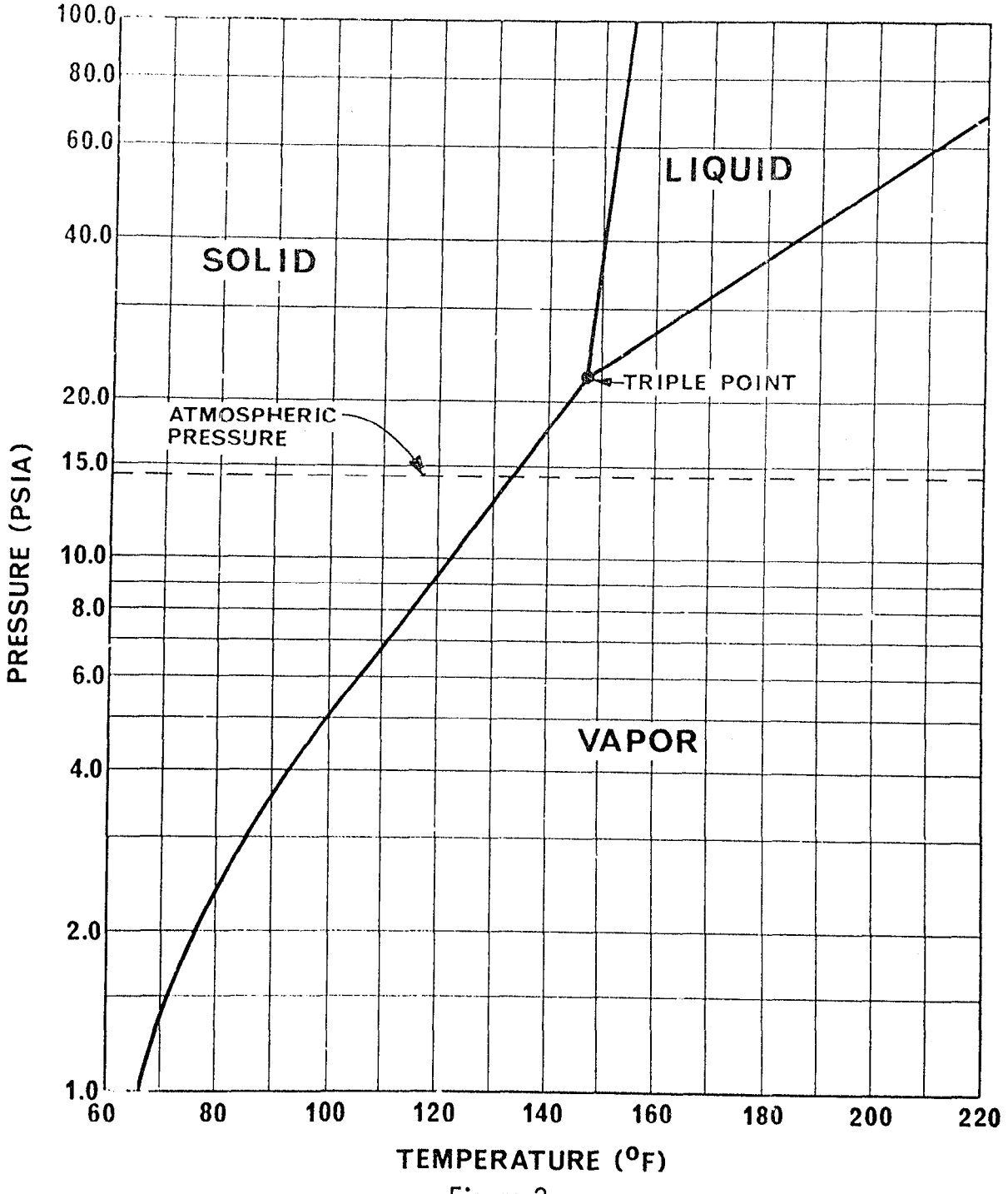

Figure 2

VAPOR PRESSURE OF URANIUM HEXAFLUORIDE

PHOTO NO. PH-72-698

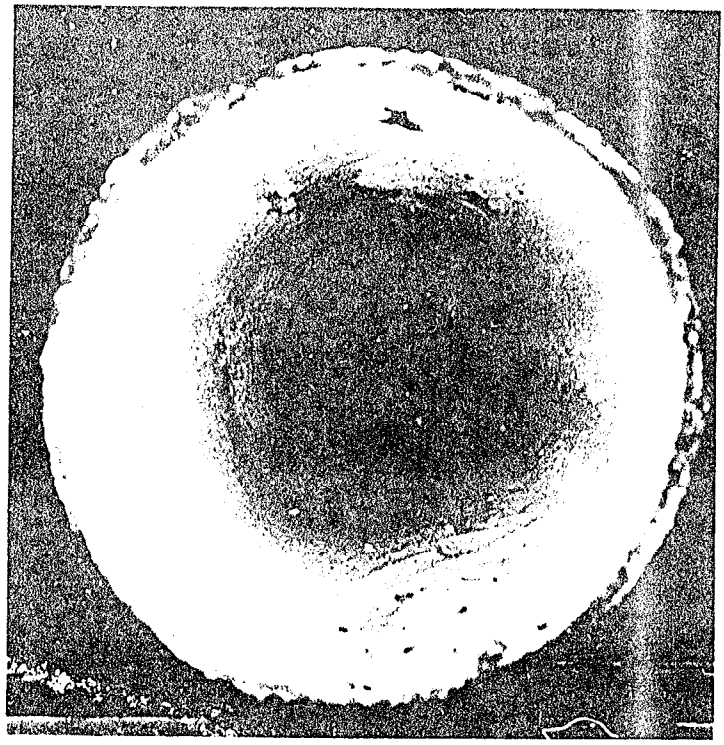

Figure 3
URANYL FLUORIDE DEPOSIT - 10-in. diam PIPE

PHOTO NO. PH-68-1556

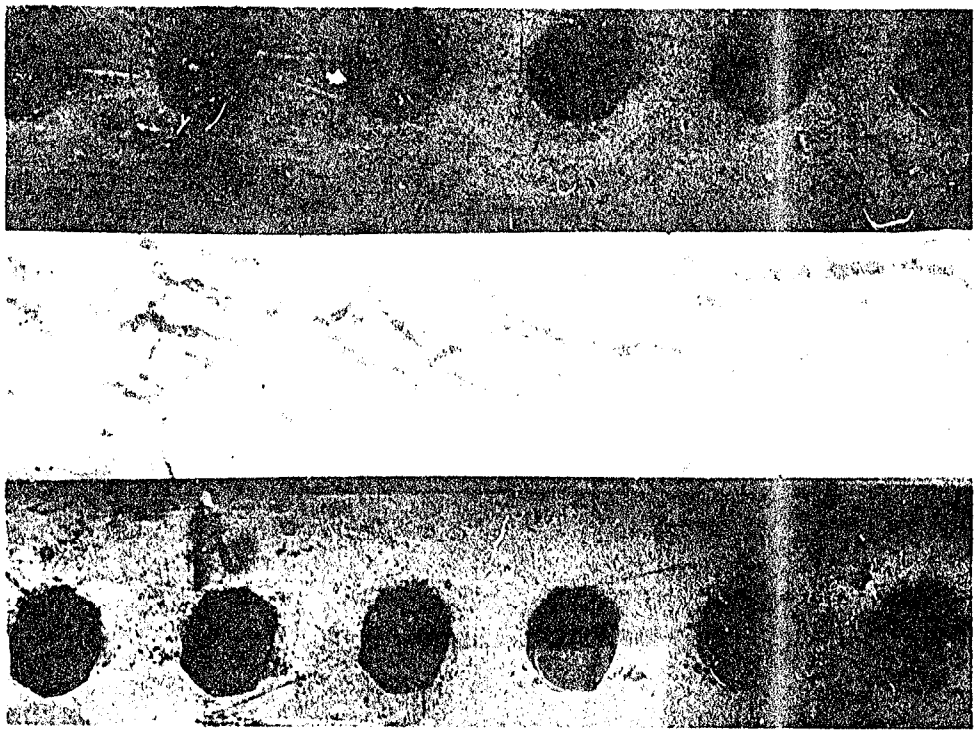

Figure 4
CONDENSED URANIUM HEXAFLUORIDE

PHOTO NO. PH-73-1055

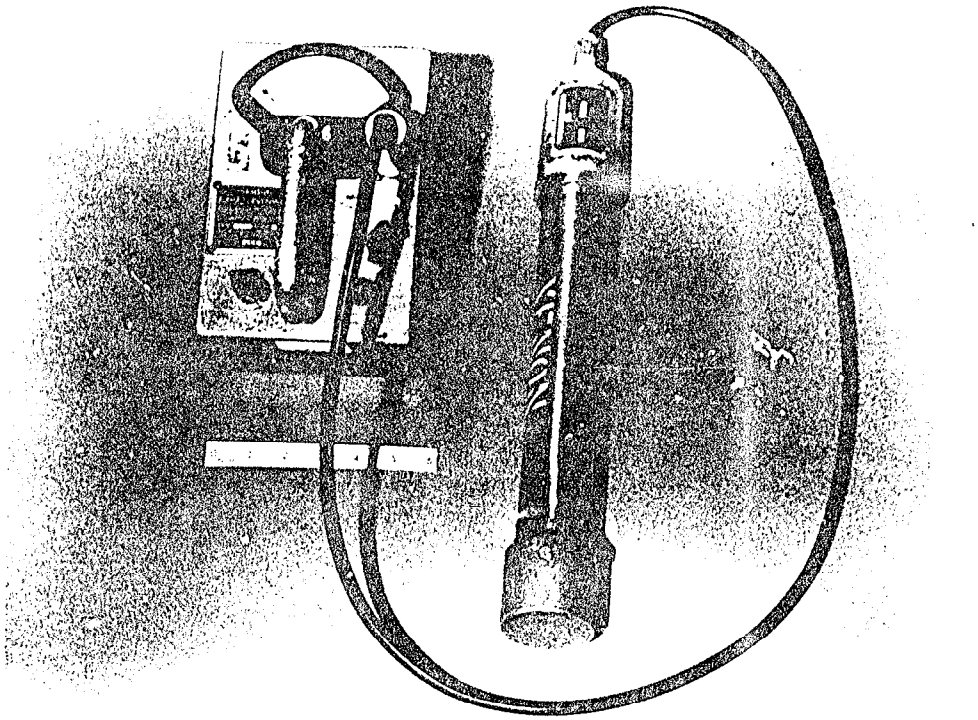

Figure 5
IONIZATION CHAMBER SURVEY INSTRUMENT

PHOTO NO. PH-73-1054

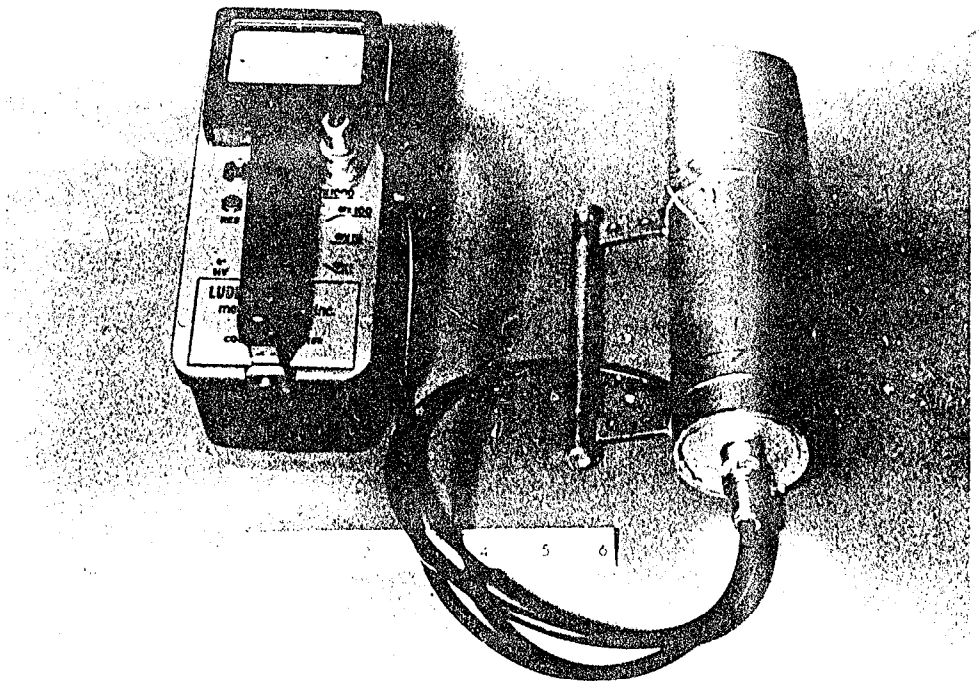

Figure 6
SCINTILLATION CRYSTAL SURVEY INSTRUMENT

PHOTO NO. PH-73-1056

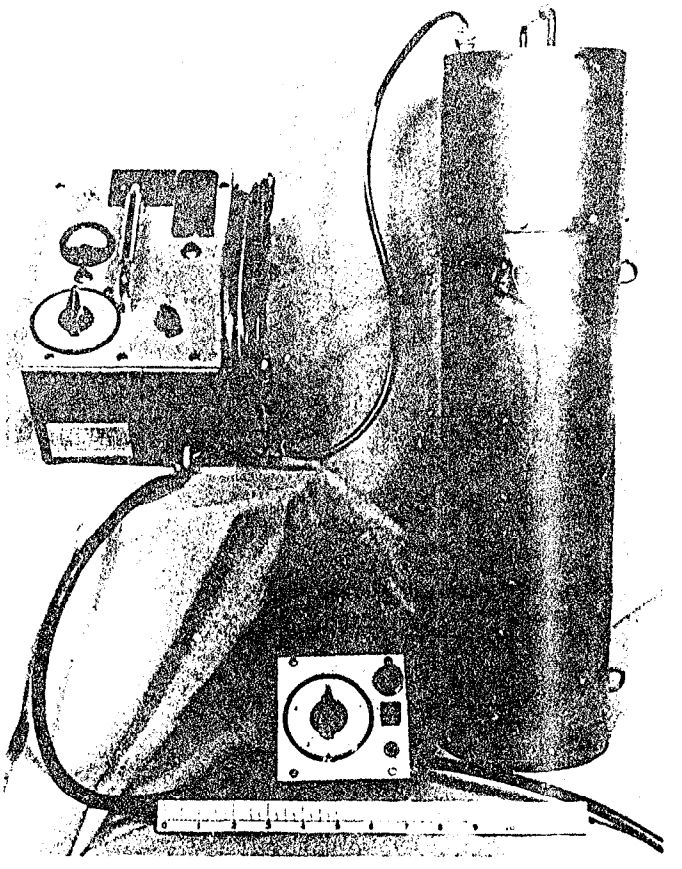

Figure 7
BF_3-FILLED NEUTRON COUNTER

DWG. NO. G-73-446

URANIUM COMPOUND	^{235}U ENRICHMENT (wt. %)	ESTIMATED WEIGHT (Pounds)			RECOVERED WEIGHT (Pounds)
		LOW	BEST	HIGH	
UF_6	0.8	-	590	-	500 ± 100
UO_2F_2	6.8	5	20	35	14.4
UO_2F_2	20	1.0	1.5	2.2	1.85
UO_2F_2	33	3.3	5.0	6.7	3.4
UO_2F_2	94	0.017	0.035	0.22	0.026
NONE (U daughters)	-	-	-	-	-

Figure 8
ACCUMULATION ESTIMATES MADE WITH BF_3-FILLED NEUTRON COUNTER

PHOTO NO. PH-65-937

Figure 9
$^{3}_{2}$He-FILLED NEUTRON COUNTER