Mound Laboratory, Miamisburg, Ohio

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TRIP REPORT To Gaseous Diffusion Plant and ORNL at Oak Ridge, Tennessee, February 26-27, 1959.

The first person I talked with was Lester Smith who heads a section in the Analytical Services Department. He said that gas sampling relative instruments using UF_6 would give greater precision analysis of uranium isotopes than a solid analysis system employing the surface ionization source. The surface ionization instrument would probably only give 1 per cent precision where there is a mixture of 1 per cent of U233 in U233-U238. The surface ionization instruments do not give a ratio reading directly but have to be shifted back and forth from one peak to another rapidly, while the emission is going down, to get readings on the various isotopes. A graph is made of the two isotope readings and a relative measurement is made. The instrument makes absolute measurements only, and it does not have much memory effect. Surface ionization instruments are probably best for plutonium.

On using a relative method of determining U235-U236, Mr. Smith said they were so close together that there would be considerable overlapping of the ion beams but that if measured against a known standard, the amounts in the mixture could be determined with good precision (0.1 per cent) if enough measurements were made on the sample to reduce the statistical error. Reports on this work are "Precisions and Biases Reports," which are issued quarterly, the latest being KL-45-5. The other is K-1369, which is unclassified, entitled "Measurement of the Isotopic Enrichment of Uranium by a Direct Comparison Mass Spectrometer Method," by V. L. Warren and L. A. Smith, which is on the same subject. I brought a copy of K-1369 back with me. UF_6 in the instrument causes considerable memory effect. They have installed a Vacion pump (~$1100) on one of their spectrometers which it is hoped will do away with using a liquid nitrogen cold trap and reduce the memory effect for UF_6 considerably. At ORNL they recommended a Veeco Ionization Gauge Type RG-75 and Control Type RG (~$425) for the same sort of job.

The uranium gas instruments used in Mr. Smith's labs are mostly G. E. Assay Machines from World War II days and two new ones from Consolidated. (Automatic Isotope Ratio Model 21-320A Lab Model 21-220A) Uranium gas instruments probably can be obtained from several companies, according to Mr. Smith. Consolidated builds a in-line instrument for $50,000 - $60,000 and a lab instrument for ~$50,000. Nuclides Analysis Associates at State College, Pennsylvania would probably build to order for $30,000 - $40,000.

The Bendix Aviation at Cincinnati, Ohio, build times-of-flight instruments and possibly would build UF_6 instruments. In the fall, after a new surface emission instrument is installed in the lab, Mr. Smith indicated that he should have a gas instrument for surplus.

There are several sources of surface ionization instruments. The Gaseous Diffusion Plant has ordered a 12-inch radius instrument from Nuclides Analysis Associates. The spectrometer has a vacuum air lock sample introduction system that allows a sample to be run every half-hour. The cost is $74,500 and delivery eighteen months, but delivery time will probably be reduced on future instruments. This company has men associated with them with considerable experience in spectrometry at various universities. C. E. C. makes a spectrometer for about $68,000 which has no vacuum air lock, and delivery in about six months. Probably, Metropolitan-Vickers makes the best instrument for about $50,000 with delivery in sixteen months which is due to instrument cost plus 28 per cent import duty, plus 6 per cent for Buy American, plus 1/2 per cent escalator, plus uncertainty in the pound and plus greater shipping cost. The instrument has the best vacuum air lock but does not have all standard parts. Mr. Smith would like to see the instrument bought in this country. The Atlas-Werke in Bremen, Germany makes an 8-inch radius instrument that Paducah liked very well. It is small but fairly adequate and has a favorable price. It can be converted to gas work readily.

Some work is being done with the emission spectrograph on isotope analysis at the Gaseous Diffusion Plant. Mr. T. E. Lee of the Works Lab in the Analytical Services Department is to give a paper at the Pittsburgh Conference on "Analytical Chemistry and Applied Spectroscopy," on March 6, at 4:00 p.m., concerning this work.

I talked to Mr. Smith and to Mr. J. C. Horton concerning problems we have had in preparing and trapping UF_6 from UF_4. In the preparation of the UF_6 there probably is a loss when the fluorine gas is pumped out since the UF_6 acts as an insulator on the wall of the condenser and there is probably some uncondensed hexafluoride. UF_6 cannot be pumped off if it is at dry ice-trichloroethylene temperature. These men believed that a 94 per cent yield was fairly good. They suggested the equipment should be pre-treated with fluorine gas according to the Hobbs-Macklin Stabilization Procedure, to reduce preparation losses due to corrosion of the metal lines by the UF_6. Reports on this procedure are K-670 and K-1132 and KLI-2823-2, "Handbook of Consumption" Ed. 2.

Fluorination of uranium compounds by liquid fluorine, as used by Mr. Smith, seems to be a simple straight forward method of preparing UF_6. The uranium compound is put into a single-end sealed copper tube which is connected to

a valve by a flare fitting. A liquid nitrogen trap is placed around the tube and fluorine liquified in the tube. The valve is closed, the liquid nitrogen trap removed and when the frost on the tube melts the uranium compound is converted to UF_6. If the uranium compound is too pure a "promoter" such as KCl is used to make the reaction go. A suitable shield must be placed around the reaction tube for protection in case of a rupture in the tube. A report describing this work is K-1267, "High Pressure Fluorination of Uranium Oxides," by R. E. Greene and G. S. Pettit.

Activated alumina and soda lime traps will remove UF_6 but there was some question as to the efficiency at reduced pressures. Reports on activated alumina and soda lime are KLI-673 and KLI-677. Sodium fluoride can be used to trap HF and reports on this are GAT-249 and KLD-328.

At ORNL, I talked to Ray McHenry of the Radioisotope Development Department, of the Radioisotope Division. Since they were in the midst of a crash program he was not able to give me as much of his time as he would have liked. As soon as the crash program is over he is going to finish a report on krypton-xenon separation, which has been partially written. There is a report CF-57-3-123, "Large Scale Separation of Fission Gases," by R. E. McHenry, Preliminary Draft. In it he discussed the various methods for separating krypton, xenon and the other gases from the fission process. He gave me a flow diagram, a process sheet known as the Product Loading Sheet-Krypton, Krypton-Xenon Process Equipment Description and Operation, and an excerpt from the report in progress which gives the "Effect of Composition," with krypton being used as the product gas. He said that the xenon gas that we are now receiving is much better than the first since they are now recycling through the carbon traps to remove carbon dioxide produced in the oxides of nitrogen decomposition. The xenon we are now receiving is about 7 parts in 10^7 krypton. He tried one more pass through the same traps and reduced the krypton to 7 parts in 10^9. To get higher purification he recommends that new carbon traps be used and at no time should traps used for colder work be used for hot work. This also, holds true for tritium work. Mr. McHenry believes the entire purification job could be done by gas chromotography. The type of carbon used is found in ORNL stores and is Fisher Scientific Company cocoanut shell activated carbon. The best they have found is Columbia Carbon Company Type G. Mesh size is 6-14. The carbon can be reactivated by heating to 500°-600°C and eluting with helium at 3 mm pressure for three hours. They use a reactor filled with carbon and heated to 1000°C to decompose the oxides of nitrogen. This trap is 5-inches in diameter and 25-inches long and filled with cocoanut shell carbon. The minimum length that is allowed for the carbon is 18-inches. They now use liquid nitrogen to cool the traps, but recommend refrigeration, as it would be much easier to control. Ion chambers and

Gow-Mac thermal conductivity cells are used as detectors for krypton-xenon and the other gases (known as air) before and after going through the traps. The cost of their chromatograph was approximately $40,000. A good portion of this money was used in lead shielding and remote controlling the process. They felt since they had built this unit on a crash basis the cost was much higher than what they could build now. ORNL-Q-1873-1 is a drawing of an ionization chamber of compact size, shielded and of good response according to McHenry. ORNL-Q-826A is a drawing of a very stable A.C. electrometer used to measure current from the ionization chamber. The one they were using was built by Nuclear Associates.

Bill Browning of Solid State Physics discussed carbon traps used in processing fission gases: the traps are used to give enough hold up time to allow the hot short lived isotopes decay. He gave me six papers on this subject, ORNL-2116, CF-58-6-6, CF-58-5-59, CF-58-12-10, CF-58-7-71 and CF-58-4-14.

In talking with Mr. George Creek it was found that ORNL probably has as much difficulty in counting gases as we do at Mound Laboratory. They do not recommend using counting tubes, which have been used for counting hot samples, for samples that have low count. A method of decontamination of the counting tubes was to alternately flush with sodium hydroxide solution and water. The counting tubes he used were of metal.

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