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SUBJECT: PROGRAM OUTLINE - DEPLETED URANIUM UTILIZATION

TO: F. L. Culler, Jr.

FROM: J. C. Bresee

ABSTRACT

The present ORNL program to develop non-nuclear uses for depleted uranium is presented in detail. The related programs at other sites are described, and a mechanism is discussed for exchange of information and presentation of a summarizing report to be written in July, 1959.

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ORNL has started a program to develop non-nuclear uses for depleted uranium.¹ Depleted uranium is being produced by gaseous diffusion and stored by the AEC in very large amounts.^{2,3} While depleted uranium may ultimately be a useful fertile material for converter (plutonium recycle) or fast breeder reactors, the nuclear consumption rate for many decades will be small compared to its present rate of production. Either non-nuclear uses must be developed or the AEC will continue to have a large and expensive uranium storage problem.

The purpose of this memorandum is to present background material, to outline the present ORNL development program as well as related programs at other sites, and to list individual responsibilities for the completion of the first phase of the program (the remainder of fiscal 1959). The first phase of the program will end with a summarizing report to be written during July, 1959. A continuation of the program will depend on the results of the first phase.

Traditional Non-Nuclear Uses for Uranium

As an element uranium has had a comparatively short history. It was discovered in 1789 but for fifty years UO_2 was thought to be the pure element. Carbon was used to reduce UO_3 , and the metallic appearance of UO_2 contributed to the confusion.

The following was found in a book published in Pennsylvania in 1824: "Uranium is a scarce metal: its ores are of a blackish colour, generally containing iron, sulphur, lead, silice, etc. The metal is of a grey colour on the outside, and pale brown inside. It is very porous, and so soft that it may be cut with a knife; but more difficult to fuse than even manganese. Specific gravity, 6.400. It combines with a few of the other metals. To obtain uranium, the ore is heated to separate the sulphur, and then carefully cleared of impurities, after which it is digested in nitric acid, which dissolves the metallic matter: the solution is then precipitated by a carbonated alkali. This precipitate, or carbonate, (which is yellow,) is then made into a paste with oil, put into a crucible lined with charcoal, and exposed to a violent heat."⁴

The result, of course, was UO_2 . It is interesting to note that in 1958 a patent application was filed on behalf of the Y-12 development group for a process for production of UO_2 by placing uranium oxide in a graphite crucible and heating it to 1800°C (a violent heat).

In 1841 the metal was prepared for the first time by reduction of the tetrachloride with potassium. The atomic weight of uranium was not measured until 1882, and it was 1896 before it was discovered that uranium was radioactive.

Prior to 1932 uranium was used primarily as a coloring agent in ceramics. During the nineteenth century, use was made of the yellow, brown, and green compounds of uranium as pigments. Certain of the salts with fluorescent properties were of special value in ornamental ceramic glazes.⁵

As early as 1897 it was reported that the French Government was experimenting with ferro-uranium alloys in ordnance. The preparation of uranium steels has been studied for many years, but the high cost of uranium limited its application. As a substitute for tungsten in high speed steels, uranium is said to impart increased hardness without an accompanying increase in brittleness due to the formation of stable but dispersed carbides. Some investigators have enthusiastically claimed, "The benefits obtained from the use of uranium certainly constitute the highest attainment of the steel-maker's art."⁶

Anyone who studied qualitative analysis before World War II probably remembers the use of uranium as an analytic reagent for the determination of sodium. The formation of characteristic sodium zinc uranyl acetate crystals is said to be the most sensitive test for the detection of sodium ions in a solution with other alkali metals. Other uranium salts have been used for quantitative analysis of phosphates, arsenates, and ferric salts in the presence of molybdic acid.

In Haber's original patent claims for ammonia manufacture, metallic uranium and uranium carbide were listed among the best catalysts for reaction of nitrogen and hydrogen at high temperatures and pressures. Aside from cost, the greatest disadvantage of uranium catalysis has been sensitivity to impurities, primarily in the hydrogen. Other catalytic uses for

uranium include reduction, polymerization, desulfurization, and cyclization of petroleum fractions.

In the electronics industry before World War II, uranium was used as a "getter" in vacuum tubes, in photoelectric tubes for ultraviolet light detection, and as a target for x-ray tubes. Another interesting electrical application was the use of UO_2 in series with tungsten in powerful incandescent lamps to smooth out current surges and to prolong the filament life.

All commercial uses of uranium began to disappear in 1938 when the fission process was discovered. Consumption decreased from a high of 100 tons/year to a low of 1500 pounds/year in 1955. The enormous difference between the current depleted uranium supply and past commercial demands may be visualized by simple, unclassified considerations. Because natural uranium is 99.3% U-238 and conversion of more than 1% to Pu-239 is impractical without plutonium recycle in nuclear reactors, essentially all domestic uranium milled each year is accumulated as gaseous diffusion cascade bottoms. The current domestic rate of production of U_3O_8 is 15,000 tons/year.⁷

Future Nuclear Uses of Depleted Uranium

It is not sufficient to compare prewar consumption of natural uranium with postwar production of depleted uranium in order to demonstrate the need for the development of non-nuclear uranium markets. A very compelling argument is one which states that depleted uranium, like thorium, is a valuable natural resource, and should be "protected" by an artificially high price so that future generations may benefit by present conservation.⁸ In theory essentially all depleted uranium and thorium can be consumed in breeder reactors with an energy potential thousands of times that available from convertor or burner reactors. But just how important is present day conservation? Most particularly, how large will the nuclear market for depleted uranium be during the next forty years?

As in all such estimates, the most important (and unknown) parameter is the build-up curve for installed nuclear power. For the purposes of this memorandum, a mean was chosen between the curves of J. A. Lane and W. K. Davis. Using this mean curve, Arnold and Ullmann predicted the

total nuclear fuel requirements for consumption and inventory between now and the year 2000.⁹

A conservative estimate of the portion of the total nuclear power which will be generated by fast breeder reactors during the next forty years is 10 per cent. Based on the fertile material inventory in a well designed fast breeder reactor, Arnold has estimated that the total requirement between now and 2000 for depleted uranium will be 28,000 tons.¹⁰

Based only on domestic uranium production, the forty year requirements could be supplied in approximately two years. Note that assuming all power reactors are fast breeders only increases the domestic production time to twenty years with no changes in current production rates.

Depleted Uranium and Conservation

Clearly there are and will continue to be large quantities of depleted uranium for uses outside of nuclear power reactors. Battelle Memorial Institute will prepare a market survey and price analysis by July 1, 1959, similar to that prepared by Armour in 1957.⁸ However, it would be useful to consider for a moment the basic philosophy behind "protection" of depleted uranium by an artificially high price.

Real conservation implies that cheaper substitutes should be employed wherever possible for all critical materials rather than nuclear fuels only. Critical materials would include those elements with unique physical or chemical properties which make them irreplaceable for certain important applications.

Consider the case of tungsten. The United States was not blessed with a large share of the world's supply of tungsten (or more properly, wolfram). In fact, more than half of the world's ore reserves are in China.

A typical high speed steel may contain 18 weight per cent tungsten. In early ferro-uranium alloy studies, it was claimed that a 3 per cent uranium, 8 per cent tungsten steel had properties equal to those of the 18 per cent steel.⁶ More than 1000 tons of tungsten are used annually for high speed steel manufacture. One could postulate a replacement of 500 tons of tungsten with perhaps 200 tons of uranium.

Although alternate fuels are available for power production, at present there is no adequate substitute for tungsten in filaments of incandescent lamps. In the example quoted, it would be possible to argue that conservation of natural resources demands that non-nuclear uses for depleted uranium be developed.

The Organization of the Present Development Program

The current development program can be divided into three parts. The first consists of a review of the toxicological and radiological properties of depleted uranium. The second and third parts are literature surveys and experimental studies of the metallurgical and chemical uses of depleted uranium.

Depleted Uranium Toxicology and Radiology - The review of the toxicological and radiological properties of depleted uranium will be carried out by B. R. Fish of ORNL and G. R. Patterson, Jr. and C. M. West of the Y-12 plant. The purpose of their review will be to establish the effect of assay on the necessary controls for safe handling of various forms of depleted uranium. They will also examine the new Code of Federal Regulations (dated May 2, 1959 - effective June 2, 1959) as it applies to the licensing agreements for industry and compare the limitations therein with those in the 1958 recommendations by ICRP.¹¹ Based on their findings, it may be possible to suggest changes in the licensing agreements which would reduce the difficulty of handling depleted uranium without increasing the danger. The results of their survey and the recommended changes will be incorporated in the summarizing report.

Handling of depleted uranium results in two types of hazards: from external radiation and from inhalation or ingestion. Uranium with an assay of 0.2% U-235 contains a minute but important quantity of U-235¹¹ (approximately 0.0015%). Having been through chemical refining and gaseous diffusion, the uranium would be essentially free of metallic impurities and in particular it would be free of daughters of radioactive decay (except for Th-234 and Pa-234 which reach equilibria in ~ six months).

It is easy to show that depleted uranium with an assay of 0.2 is only a minor hazard as a source of external penetrating radiation. Consider an

infinitely long cylinder of depleted uranium of infinite wall thickness and a two foot inside diameter chamber¹¹ in which a man might be unlucky enough to find himself. The Th-234 and Pa-234 beta particle emissions make up essentially all of the penetrating radiation in the chamber. The 2.32 mev β^- from Th-234 alone contributes 240 mrem/hr while the 0.2 mev β^- from the Pa-234 adds 20 mrem/hr.¹² All gammas from all disintegrations are quite soft and add only 5 mrem/hr to the total of approximately 265 mrem/hr. A recommended maximum occupational exposure in 13 weeks of 3 rem would be accumulated in slightly over 11 hours.¹¹

It is difficult to imagine a more favorable geometry for maximizing total body irradiation. All actual working environments (e.g., during the fabrication and installation of small uranium counterweights) would result in decreases in penetrating radiation intensities of several orders of magnitude.

The dangers from inhalation or ingestion of depleted uranium depends on the solubility of the inhaled or ingested material. Soluble uranium is absorbed in the kidney and acts as a chemical poison, blocking the reabsorption of vital constituents of the blood and contributing to a loss of body fluids. The damage is reparable by the body and the biological half life for uranium removal from the kidney is only 6 days.

Insoluble depleted uranium is only dangerous to the lungs and the mechanism of damage is from alpha bombardment of the tissue. The repair of such damage is slow, and the biological half life for uranium removal is much longer (approximately 120 days).

The control of inhalation or ingestion is basically the control of air-borne uranium. According to the latest recommendations of the International Commission on Radiological Protection, the maximum permissible concentration of natural uranium (soluble or insoluble) in the air breathed 40 hours per week is 200 micrograms per cubic meter.¹¹ Since the alpha activity of pure U-238 is approximately a factor of two lower than that of natural uranium, there would be no substantial increase in the limitations for insoluble and no change for soluble depleted uranium.

It is instructive to compare the limitation on air-borne quantities of natural uranium to those of two other common heavy elements: lead and

mercury. The maximum permissible concentrations of these materials in air are 150 and 100 micrograms per cubic meter.¹³ Thus controls normally applied to the industrial uses of these materials would be entirely adequate for depleted uranium.

Depleted Uranium Metallurgy - The current metallurgy development program may be subdivided into AEC associated programs and independent research and development efforts. The role of ORNL will be to collect and analyze metallurgical data from the literature and to share information with the other sites.

A representative of the ORNL Metallurgy Division, Professor Gregg of Cornell has undertaken a literature survey of uranium alloys. It is not known at present whether his survey will be available before the end of the first phase of the metallurgy development program (the remainder of fiscal 1959), but if the program is continued next year the information will be quite valuable for all in AEC associated programs.

S. D. Clinton will be the ORNL metallurgy coordinator during phase one. He will be responsible for collecting, analyzing, and interpreting data from the survey programs at Y-12 and ORGDP and for comparing the results of development studies at various Bureau of Mines stations. The mechanism for the sharing of information from ORNL and the Bureau of Mines is being arranged through the AEC Oak Ridge Operations Office. His findings will be reported in the summarizing report.

The Y-12 survey program is under the direction of C. P. Coughlen. For several years he has collected data on uranium metallurgy and the properties of various uranium alloys. In the course of his work he prepared a useful analysis of shipping cask factors as part of a program proposal for the development of depleted uranium shielding for fuel element shipping casks.¹⁴ He will attempt to extract unclassified data from his notes in a form useful for the preparation of a report at the end of phase one.

The ORGDP survey program is the responsibility of C. H. Mahoney and W. S. Dritt of the K-25 Metallurgy Department. Several months ago they requested the technical library of ORGDP to prepare a bibliography of uranium metallurgy, and they will screen the collection for information

useful to the current program. Several of the most important collections of data on uranium and its compounds have been prepared at K-25 in the past.^{15,16,17}

The Bureau of Mines stations at Rolla, Missouri and Albany, Oregon are engaged in several interesting metallurgical studies of uranium.¹⁸ The Rolla studies are under the joint sponsorship of the AEC and the Bureau of Mines, while the Albany studies are supported by the Bureau alone.

The work at Rolla is under the direction of Dr. James Rowland, Chief Metallurgist, with the assistance of Dr. D. S. Walsh, formally of the Missouri School of Mines. They are investigating the preparation and properties of copper, tin, and lead alloys of uranium as possible constituents of soft, Babbitt-type bearings. Even though phase diagrams are available for essentially all mixtures of interest, little is known about the crystal habits and the means of modifying the crystal structure of the mixtures.

Depleted uranium metal is being considered at Rolla as a replacement for magnesium for cathodic protection of underground pipe lines and similar iron or steel structures. Even though there is little possibility that uranium would ever be as cheap as magnesium, particularly on an equivalent weight basis, the major cost of cathodic protection is said to be the cost of installation and maintenance of the anodes. The electrochemical potential of uranium relative to steel will be measured under simulated natural conditions (using, for example, earth boxes). The minimum anode area will be determined for adequate corrosion protection.

At Albany, Dr. Alva Roberson, Supervising Metallurgist, and Dr. Lloyd H. Banning, Project Coordinator for Ferrous Metallurgy, are carrying out an investigation of the ferrous alloys of uranium. As was mentioned earlier, the early literature contains reports of ferro-uranium alloys of remarkable properties.¹⁹ Their usefulness as high speed tool steels was strongly suggested. The Albany program will be sufficiently broad that these and other uses for uranium steels will probably be considered.

One problem associated with the use of uranium in the steel industry should be pointed out. A large fraction of the steel produced each year

is steel recycled as scrap. If special melts of uranium steels are produced for particular applications, care will have to be taken to prevent uncontrolled recycle of the steel. Even though the total U. S. steel production exceeds 100 million tons, small amounts of uranium in steel could have serious effects in certain application (e.g., steel cans for photographic film). Even the danger of radioactive steel could affect the steel market adversely.

Among the remaining independent research and development studies of the metallurgical uses of depleted uranium, the most important are those of the Nuclear Metals Division of National Lead Company.²⁰ With the Company's years of experience in lead metallurgy and uranium refining, it is logical that immediate consideration has been given to depleted uranium shielding. One potential application under consideration during the past year was the use of depleted uranium in the reactor shielding for the Nuclear Ship Savannah. Another very interesting development was the possible substitution of depleted uranium for tungsten alloys as counterweights for airplane ailerons. The latter application has been hampered by current uranium licensing restrictions.

It has been recently disclosed that U. S. Steel has been studying uranium steels at their Monroeville, Pa. laboratory.²¹ No results are available from their work.

Chemical Uses of Depleted Uranium - The chemical studies of the application of depleted uranium are being carried out at many AEC sites, in a joint program with the Bureau of Mines, and in an independent Bureau project.

At ORNL, P. A. Haas and S. Peterson share the responsibility for a thorough survey of the literature, the evaluation of available results from experimental programs conducted here or other sites, the conducting of limited but definitive experiments during the remainder of the current fiscal year, and the preparation of a summarizing report during July.

A group headed by Dr. R. V. Lundquist, Project Leader, is studying uranium solutions and slurries for heavy-media separations of minerals of different densities at the Boulder City, Nevada station of the Bureau of Mines in a program jointly sponsored by the Bureau and AEC.

In a parallel program at ORNL, Peterson has concentrated his efforts thus far on (a) the assembly of a file of methods of preparation and properties for all uranium compounds, and (b) the preparation of dense aqueous solutions of uranium halides.

Three grams per cubic centimeter is a useful density at room temperature for an ore dressing heavy media. The density of individual sand particles is slightly less than three, so it would be possible to float sand from heavier minerals with such a liquid. In preliminary experiments, Peterson has prepared a stable uranyl bromide solution with a density of 3.3 g/cc.

Another very attractive chemical use for depleted uranium is as a catalyst. At the Laramie (Wyoming) Petroleum Research Center, the Bureau of Mines is conducting an independent investigation of the use of uranium as a hydrogenation catalyst. The project leader is Dr. P. L. Cottingham. The chemical form of the uranium is an oxide supported on alumina, and the reactions of interest are the cracking and hydrogenation of high sulfur and nitrogen petroleum and shale oils.¹⁸ Although such an observation may be premature, the large scale use of uranium as a catalyst in the petroleum industry would create a very large market for depleted uranium.

P. A. Haas is responsible for the ORNL program on the catalytic uses of depleted uranium. In addition to conducting a thorough literature survey, he has recently written to six large commercial catalyst manufacturers, outlining the ORNL program and soliciting expressions of interest in conducting independent research with uranium compounds provided by ORNL.

University research, particularly doctoral research, may be the most appropriate method of carrying out basic research on uranium catalysis. In answer to an inquiry from Haas, Dr. L. Berg of Montana State College proposed an AEC-sponsored program on uranium catalysis as applied to petrochemicals, with two doctoral thesis to begin next year and a total annual program cost of eight to ten thousand dollars.²² Due to the uncertainty of next years program, it was suggested to him that he enter a separate proposal to the AEC Division of Research.

Another interesting possibility involves Haas directly. During the current calendar year, he will begin a doctoral thesis in chemical engineering under the supervision of Professor H. F. Johnson of the University of

Tennessee. One possible thesis topic is the investigation of the use of uranium catalysis in certain industrially important chemical reactions.

Two other general classes of chemical use of depleted uranium will be considered by Peterson and Haas. These are inorganic ion exchange and the preparation of ceramics.

In his letter of April 13, 1959, George Rennich suggested the use of uranyl and silver salts to desalt sea water.¹⁸ Advantage would be taken of the formation of insoluble sodium uranium compounds and silver chloride.

The following is an example of how a uranium compound might be used as an inorganic ion exchange reagent. Hydrated UO_3 may be thought of as precipitated uranic acid - H_2UO_4 or $\text{H}_2\text{U}_2\text{O}_7$ ($\text{UO}_3 \cdot \text{H}_2\text{O}$ or $2 \text{UO}_3 \cdot \text{H}_2\text{O}$). If a hydrated UO_3 slurry were titrated with caustic, formation of insoluble sodium uranate or diuranate would result in the replacement of sodium ions in solution with hydrogen ions, a basic requirement for desalting. Sodium hydroxide could be produced from salt water by the adsorption of chloride ions on hydrated silver oxide.

Regeneration of the silver and uranium "resins" obviously would be necessary. However, the two regeneration processes would not be equally easy. Although chloride could probably be removed from hydrated silver oxide by repeated strong caustic washes, the removal of sodium from uranium oxide with an acid would probably result in the dissolution of the uranium. The example quoted may not be technically feasible and, on the surface at least, appears to be uneconomic.

Another proposed use of uranium ion exchange made recently by H. B. Whetsel appears to be both feasible and practical.²³ In Whetsel's proposal, advantage is taken of the insolubility of the mixed uranium cesium phosphate: UO_2CsPO_4 . Since the cesium compound is less soluble than its sodium counterpart (UO_2NaPO_4), uranyl sodium phosphate would probably adsorb cesium-137 ions from radioactive waste solutions, and the "resin" could be regenerated with strong caustic.

Uranium oxides are possible components of interesting ceramic systems for non-nuclear uses. In addition, pure UO_2 which melts at 2700°C may be a useful ceramic for high temperature coatings, crucibles, or fire bricks. A depleted UO_2 crucible might be a convenient container for melting and holding alloy mixtures of depleted uranium metal.

Finally, a chemical use for depleted uranium might be developed by utilizing UF_6 as a source of fluorine. This potentiality has aroused the interest of several sites associated with AEC programs. The ORNL fluoride recovery program is jointly the responsibility of C. D. Scott ($\text{UF}_6 \longrightarrow \text{UO}_2$) and S. Peterson (UF_6 as a fluorinating agent).

Using existing technology, it is relatively simple to use the dry reaction developed at ORGDP: $\text{UF}_6 + \text{H}_2 \longrightarrow \text{UF}_4 + 2\text{HF}$ to recover one third of the fluorine in UF_6 as anhydrous HF. Recovery of the remainder in a useful form is quite difficult, since UF_4 is an unreactive compound. Pyrohydrolysis to UO_2 : $\text{UF}_4 + 2\text{H}_2\text{O} \longrightarrow \text{UO}_2 + 4\text{HF}$, requires excess water and produces dilute hydrofluoric acid. However, azeotropic distillation as at Weldon Springs, Missouri might be used to recover strong hydrofluoric acid.

A much more interesting and potentially cheaper reaction is the one-step reduction and hydrolysis of UF_6 : $\text{UF}_6 + 2\text{H}_2\text{O} + \text{H}_2 \longrightarrow \text{UO}_2 + 6\text{HF}$. It may be possible to produce greater than ninety per cent hydrofluoric acid (above the azeotrope) by the selection of the proper temperature and pressure for the reaction. The one-step reaction is currently being studied at the Paducah Gaseous Diffusion Plant, where the work began in 1958, at ORGDP, at Argonne, and at ORNL. The approach that C. D. Scott has taken at ORNL is to hydrolyze UF_6 continuously to a uranyl fluoride solution and to spray the solution into a hot fluid bed of UO_2 in hydrogen.

Scott is planning a trip late in June to all the sites studying the reaction in order to compare the approaches and to summarize the status of the work at the end of the fiscal year.

The possibility of utilizing UF_6 as a fluorinating agent is being investigated by S. Peterson. There is a great deal of appropriate early project literature in that much attention was paid to the stability of organic compounds in the presence of UF_6 . The most common reason for rejecting a compound was a reaction to form a fluorocarbon. The research led to the development of a new industry and important plastics such as Teflon and Kel-F.

Uranium hexafluoride may react with unsaturated compounds such as acetylene to form useful raw materials for the present fluorocarbon industry. The

uranium product would doubtless be UF_4 , but the recovered fluorine might have a higher value than anhydrous HF.

The Preparation of the Summarizing Report

The purpose of the summarizing report will be to present the current status and to recommend the future of the AEC depleted uranium program. The report will be written during the month of July, 1959 and will cover the topics discussed in this memorandum plus others added during the month of June.

The editor of the report will be J. C. Bresee and the main contributors will be S. D. Clinton, P. A. Haas, S. Peterson, and C. D. Scott. The mechanism for an exchange of information with sites outside Oak Ridge will be trips to Paducah, Weldon Springs, and Argonne by C. D. Scott and trips to Albany, Oregon; Boulder City, Nevada; Laramie, Wyoming; and Rolla, Missouri by S. D. Clinton, P. A. Haas, and S. Peterson. All trips will be made during the period: June 18 - 28, 1959.

The necessary arrangements for information exchange with the Bureau of Mines stations will be made by the Oak Ridge Operations Office of AEC. The visits by Scott are being arranged directly by letter.

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