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TRIP REPORT - MEETING ON ISOTOPE SEPARATION

ARGONNE NATIONAL LABORATORY - MAY 8, 1961

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AUTHOR

E. E. Voiland

and

R. E. Tomlinson

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TRIP REPORT - MEETING ON ISOTOPE SEPARATION

ARGONNE NATIONAL LABORATORY - MAY 8, 1961

by

R. E. Tomlinson
Research and Engineering
CHEMICAL PROCESSING DEPARTMENT

and

E. E. Voiland
Chemical Research and Development
HANFORD LABORATORIES OPERATION

May 12, 1961

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HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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TRIP REPORT - MEETING ON ISOTOPE SEPARATION

ARGONNE NATIONAL LABORATORY - MAY 8, 1961

by

R. E. Tomlinson and E. E. Voiland

This meeting was called by Ed Grabowski of the AEC Division of Production, Washington. The principal purpose was to scope the chemical problems pertaining to the separation of plutonium isotopes by the gas centrifuge.

Grabowski gave a brief summary of the centrifuge program as it applies to uranium isotope separation. Briefly, it consists of the following:

1. At the University of Virginia (Kuhlthau), fundamental studies of centrifuges and properties of spinning rotors.

2. At Oak Ridge Gaseous Diffusion Plant (Garrett), studies of cascade parameters using current-design centrifuges.

3. Advanced centrifuge study including design, engineering and testing of an improved machine. This program has not been assigned. Seven firms, including General Electric (GEL) are being considered.

4. At Yale University (Onsager), theoretical hydrodynamic studies aimed at the flow problem within the centrifuge.

The program is under the administrative responsibility of the Oak Ridge Operations office. Its cost is $6 million. The goal completion date is the end of CY 1963. (The program originated under the Division of Research but in August, 1960, responsibility was transferred to the Division of Production.)

Grabowski stated that in their consideration, the most reasonable application of centrifugation as a means of separating isotopes was to plutonium. He also stated that the Division of Production wants to support work in this field.

After his opening remarks, Grabowski inquired what each group was doing concerning the plutonium problem. Oak Ridge had considered some of the problems, but had done no work and wanted to do no work with plutonium hexafluoride. From Burney's lack of participation, it is inferred that Savannah had little active interest. Argonne had done nothing directly supporting the plutonium isotope separation program, except, perhaps, extending their thermal decomposition studies down to 80 C. (This was a result of our visit to ANL in February.) Their PuF_6 program and earlier work in the Chemistry Division, however, has provided answers to many of the questions involving this application. In addition, their available facilities and experienced personnel put them in a strong competitive position for work of this sort. At Hanford, of course, no experimental work has been done. However, we appeared to be as well informed on the whole process and the problem areas as anyone present.

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At Grabowski's request, Vanstrum had prepared some information on the nature of the volatile material required, on possible alloys for construction and their limitations, and possible solutions to the corrosion problem by using throw-away liners or plated surfaces. There was nothing new here except that some of the more promising alloys are limited to operation below 200°F because of the type of heat treatment. He also presented the problem of removal of deposited PuF_4.

Fischer of ANL reviewed their work on decomposition, transport and fluorination of PuF_4. This work has appeared in published form and is reviewed in HW-68727. New work at 80 C showed only about one-fourth the thermal decomposition of PuF_6 expected from higher temperature studies. Thus, it appears that the principal decomposition will be alpha radiation induced.

Martin Steindler reviewed the radiation hazard involved in handling plutonium fluorides and presented an analysis based on Bill Roesch's studies. Calculations for plutonium derived from 10,000 MWD/T Dresden fuel inciated the following radiation levels close to a massive piece: 5.2 r/hr for gamma, and 4.7 rad/hr from fast neutrons. Also given were experimentally determined radiation levels from samples of PuF_4.

The alpha induced decomposition rate of PuF_6 was discussed by Malm, who pointed out the reaction is a function of composition and pressure. Of greater significance, however, is the dependence on fluorine pressure. In the presence of a small amount of fluorine, a steady state condition occurs at low temperatures wherein the PuF_6/F_2 pressure ratio is considerably greater than expected from thermodynamics. The importance of knowing more about this reaction is obvious, especially since deposition of AmF_3 in a centrifuge may perturb the intensity of the radiation field to which PuF_6 is exposed.

Discussion of the direction of future work occurred and was summarized as follows:

1. Continuation of decomposition studies of PuF_6; emphasis on 25-100 C range, pressures 0-1000 mm Hg, distinction of alpha-induced and thermal effects, surface effects on heterogeneous reaction and effect of materials on decomposition rate, etc. Basic thermodynamic studies.

2. Engineering feasibility studies particularly relating to containment and shielding. (Tomlinson)

3. Recovery or removal of PuF_4 (and AmF_3) from surfaces by chemical, mechanical, or fluorination techniques. Also, production, transport and handling of PuF_6.

4. Structural materials: strength, corrosion characteristics.

No work was suggested on other volatile plutonium compounds, which was in accord with our conclusion (from calculations based on the ANL kinetic studies) that PuF_6 would have sufficient stability. (For a typical centrifuge operating with a one percent per day decomposition rate, operation for 100 days would result in deposition of a quantity equal to the hold-up, or no more than 10 grams. The resulting deposit would be about $1 mg/cm^2$.)

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No definite allocation of the work was made, however, there was strong indication that ANL would be requested to submit a program on the kinetic studies. Steindler discussed a projected study on containment of PuF_6 and decomposition dusts that might form. He stated that they were doing the work with reluctance and would rather not be involved in the study but felt it is necessary. It is possible that Hanford might undertake this work, particularly if the engineering studies are assigned here.

The following individuals attended the meeting:

ANL - Chemical Engineering:

Jack Fischer
Stephen Lawroski
Martin J. Steindler
Albert A. Jonke
Victor H. Munnecke
Richard C. Vogel

ANL - Chemistry:

Paul R. Fields
John G. Malm

DuPont - SRL:

Glenn A. Burney

GE - HAPO:

Roy E. Tomlinson
Eugene E. Voiland

ORNL:

George I. Cathers

UCNC - ORGDP:

Harvey A. Bernhardt
George A. Garrett
Paul R. Vanstrum

USAEC - Chicago Operations:

Bruce Anderson, Jr.

- Oak Ridge Operations:

Walter E. McMahon

- Washington:

John E. Bigelow
Edward J. Grabowski

5-12-61

RE Tomlinson and EE Voiland:pct