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Nonproliferation Alternative Systems Assessment Program (NASAP)—Preliminary Environmental Assessment of Thorium/Uranium Fuel Cycle Systems

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NONPROLIFERATION ALTERNATIVE SYSTEMS ASSESSMENT PROGRAM (NASAP) - PRELIMINARY ENVIRONMENTAL ASSESSMENT OF THORIUM/URANIUM FUEL CYCLE SYSTEMS

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ABSTRACT

This report presents a preliminary assessment of the quality of existing information available for the evaluation of potential environmental impacts resulting from large-scale implementation of a thorium-based fuel cycle. The report's purpose includes (1) assistance in the development of a hazard assessment policy for the Nonproliferation Alternative Systems Assessments Program (NASAP) sponsored by the Department of Energy, and (2) identification of areas in which further research is necessary to allow detailed evaluation of the environmental hazards associated with thorium fuel cycles in general. Both the hazard assessment data base and the available assessment methodology are evaluated.

While this report does not present coverage of all issues pertaining to practical thorium fuel cycles and pertinent literature, it is an attempt to specify those issues likely to appear to be significant during an exhaustive hazard analysis.

INTRODUCTION

This report has been prepared in response to a request from the Division of Nuclear Research and Applications of the U.S. Department of Energy (DOE). The purpose of the report is to present the results of a survey of existing information on the environmental impacts of thorium/uranium commercial nuclear reactor and fuel cycle systems, examining the current state of knowledge of potential environmental impacts for these systems.

The report consists of a discussion identifying significant differences between conceptual uranium-plutonium (U/Pu) and thorium-uranium 233 (Th/^{233}U) breeder fuel cycles, with emphasis on those aspects which appear to require additional field/laboratory research effort. Given the time constraints under which this preliminary document has been prepared, it should be viewed as an identification of major research needs, rather than as a detailed evaluation. It is anticipated that ongoing assembly and review of pertinent literature at Oak Ridge National Laboratory (ORNL) will result in a comprehensive hazard evaluation of the Th/^{233}U fuel cycle, including a detailed thorium mining and milling assessment to be published in 1978.

While significant generic differences do exist between the U/Pu and the Th/^{233}U fuel cycles in terms of potential environmental hazard, it is not anticipated that serious difficulties in the licensing of system components will be encountered for either fuel cycle. When compared to U/Pu systems, Th/^{233}U concepts may possess the prospect of reduced

hazards in certain portions of the fuel cycle, including the mining and milling of ores, and the long-term management of high level wastes. With respect to other fuel cycle operations, including fuel processing and fabrication, transportation, and reactor operation, variations in hazards exist, but appear to be minor in magnitude. It is clear, however, that current interest in thorium-based fuel cycle systems requires additional research and analysis before complete confidence in these initial conclusions is possible. To assist in defining this research, a compendium of Th/^{233}U-related work has also been prepared, entitled, "Biomedical and Environmental Aspects of the Thorium Fuel Cycle".[1]

PRELIMINARY ASSESSMENT OF THORIUM/URANIUM FUEL CYCLE ENVIRONMENTAL HAZARDS

Mining and Milling

Currently the major foreign availabilities of thorium for the U.S. are Malagasy and the Elliot Lake area in Canada. Thorium deposits in the U.S. include monazite beach placers along the Atlantic Coast, and thorite veins in the western mountain states.[2] The major U.S. uranium deposits occur in New Mexico and Wyoming. The mining of western thorite deposits would involve open pit or deep-mine methods similar to those for uranium mining. If thorium monazite deposts along the Atlantic Coast are to be exploited, however, the potential ecological and socio-economic impacts are likely to be considerably different from those encountered in the western U.S., for reasons of climate, population density, etc. It is evident, therefore, that the future demand for

thorium may qualitatively as well as quantitatively affect the environmental impacts of thorium mining and milling.

Few analyses considering the environmental impact of mining and milling thorium ore are available, although interest in this area is currently increasing. The unpublished document, "Environmental Survey of the High-Temperature Gas-Cooled Reactor Fuel Cycle - Thorium Mining, Milling, and Refining,"[3] has been found to be incomplete. The model facility design in this study was not clearly documented and utilized assumptions possibly oversimplifying analyses of problems associated with mining and milling. The Science Applications Inc. draft final report, "Environmental Impact Analysis of Thorium Nuclear Fuel Cycles,"[4] has been reviewed in detail at ORNL, and was found to focus insufficiently on the unique aspects of thorium mining and milling.

The most comprehensive generic assessment to date of mining and milling of thorium and uranium ore is presented in ERDA 1541,[2] the light-water breeder reactor (LWBR) impact statement. In that report, a combined mine-mill complex is conceptualized for the purpose of estimating environmental effects. Key environmental impact variations in a comparison of mining and milling of thorium vs. uranium ores are found to include land use and quantity of liquid waste discharged. It is estimated that a significantly smaller land area (per unit energy-equivalent produced) would be disturbed for the LWBR thorium cycle since the grade of the thorium ore is higher, the ore is contained in veins of substantial width, and a larger number of veins lie parallel within a typical work pit area. The amount of waste water discharged (also on a per-unit-

energy basis) from a thorium mine is also estimated to be considerably reduced, indicating that off-site effects from discharge may be reduced depending on site-specific tolerances to mining activities. Due to the low solubility of thorium and its daughters in water, concentrations of radionuclides in liquid effluent streams from the thorium mine and mill may be orders of magnitude less than for uranium mine liquid effluents, although a study to compare suspended particulate concentrations is necessary to supplement this estimate.

It was estimated in ERDA 1541 that the quantities of radioactive materials released to the atmosphere during conceptualized thorium mining and milling would be greater than those released from comparable uranium facilities. A hazard in both mining and milling of thorium ore might be the radiological impact resulting from the release of ^{220}Rn and daughter particulates to the atmosphere; however, the short radiological half-life of ^{220}Rn reduces this hazard.

Although the assessment of thorium mining and milling presented in ERDA 1541 is a good overview with respect to the Light-Water Breeder Reactor Program, radiological dose estimates specific to the mine/mill components of the fuel cycle are not calculated in the report. Current emphasis on thorium, as utilized in one or more of the various reactor cycles, requires the completion of a generic mining and milling document.

In view of similarities in mechanical and chemical methods between mining and milling thorium and uranium ores, sufficient data are probably available[5] to allow reliable estimates of thorium mine and mill radionuclide and chemical effluent releases to the environment. Although

uncertainties still exist in environmental transport data and dose conversion factors for certain radionuclides, as discussed later in this report, it appears that a useful generic environmental assessment can be assembled at this time. Additional research on the environmental and metabolic behavior of certain radionuclides, as outlined within this report, should result in substantial reduction of uncertainties associated with the estimation of environmental hazards in such an assessment.

Fuel Reprocessing

A comparison of U/Pu versus Th/^{233}U reprocessing indicates few significant differences in anticipated environmental hazard levels. It may be confidently assumed in either case that routine operation of reprocessing facilities would result in: 1) no radioactive solid wastes buried on site, 2) no radioactive liquid releases, 3) controlled radioactive gaseous releases, following cleanup to ensure that releases are well below regulatory limits, and 4) minor differences in hazard associated with chemical effluents or release rates.[6-11]

The most important volatile or gaseous radioactive isotopes expected to be emitted from Light-Water Reactor (LWR) reprocessing plants are ^{3}H, ^{85}Kr, ^{14}C, and ^{129}I.[11,12] Tertiary fission in fast reactors may result in greatly increased ^{3}H levels at the reprocessing facility. High Temperature Gas-Cooled Reactor (HTGR) plants would release additional ^{14}C via the burning of the graphite matrix of irradiated fuel elements. Due to the fission product spectrum modifications resulting from the use of ^{233}U rather than ^{235}U fuel, a Th/^{233}U reprocessing facility would be expected to release increased quantities of ^{85}Kr and iodine. Table I

indicates estimated relative fission and activation product inventories for HTGR versus LWR fuel elements. Given the range of Th/^{233}U options under current consideration, this table provides a comparison adequate for the purposes of this assessment.

Table I. Estimated major volatile fission and activation products in fuel elements (180 days after reactor discharge)

Isotopes	Ci/(GW_eY)	
	HTGR[a]	LWR
^{3}H	13,300	22,900
^{14}C	159	16
^{85}Kr	711,000	372,000
^{129}I	2.0	1.3

[a] Ref. 12.

Containment of gaseous ^{3}H and ^{85}Kr is deemed feasible.[12] Containment of the iodine isotopes is a more difficult problem but also appears to be feasible. The separation of $^{14}CO_2$ from large quantities of inactive CO_2 involves greater technical difficulty and may not be economically justifiable.[12,13] A thorough study of population doses resulting from large scale use of HTGR's is, therefore, of importance. Current models

do not include sufficient complexity to reliably model worldwide distribution, fixation, and population doses related to $^{14}CO_2$ (ref. 2), although significant progress is evident.[13]

With respect to particulate releases, conceptual Th/^{233}U reprocessing facilities utilize deep-sand and HEPA filtration techniques to reduce particulate emissions by at least 10^{-9}(ref. 2,14,15), sufficient to comply with current standards. Additional isotopes which may be unique to HTGR reprocessing are volatile forms of ^{35}S and ^{33}P, (ref. 12) both pure beta emitters, created by neutron activation of the HTGR fuel pitch binder. Further research is necessary to determine the levels of release and environmental hazard imposed by these nuclides and suitable control measures.

Research investigating potential environmental hazards resulting from deliberate production or introduction of gamma-emitters into fuels (for safeguards purposes) prior to refabrication is necessary, as is a thorough investigation of the hazards related to prolonged or repeated irradiation of recycle materials, with consequent buildup of low cross section transmutation products.

A hazard introduced by Th/^{233}U fuel reprocessing and refabrication (as compared to U/Pu facilities) would be accidental release of ^{232}U, ^{233}U, ^{228}Th, ^{232}Th and daughters. Behavior of these isotopes in man and the environment is insufficiently understood, as discussed in following pages. Increases in hazard from the introduction of these isotopes might be offset to an unquantified extent by a significant reduction (80-90% in LWBR fuels) in quantities of transuranic isotopes produced,

including neptunium, plutonium, americium and curium.[16] Plutonium, for example, will be present in LWBR spent fuel only as a trace, due to a reduced production cross section via ^{232}Th. A relative hazard analysis of the cycles, based on this tradeoff, should be performed. The effects of ^{238}U if added as a denaturant must be included in this analysis.

In the past, relatively short-term storage of spent fuel elements was envisioned prior to reprocessing. Experience with LWR fuel indicates that a reevaluation of spent fuel average storage time may be in order. Increased storage time, prior to reprocessing, effectively reduces hazards from short and intermediate half-life isotopes, including ^{131}I, ruthenium, ^{95}Zr-^{95}Nb, and certain transuranics. In a comparison of 160-day-versus three-year-stored LWR spent fuel, total radioactivity fed to a reprocessing plant was estimated to be reduced by a factor of about 5 (ref. 17). The likelihood of a similar reduction in hazard for the Th/^{233}U cycle should be considered.

Solid Waste Management

A unique hazard introduced by the Th/^{233}U fuel cycle involves the additional uranium isotopes (^{233}U and ^{232}U), thorium isotopes (^{228}Th and ^{232}Th) and their daughters (the thorium chain). The introduction of these isotopes may be offset by a significant reduction in transuranic wastes. In a recent comparison[16] including U/Pu and ^{233}U fuel cycles, it is concluded that waste "radiological hazard" from Th/^{233}U (primarily fission products for the first 300 years) may be up to three orders of magnitude less than from corresponding U/Pu cycle wastes. Alpha heat production, a significant storage problem, may be reduced by up to five

orders of magnitude when compared to U/Pu cycle wastes. Comparative analysis of long-term effects for the two generic cycles' waste management policies may, therefore, be possible at this time. Again, the effects of denaturant ^{238}U must be considered in this context.

Weinberg et al.[18] have prepared an excellent comparison of various fuel cycle technologies, including estimates of long-term hazards of concern in this context.

Specific research needs essential to reliable estimation of waste management environmental hazards also include the development of data quantifying the environmental and metabolic behavior of the uranium and thorium chain radionuclides, and calculation, based on available geochemical data, of uranium, thorium, radon, and thoron and daughter geotransport coefficients. Continued development of methodology appropriate to ^{3}H, ^{14}C, and ^{85}Kr waste hazard assessment is urged. Entrapment of these nuclides during reprocessing operations will result in chemically fixed or physically compressed materials for storage. These entrapped radionuclides may be gradually released, and population dose estimate methodologies should be perfected to handle large-scale storage and subsequent potentials for release.

A Discussion of Research and Methodology Development Needs

Atmospheric transport

Most computer-implemented models used for estimating doses to man from atmospheric releases of radionuclides do not provide for contributions to external doses resulting from daughter buildup in the plume. Daughters which attain secular equilibrium with their parents very quickly after

release of the parents can be simply treated by adding them to the source term itself. Cases in which a very short-lived parent produces a longer-lived daughter can also be handled for specific downwind distances in an approximate manner by adding the daughter to the source term at an appropriate release rate. More serious problems arise, however, in cases for which the parent and daughter half-lives are such that secular equilibrium is not achieved within the dose assessment area or in cases in which conversion of the parent to a longer-lived daughter is far from complete by the time the plume reaches a point of interest. The existence of a chain of daughters can complicate these problems. The decay of ^{220}Rn ($T_{1/2}$ = 55s) is a case in point. There is a need to be able to treat the buildup of ^{212}Pb ($T_{1/2}$ = 10.64 h), ^{212}Bi ($T_{1/2}$ = 60.6 m), and ^{208}Tl ($T_{1/2}$ = 3.1 m) in the plume. The computer modeling problem is not trivial because, for example, plume depletion through dry deposition and scavenging processes, included in advanced atmospheric dispersion computer codes, involves use of time-dependent deposition and scavenging coefficients.

Another related and non-trivial problem involves the applicability of current atmospheric transport models to mountainous terrains, such as those surrounding many western potential mine/mill sites. Currently applied dispersion codes do not consider the effects of terrain variability on pollutant dispersion.

Accurate modeling of radiological doses from alternative nuclear fuel cycles, therefore, requires additional effort in estimation of time dependent coefficients, in model development, and in validation of resultant methodology.

Ecosystem transport

Because high specific activity uranium ($^{233}U/^{232}U$) and thorium (^{228}Th) are present extensively within thorium fuel cycles, the environmental transport of these radionuclides requires thorough evaluation. The lack of field data on the behavior of uranium and thorium in aquatic and terrestrial food chains has been a major shortcoming in assessments of environmental transport of these elements. Dose estimates to individuals and populations in the vicinity of facilities releasing these materials contain considerable uncertainty as a result. Aside from geochemistry, where extensive documentation of uranium and thorium behavior exists, little is known about the ecosystem dynamics of natural uranium and thorium. In recent years, the research emphasis on plutonium as a potential environmental problem from the Liquid Metal Fast Breeder Reactor (LMFBR) fuel cycle has overshadowed research on the environmental behavior of other actinides.

Geochemically, uranium and thorium in the +4 oxidation state have similar properties,[19] but uranium in the +6 state (UO_2^{+2}) is significantly more soluble than thorium. Thus, the behavior of uranium in the freshwater and marine environment is not easily predictable. Regardless of the ionic aquatic environment, thorium, like plutonium, tends to accumulate in organic and inorganic sediments more than does uranium. In soils, thorium is geochemically less mobile than uranium.[20] The movement of uranium from soils to the oceans is relatively rapid.

Soil to plant transfer coefficients for impact assessments are typically based on naturally occurring concentrations of uranium and thorium

in soils and plants. Terrestrial plants preferentially assimilate uranium over thorium, but fractional uptake by plants for total soil uranium or thorium is unknown. Isotopes of uranium or thorium freshly added to soil by fuel cycle operations may be more available than natural uranium and thorium; more research in the area of soil to plant transfer is needed. While concentration ratios for plutonium have now been measured in a variety of terrestrial environments in the U.S.[21] and for a variety of plant species, there are few comparable studies of soil to plant transfers for uranium or thorium. The geographic range in plutonium concentration ratios is 10^{-4} to 10^{-1}, and similar variations might be expected for uranium and thorium.

Even less research has been attempted on the potential transfer of uranium and thorium from plants to animals or animal products. It is anticipated that the fractional transfer of uranium, thorium, and plutonium to milk and meat is small; estimates of these factors are available.[22] A similar lack of data is evident regarding uranium and thorium transfer to poultry and eggs, the necessity for field research is evident. An equally important shortcoming in predicting food chain transport to man is the lack of information on thorium concentrations in human foods.

Little information is available on the aquatic food chain transfer of uranium and thorium to man. There is no evidence for biomagnification of thorium or uranium in aquatic food chains, but thorium/uranium ratios are typically greater than unity at each trophic level. Uranium and thorium concentration factors for fish and seafoods consumed by man are virtually unexplored.

Finally, many of the parameter values required to predict the environmental transport of uranium and thorium using available methodologies have not been measured specifically for these elements. Consequently, generic parameter values are frequently substituted into models of food chain transport. Given that the consumption of plant foods is a primary route of the actinides ingested by man,[23-25] parameters used to predict uranium and thorium concentrations in vegetation are of considerable importance. For example, deposition velocities (Vg) are commonly used to predict radionuclide concentrations in vegetation via air concentrations. There are no field measurements of deposition velocities specific to uranium, thorium, or plutonium compounds under a variety of atmospheric conditions. Additionally, there are no measurements of effective half-time (a function of physical half-life and weathering half-time) for plutonium, uranium, or thorium deposited on vegetation.

It is clear, therefore, that research on food chain transport of uranium and thorium compounds is required for more accurate estimation of hazards via environmental transport.

Further research on the environmental behavior of radionuclides other than thorium/uranium and daughters would also contribute significantly to a more thorough assessment of thorium fuel cycle systems. These radionuclides include ^{14}C and ^{99}Tc as well as several of the lesser known actinides, such as americium, neptunium, and curium.

Metabolic and toxicological studies

Extensive use of ^{232}Th as a fertile material implies exposure of the occupational worker and the general public to several radionuclides

(previously noted) not normally present in large quantities in uranium/plutonium fuel systems. Several areas of research have been identified which, if undertaken, would contribute to reduction of uncertainties associated with the assessment of potential environmental impacts of thorium fuel systems. The following summarized recommendations were largely presented at the Conference on Occupational Health Experience with Uranium,[26] but have also been suggested by Till[27,28] and in the Light-Water Breeder Reactor Program Final Environmental Statement.[2]

Metabolism of uranium and thorium. For protection of both occupational workers and the general public, it is important that we obtain more accurate estimates of parameters in uranium and thorium metabolic models, such as (1) the absorption of uranium and thorium from the gastrointestinal tract, (2) the long-term residence characteristics of uranium and thorium in bone (probably requiring research with a long-lived mammalian species), and (3) the quantities of uranium and thorium in soft tissues after prolonged exposure. Human studies based on large numbers of individuals via whole-body counting and post-mortem examination should assist in overcoming familiar extrapolation problems associated with animal studies.

Chemical toxicity. Animal experiments are needed to examine the following: 1) the longevity, functional status, and ability of the kidneys to withstand further stress after acute or chronic uranium poisoning, and 2) the separate and/or synergistic effects of chemical toxicity and radiation damage due to isotopes or mixtures of isotopes of high and moderate specific activity, such as ^{233}U, ^{232}U and highly enriched uranium containing ^{234}U and ^{235}U.

Radiation effects. Because up to now only low-specific-activity uranium isotopes and mixtures have been encountered in the nuclear industry, their radiation effects have generally been ignored. Long-term radiation effects in bone and kidney for the high-specific-activity isotopes ^{232}U and ^{233}U have been studied only in mice,[29,30] and need to be studied in large, long-lived species such as dogs or monkeys. The dose-response relationships in lung need to be determined for a range of specific activities of inhaled insoluble uranium compounds, to aid in understanding the general problem of the effects of inhaled insoluble alpha-emitters.

It has been noted[2,31] that we currently assume all bone-seeking radionuclides to be five times more effective in inducing bone tumors than ^{226}Ra (i.e., n = 5). Only a limited number of studies have been conducted with ^{233}U and only one with ^{232}U (ref. 29). These limited data suggest that for high specific activity uranium, setting n = 1 for ^{233}U and n = 2 for ^{232}U may be justified, effectively increasing exposure limits for these isotopes.

In conclusion, it appears that additional metabolic and toxicological data, both human and animal-derived, focusing on those radionuclides unique to thorium fuel systems, would be helpful in assessing the potential environmental impact associated with the use of thorium fuels.

Dose conversion factors

Dose conversion factors used in environmental assessments often involve large uncertainties. Outdated metabolic and retention models are too frequently employed in the computation of these factors. For

many elements of importance in the thorium and uranium fuel cycles, no more recent metabolic models are available than those of the 1959 ICRP Publication 2.[32] Metabolic data and some dose conversion factors for radionuclides which would be released to the environment in the event of a nuclear power plant accident have been compiled by the Nuclear Regulatory Commission in the WASH-1400 report.[33] Other tabulations of dose conversion factors include that of Killough and McKay,[34] the NRC's Regulatory Guide 1.109,[35] and NUREG-0172 (ref. 36), a compilation of age-dependent factors. Frequent discrepancies in the values from these various sources are evident and can often be traced to differences in metabolic models and assumptions.

There is a need for further development of dose conversion factors applicable to age groups in the population other than the reference adult. The approach to this problem has generally been one of extrapolating the adult dose conversion factors to the child by means of a simple ratio of organ masses. However, other physiological, metabolic, and dietary differences would likely produce variations in uptake and retention of radionuclides among age groups.

The internal dosimetry of thorium and uranium isotopes and their progeny has many complexities; the dose conversion factors for these radionuclides may require reevaluation. One particularly difficult problem is the handling of radon species produced in these decay chains. Radon-220 and 222 are important progeny in the ^{232}Th and ^{230}Th decay chains. There are major uncertainties in the dose conversion factors associated with these radionuclides. There is also uncertainty (ERDA

1541, 1976)[2] concerning the extent of translocation of the ^{232}Th daughter, ^{228}Ra, from bone, recirculating via the blood, and the related potential could exist for up to an order-of-magnitude decrease in ^{232}Th exposure limits. On the other hand, Marshall et al. in a 1972 ICRP report[37] proposed a model and parameters leading to an increase in ^{232}Th exposure limits. It is evident that additional data should be collected on ^{232}Th and daughter behavior.

BIBLIOGRAPHIC LITERATURE SURVEY

An annotated bibliography entitled, "Biomedical and Environmental Aspects of the Thorium Fuel Cycle" has been prepared to supplement this report. The purpose of the bibliography is to assist in evaluation of the consequences of high specific activity uranium and related nuclides released to the environment. The document includes abstracts of studies in the following subject areas: Biological; Medical; Radiation Safety and Control; Ecological; Monitoring, Measurement and Analysis; Chemical Analysis; Production; Waste Disposal and Management; and Energy. A majority of the references deal with the bioenvironmental aspects of ^{232}Th-^{228}Th in man and animals. The abstracted references are arranged by subject category; indexes are provided for: (1) authors, (2) title, and (3) keywords. The bibliography may be searched via a computerized information file in the Ecological Sciences Information Center at Oak Ridge National Laboratory and is available for searching upon submission of specific requests.

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