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IONIUM, URANIUM-232, AND THORIUM-228
PROPERTIES, APPLICATIONS AND AVAILABILITY

by

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INTRODUCTION

Although uranium-232 and its daughter thorium-228 have extremely unusual properties which may find very useful applications, consideration for such uses (applications in which the intense heat, alpha emission and neutron generating potentials of these isotopes could be utilized) have been largely neglected because of the assumed unavailability of the basic source material thorium-230 (ionium). The expanded and diverse uranium milling industry particularly with many ores in the U.S. indicates that ionium may indeed be available in quantities sufficient to justify consideration of the production of substantial amounts of these potentially useful isotopes. The following charts present information which illustrate the properties of these materials, compares these properties with competitive isotopes, and reviews the availability of the parent material ionium (thorium-230).

These charts, diagrams and tables are the result of modest efforts conducted over the past year within the Hanford Laboratories to support a proposed research and development program for which funds have been requested. Qualified personnel highly experienced in chemical processing techniques, fuel fabrication, work with alpha emitting materials (plutonium) and in irradiation processes together with related experimental and production facilities essential for study of these materials make for a logical decision to conduct this proposed work at Hanford.

CHART INATURE OF THE DECAY CHAIN FOR URANIUM-232

Whereas the common radioisotopes which are receiving most attention as sources of heat (Sr-90, Pm-147, Pu-238, Ce-144) involve a single or at most only two steps in their decay to stable materials, uranium-232 goes through ten steps of radioactive decay. Seven of these steps involve energetic alpha emissions. Thorium-228 is the first member of this decay chain and is the only one other than the parent material (U-232) to have a half-life greater than a very few days, most being measured in seconds and minutes. It is of considerable significance as shown on the chart that one kilogram of U-232 in one year produces 7.7 grams of thorium-228. In view of this phenomenon it is considered feasible to assume a stored stock of uranium-232 as a source of multi-gram amounts of thorium-228.

RADIOACTIVE DECAY OF U-232

* 7.7 GRAMS Th-228 PER YEAR FROM STORAGE
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Whereas the common radioisotopes which are receiving most attention as sources of heat (Sr-90, Pm-147, Pu-238, Ce-144) involve a single or at most only two steps in their decay to stable materials, uranium-232 goes through ten steps of radioactive decay. Seven of these steps involve energetic alpha emissions. Thorium-228 is the first member of this decay chain and is the only one other than the parent material (U-232) to have a half-life greater than a very few days, most being measured in seconds and minutes. It is of considerable significance as shown on the chart that one kilogram of U-232 in one year produces 7.7 grams of thorium-228. In view of this phenomenon it is considered feasible to assume a stored stock of uranium-232 as a source of multi-gram amounts of thorium-228.

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CHART II

HEAT GENERATING CHARACTERISTICS OF URANIUM-232 AND THORIUM-228

Isotopes considered for heat generation purposes can be grouped into two general classes, according to half-lives.

Among those with short half-lives, (less than three years) thorium-228 is not only the most energetic emitter, but its half-life is more than five times greater than that of its nearest competitor, polonium-210.

Among isotopes with long half-lives, (more than ten years) U-232 is almost nine times as energetic as most competing products.

The principal deficiency of U-232 and Th-228 as heat sources relates to their gamma energy component. However, on the basis of total heat output U-232 and Th-228 are slightly less gamma active than cerium-144, which is now receiving major consideration as a heat source. Among isotopes with little or no gamma energy the heat potentials are relatively low.

In the utilization of all alpha emitting isotopes provision must be made for helium gas evolution. The alpha particles from the U-232 and Th-228 decay will require that the device in which these are used can handle the released helium gas.

COMPARISON OF SPECIAL ISOTOPES

ISOTOPE	HALF-LIFE	HEAT GENERATION, WATTS PER GRAM			
		ALPHA	BETA	GAMMA	$\alpha + \beta + \gamma/2$
SHORT HALF - LIVES					
THORIUM - 228 *	1.9y	153.2	3.2	9.1	161.0
POLONIUM - 210	138.0d	141.0	—	—	141.0
CURIUM - 242	163.0d	119.7	—	0.2	119.8
CERIUM - 144 (PURE) *	285.0d	—	20.25	1.25	20.9
CERIUM - 144 (RECOVERABLE) *	285.0d	—	4.7	0.3	4.8
PROMETHIUM - 147	2.6y	—	0.4	—	0.4
LONG HALF - LIVES					
URANIUM - 232 *	74.0y	4.17	0.07	0.21	4.34
STRONTIUM -90 (PURE) *	28.0y	—	0.78	—	0.78
STRONTIUM-90 (RECOVERABLE) *	28.0y	—	0.5	—	0.5
PLUTONIUM - 238	90.0y	0.54	—	—	0.54
TRITIUM	12.3 y	—	0.34	—	0.34
KRYPTON - 85	10.4 y	—	0.53	0.01	0.53
RADIUM - 226 *	1620.0y	0.15	0.004	0.03	0.17

*INCLUDES OUTPUT OF RADIOACTIVE DAUGHTERS

CHART IIISUSTAINED HIGH HEAT OUTPUT OF URANIUM-232

In heat generating characteristics U-232 is unique in that it maintains a high level of output for a large fraction of its half-life. However, it does not reach the maximum level until after about five years since its date of recovery (chemical separation). This latter deficiency can be readily overcome by blending initially a small amount of Th-228 with the freshly recovered U-232. The accompanying chart illustrates this technique.

As can be seen, a device containing U-232 plus a small amount of Th-228 could achieve a steady heat output (plus or minus ten percent) for a period of over 30 years. This feature may be especially advantageous in certain applications.

CHART XIIRRADIATION CHARACTERISTICS OF IONIUM AND THORIUMTHE NEED FOR HIGH IONIUM CONCENTRATION IN RECOVERED THORIUM

Because of the wide difference in thermal neutron absorption cross sections the irradiation process by which ionium would be converted to uranium-232 (via Pa-231) will tolerate ionium which may be highly contaminated with natural thorium without excessive wastage of neutrons in the contaminating materials. The accompanying chart illustrates this problem. Although there is a fortunate relationship in the cross sections which favors ionium conversion the desirability is still strong for ionium with as little thorium in it as possible for the first irradiation stage in which protactinium-231 with a minimum amount of Pa-233 is desired with also a minimum possibility for losses in early formation of unsegregated U-232.

CHART XI**IRRADIATION CHARACTERISTICS OF IONIUM AND THORIUM - 232**

CHART XIIORIGIN AND FATE OF THORIUM AND IONIUM IN ORES AND MILL STREAMS

Insufficient data are at hand to establish precisely the fate of thorium and ionium in the mill process, however, the accompanying chart is considered as representative of the general situation.

A large portion of U.S. uranium ores are considered to have originated by displacement and deposition from percolating waters. Thorium is not regarded as having the geologic mobility characteristic of uranium. The uranium was therefore deposited in structures in which natural thorium already existed. Since deposition then, ionium "grew in" in close association as the daughter of uranium-238 (via U-234).

In the ore mill process (sulfuric acid leach) the ionium would be expected to go into solution along with the associated uranium. However, any resident thorium which may exist as thorite (a silicate) or monazite (a phosphate) would be expected to go into solution only partially. Thereby a degree of isotopic enrichment of the ionium-thorium mixture would be achieved in the mill dissolution process. Some analytical results support this expectation.

Most of the thorium is expected to appear in the waste liquors from the uranium extraction step (as either solvent extraction or ion exchange wastes). A portion may follow the uranium product. This could very probably be the situation, if the mill process used an alkali to remove the uranium from the solvent. Another portion may be retained by the stripped solvent and then be recycled or removed in any solvent treatment step. Further study is needed to establish conclusive and useful data on this feature of the problem.

CHART XII

DISTRIBUTION OF IONIUM IN AN ACID URANIUM MILLING PROCESS

CHART XIIIIONIUM IN URANIUM MILL WASTES AS A FACTOR IN
RADIOLOGICAL HAZARDS TO THE PUBLIC

Although no maximum permissible concentration figure has been formally established for ionium, 5×10^{-8} microcuries per milliliter has been proposed. The figure for radium is 4×10^{-9} . The much higher solubility of ionium in the acid leach uranium ore milling processes appears to overbalance the more liberal proposed limit such that for acid mill wastes the alpha hazard from ionium may be higher than that from radium. This may, however, not be the case in the alkaline leach processes in which ionium would be expected to have minimum solubility. Work of others (WIN-112) shows no thorium to be solubilized in the alkaline leach process.

The alpha emission characteristics of equilibrated uranium ores are complex and involve at least two radioactive decay chains (and three if the ore is highly contaminated with natural thorium (Th-232)). It should be understood that in equilibrated ores the alpha activity of uranium-238, ionium and radium are equal. Because of the much shorter half-lives of members of the U-235 decay chain, Th-227 and Ra-223 are not considered to be significant in evaluation of the long term public health aspects of mill waste disposal. Although it has a long half-life, the apparent immobility of Pa-231 justifies neglecting it also in this consideration. The expected distribution of the alpha emissions from uranium-238, ionium (Th-230) and radium-226 in a typical one ton per day acid leach uranium mill is shown on the accompanying chart. The importance of ionium as the major source of alpha activity in soluble wastes is thus conclusively exhibited.

CHART XIII

DISTRIBUTION OF ALPHA EMISSIONS FROM URANIUM-238, IONIUM,
AND RADIUM-226 FROM A TYPICAL ACID LEACH MILL PROCESS

Basis: One short ton of uranium per day.

Alpha emissions per minute from U-238, Io (Th-230) and Ra-226.

*This shows that in the soluble waste fraction the alpha activity from the ionium (Th-230) may be ten times that from the radium. In a process where greater than 50% of the ionium is solubilized the alpha activity from ionium in the extraction wastes would even be greater.

CHART XIVMANUFACTURING COST ESTIMATES

The use of ionium (comprising a mixture containing about 10% ionium and 90% natural thorium) for the production of U-232 from which Th-228 may be periodically recovered could involve a complex series of steps with many possible alternate routes. There is little background on which to base estimates of costs for such a production scheme. The accompanying table, however, is an effort at arriving at a realistic figure. These figures are considered to be high and assume use of existing facilities under government sponsorship. No credit is taken for the pure U-233 which may be produced as a by-product, neither amortization, cost of special facilities such as shipping casks, nor profit are assumed in these costs. These figures should not be used as the basis for the study of routes which may be felt to be more economical. As our own studies become more firm and as more information is obtained which may influence costs our estimates will be revised. Once an accumulation of U-232 has been obtained, very little extra cost will be involved for the recovery of the Th-228. Th-228 then should cost only slightly more per gram than U-232. We have considered a figure of \$20/gram of Th-228 as appropriate. Thus the estimate indicates a cost of \$390 per gram for uranium-232 and \$410 per gram for Th-228. This figure should be compared with the "values" shown on the earlier charts. It is of interest to recall that recently British workers recovered about one hundred grams of Pa-231 from uranium ore processing residues. The cost of this material was listed as \$2,800 per gram. The U.S. AEC was negotiating for the purchase of about half of this material. Ionium therefore may also be an attractive source of relatively inexpensive Pa-231 (\$230 per gram).

CHART XIVURANIUM-232 (AND THORIUM-228) PRODUCTION COST ESTIMATE

Bases: The raw material will be ionium in the form of a Th-232-Th-230 mixture containing 7 to 10% ionium. Such mixtures will cost \$20 per gram of contained ionium.

For each gram of U-232 recovered two grams of ionium will be consumed.

Each gram of Pa-231 will be converted to one gram of U-232.

<u>Operation</u>	<u>Cost Basis</u>	<u>Cost/Gram of U-232</u>
Ionium (raw material)	\$20/gm Th-230	\$ 40.00
Fabrication of Ionium "Fuel"	\$10/gm Th-230	20.00
Irradiation (Th-230 to Pa-231)	\$120 (cost of neutrons)	120.00
Age and Separate (Pa-231 and Recycled Th-230)	\$25/gm Th-230	50.00
Fabrication ("Hot") (Pa-231 and Th-230)	\$50/gm Th-230 / Pa-231	100.00
Irradiate (Pa-231)	\$30 (cost of neutrons)	30.00
Age and Separate (U-232 and Recycled Pa-231)	\$30/gm	<u>30.00</u>
Total Cost of U-232/gram		\$390.00
Age and Separate (Th-228 from U-232)	\$20/gm Th-228	<u>20.00</u>
Total Cost of Th-228/gram		\$410.00

CONCLUSION

This preliminary survey including chemical analysis of uranium mill ore feeds and process liquors strongly indicates that ionium concentrates could be recovered at relatively low cost from a large fraction of the U.S. uranium ore milling industry. With such material thus suggested to be available annually in multikilogram quantities consideration has been given to applications, specifically for those new products, uranium-232 and its daughter thorium-228 which could be produced via neutron irradiation of ionium. These latter materials have extremely unique properties for use as long-lived heat sources for thermoelectric power generation; they are intense alpha emitters which may find wide application as economical neutron sources; they may also find use in special luminescent materials and in radiography. These outstanding and unusual properties justify continued and expanded efforts on the recovery and exploitation of ionium and its related conversion products.

The skills and equipment available within the Hanford Laboratories particularly with regard to the processing, manipulation and utilization of alpha emitting elements together with the outstanding irradiation facilities in the adjacent research and production complex strongly support the continuation of research and development and production studies on ionium and its unique irradiate products by the Hanford Laboratories.