An Assessment of U.S. Domestic Capacity for Producing Reactor- Grade Thorium Dioxide and Controlling Associated Wastes and Effluents

by
W. I. Enderlin

February 1978

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SUMMARY

This report presents an assessment of the U.S. domestic capacity for 1) producing reactor-grade thorium dioxide (ThO_2) and 2) controlling the associated wastes and effluents.

A significant increase in the demand for reactor-grade ThO_2 is likely to develop as a result of the growing interest in the application of the thorium-uranium fuel cycle to nuclear reactors. On the basis of this study, it is concluded that a 28,000 MWe thorium-uranium fuel cycle can be sustained using domestic ThO_2 byproduct, providing that the existing purification capacity is expanded, the market for rare-earth products remains stable, and sufficient quantities of imported monazite continue to be available.

More specifically, an analysis of the information obtained in the course of this investigation gave the following results:

- The wastes and effluents identified with the production of ThO_2 from monazite sand are waste water, tailings, dust, smoke and gas, and radio-nuclides (primarily, ^{232}Th and ^{226}Ra). These wastes and effluents are quantified in Tables 6 and 7 of this report.
- There are currently an estimated 1,500 short tons of crude thorium hydroxide byproduct that can be readily converted to reactor-grade ThO_2 that is impounded with rare-earth waste products at Chattanooga, Tennessee.
- Crude thorium byproduct is currently produced at a rate of about 250 ton/year of equivalent ThO_2 . Of this material, 81% is produced from imported monazite sand, whereas 19% is from domestic monazite sand. The technology for extracting ThO_2 from resources other than monazite sand is not fully developed.
- The present maximum domestic capacity for producing reactor-grade ThO_2 is about 65 to 100 ton/year.

- The current domestic capacity for producing reactor-grade ThO_2 is sufficient to sustain a thorium-uranium fuel cycle of up to 11,000 MWe without recycling thorium, depending on the mix of reactor types selected. This range can be increased to 28,000 MWe by expanding ThO_2 purification capacity to match the current production rate of crude thorium byproduct.
- Potential constraints identified which may impact the expansion of domestic ThO_2 production are: 1) uncertainty in the marketplace, 2) limited available thorium for production of reactor-grade ThO_2 , 3) limited production capacity, and 4) mounting public concern over current levels of radioactivity detected at various points in the production process of thorium and uranium products.

If the demand for domestic ThO_2 exceeds 250 ton/year, expanded research into the following areas is required:

- the potential cost benefit of expanding ThO_2 production capacity and of producing ThO_2 from domestic ores other than monazite,
- the extent of domestic thorium reserves,
- the extraction of thorium from complex ores,
- the environmental impact of increasing domestic ThO_2 production rate from monazite sand and other ores, and
- the evaluation of potential constraints on ThO_2 production and the development of strategies to minimize their impact.

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INTRODUCTION

There is presently little demand for reactor-grade thorium dioxide (ThO_2) in the U.S. The current domestic production rate is about 10 ton/year. However, because of the current resurgence of national interest in the application of the thorium-uranium fuel cycle to nuclear reactors, it is quite possible that a significant increase in demand for this material may develop in the near future. This renewed interest has brought with it a mounting concern over the environmental consequences of large-scale production within the U.S. of reactor-grade ThO_2 from domestic resources.

There is little data or information in the current open literature pertaining to: 1) the extent of domestic thorium resources, 2) the quantity of thorium readily available for conversion to reactor-grade ThO_2 , 3) the current domestic capacity for producing this material, 4) the nature and quantity of wastes and effluents generated as a result of producing the material, 5) the quantity of ThO_2 required to sustain a thorium-uranium fuel cycle, and 6) the constraints which may be encountered with regard to increasing the domestic ThO_2 production rate. This information is necessary to provide a basis for complete evaluation of the thorium-uranium fuel cycle.

PURPOSE

The purpose of this report is to provide an overview of the current capability for large-scale production of reactor-grade ThO_2 in the U.S. from domestic resources, to set forth an initial data base for further assessment of the anticipated environmental impact resulting from increased production from monazite sand, and to identify further research needs.

SCOPE

The scope of this report is limited to the existing capability for producing reactor-grade ThO_2 from domestic thorium resources, using existing facilities and methods for controlling and treating wastes and effluents.

OBJECTIVES

The specific objectives of this study were:

- to identify readily available domestic thorium resources,
- to describe the mining, beneficiation, extraction, and purification processes currently employed in the U.S. for producing reactor-grade ThO_2 from monazite sand,
- to identify the types and magnitudes of wastes and effluents generated as a result of these processes and the control and treatment methods employed,
- to ascertain the current domestic production level of ThO_2 , the existing capacity for increased production, and the associated power production potential, using the thorium-uranium fuel cycle.
- to compare the production processes, the generation of wastes and effluents, and the associated health hazard for producing ThO_2 from monazite sand with those for producing U_3O_8 ,
- to identify the constraints which may be encountered in expanding ThO_2 production.
- to identify areas requiring further investigation.

METHODOLOGY

The methodology used in conducting this study was as follows:

- A comprehensive search of the open literature was conducted.
- The author toured heavy-mineral placer mining and beneficiation operations in Florida and Georgia and visited a rare-earth processing facility in Tennessee.
- Representatives of state environmental monitoring and enforcement agencies, the EPA, and private industry were interviewed.

- Plant production and environmental monitoring data were obtained from the files of state agencies and from private industry.
- Data and information gathered in the course of this study were analyzed to attain the previously stated objectives.

BACKGROUND

The resurgence of interest in the thorium fuel cycle is attributed primarily to the ever increasing price of uranium, the potential for using this valuable resource more effectively with the thorium cycle, and a desire to use non-proliferation fuel cycles. In his paper, "Thorium Utilization Re-examined," Stewart⁽¹⁾ concluded the following:

- U^{233} bred from thorium is generally a better fuel in thermal-spectrum reactors than plutonium bred from U^{238} .
- The recent large escalations in U_3O_8 prices provide a very significant new economic incentive for thorium utilization in Resource-Efficient Converter Reactors (RECR's).
- A breakthrough in laser enrichment would probably provide still more economic incentive for the use of the thorium cycle.
- The combination of Laser-Isotope Separation (LIS) technology and thorium utilization in RECR's could postpone any critical need for breeders for many years.

Thorium dioxide is currently considered the best ceramic compound of thorium for use as nuclear fuel.

Although the market for reactor-grade ThO_2 (99.8% pure) is currently suppressed (due to cancellation of all high temperature gas reactor (HTGR) orders in this country), the potential does exist for a substantial increase in the demand for this fuel in the U.S. in the foreseeable future. The Department of Energy has continued studies of the HTGR steam cycle.⁽²⁾ Moreover, research has also continued on the development of a thorium-fueled, light-water breeder reactor (LWBR) and a gas-cooled, fast-breeder reactor (GCFR) which breed U^{233} from Th^{232} . Also under consideration by the Federal government is a modified Canadian-deuterium-uranium (CANDU) reactor which would consume thorium.⁽³⁾ Large-scale development of any of these reactor systems could substantially increase the demand for thorium.

Should the demand for reactor-grade ThO_2 increase significantly, it would become necessary to develop a domestic capability for large-scale production of this material, or to rely on imports of either reactor-grade ThO_2 , crude thorium nitrate or thorium ore.

History shows foreign imports to be an unreliable source of ThO_2 , because national controls and agreements have become a part of the international traffic in monazite. Monazite is a primary source of rare-earth materials and is currently the only commercial source of ThO_2 . The U.S., Brazil, India, and the Republic of South Africa have been the chief sources of monazite, but small amounts have come from many other countries. In the past many of these countries have restricted exports of monazite. In 1944, Australia forbade shipments of monazite to markets outside the United Kingdom and the U.S. The Republic of South Africa had similar regulations. India restricted exports in 1946. At the beginning of 1951 Brazil ceased to issue export licenses, but was, like India, considering offers of foreign firms to process monazite within the country. Rare earths extracted from monazite were to be exported and the thorium retained in the country of origin.⁽⁴⁾ Also in the past, the price of monazite was affected by political controls. Only India and Brazil still maintain embargoes on the export of monazite because of its thorium content.

The domestic potential for the large-scale production of reactor-grade ThO_2 and the nature and magnitude of the resulting effluents and wastes are not well understood.

There are considerable thorium-bearing mineral resources in the U.S.; however, due to the lack of market of thorium, these resources have not been developed. Moreover, commercial extraction processes for thorium minerals other than monazite have not been fully developed.

Domestic production of ThO_2 is currently tied to both the market for titanium and the market for rare earths. A small amount (19%) of the monazite processed in the U.S. comes from domestic heavy-mineral placer mining operations. The balance (81%) of the monazite processed is imported from

Malaysia and Thailand.⁽²⁾ Currently, successful commercial exploitation of domestic monazite hinges on beneficiation of heavy multimineral concentrates, which include monazite, ilmenite, rutile and other heavy minerals of commercial value. Ilmenite and rutile are primary sources of titanium. The monazite sand fraction is separated and marketed for its rare-earth content. As demand requires, ThO_2 is extracted from empounded rare-earth residue and purified to reactor grade.

The Federal government currently maintains a surplus of monazite sand to build stocks of rare-earth materials.

Because of the extremely small production of ThO_2 in the past, there has been little effort in the U.S. to identify and characterize wastes and effluents generated as a result of producing reactor-grade ThO_2 . For the same reason, there has been little effort to assess the potential impact of these wastes and effluents on the surrounding environment and the necessary action required to minimize that impact, should large-scale production become necessary. Albert⁽⁵⁾ and others have assessed the industrial hygiene aspects of producing thorium and its compounds from monazite sand. However, these works are primarily concerned with refining operations. They do not cover mining and beneficiation operations or impact on the quality of the environment external to the processing plants. The existing data base for assessing potential impact of producing ThO_2 on the quality of the surrounding environment is limited and to date has not been compiled into a single source document. State (Florida, Georgia, and Tennessee) environmental health agencies, in conjunction with mine and mill operators, monitor the quality of surface water discharged to the environment from heavy-mineral mining and rare-earth processing operations. The state agencies have on occasion also measured the level of radioactivity at various points in the mining, beneficiation, extraction and purification processes. The raw data is contained in the files of the various state agencies. Currently the Federal government does not monitor these operations but defers this function to the applicable state agencies.

CONCLUSIONS AND RECOMMENDATIONS

CONCLUSIONS

Based on the information set forth in this report, the following is concluded:

- Application of the thorium-uranium fuel cycle in the U.S. in excess of 1,000 MWe to 11,000 MWe, depending on the mix of reactor types selected, would create a demand for reactor-grade ThO_2 that exceeds current domestic production capacity.
- A modest thorium-uranium fuel cycle could be sustained by making minor adjustments to current ThO_2 production facilities.
- Adequate foreign sources of ThO_2 or ThO_2 ore may not always be available to the U.S.
- There may be considerable domestic thorium resources; however, the supply of thorium readily available as a byproduct for consumption by the power industry is limited to 250 ton/year ThO_2 . The full extent of the domestic resource is still unknown.
- Thorite (ThSiO_4) has the highest thorium content (25.2% to 62.7%) of any of the currently identified domestic thorium-bearing minerals and is the primary mineral other than monazite that is likely to be exploited in the event of an increase in the demand for domestic thorium.
- The technology for extracting thorium from ores other than monazite is not fully developed.
- Because of the present uncertainty of a future market for thorium, there is currently no incentive for industry to develop known domestic thorium resources, to expand ThO_2 production capacity, or to explore for additional resources.
- There is currently insufficient data available for assessing the environmental impact of large-scale reactor-grade ThO_2 production.

- If the demand for ThO_2 exceeds 250 ton/year, research is required in the following areas: the extraction of thorium from complex ores; environmental impact assessment of increasing domestic ThO_2 production; potential cost benefit of producing reactor-grade ThO_2 from domestic resources; and reassessment of domestic thorium reserves.

RECOMMENDATIONS

The following is recommended:

- Determine potential demand for reactor-grade ThO_2 .
- If it is anticipated that the demand for domestic ThO_2 will exceed 250 ton/year, research should be conducted in the areas identified above.
- Determine the impact of the constraints identified in this report on the development of the thorium-uranium fuel cycle and, if necessary, develop strategies for mitigating the impact.

DOMESTIC THORIUM RESOURCES

The domestic sources of thorium are primarily thorium-bearing minerals shown in Table 1,^(6,7) impounded residue from rare-earth processing, and monazite sand stockpiled in the U.S. Identified domestic thorium resources recoverable primarily as a byproduct or a co-product have been estimated at 46×10^3 short tons ThO_2 , of which 16×10^3 short tons are in Atlantic coast beach placer deposits.⁽⁹⁾ Domestic resources recoverable primarily for ThO_2 of grade less than 0.1% have been estimated at 106×10^3 short tons; whereas, those of grade greater than 0.1% have been estimated at 142×10^3 short tons.⁽⁹⁾ These data, however, are even less certain than those published for domestic uranium resources. As a result, the USGS is currently upgrading these estimates to a higher level of validity and a new report is expected by mid-1978. Also, the Department of Energy (DOE) established a regional office in Atlanta, Georgia to evaluate the potential of uranium and thorium resources in the Southeast.

TABLE 1. The Major Domestic Thorium-Bearing Minerals^(6,7)

<u>Mineral</u>	<u>Chemical Formula</u>	<u>Thorium Content, %</u>
Allanite	$(\text{Ca}, \text{Ce}, \text{Th})_2(\text{Al}, \text{Fe}, \text{Mg})_3\text{Si}_3\text{O}_{12}(\text{OH})$	Up to 3.2
Monazite	$(\text{Ce}, \text{La}, \text{Y}, \text{Th})\text{PO}_4$	uncertain, perhaps up to 26
Thorite	ThSiO_4	25.2-62.7
Xenotime	YPO_4	up to 2.2

One dimension of the evaluation of the use of thorium for reactor fuel cycles would be the amount of uranium present in thorium-bearing ores. Thorium contained in ores that contain no uranium is 100% ^{232}Th . This is the fertile isotope that produces fissile ^{233}U when placed in a fission reactor. However, ores that contain both Th and U contain some ^{230}Th which

is produced in the ^{238}U radioactive decay chain. When irradiated, ^{230}Th produces ^{232}U which is chemically inseparable from the fissile ^{233}U product. Moreover, this isotope decays rapidly to ^{208}Tl which β decays with a high energy 2.6 MeV γ . The presence of ^{230}Th , therefore, significantly affects the inherent radioactivity of uranium that could be recovered from irradiated thorium fuels and recycled as fissile fuel. This dimension of the evaluation of thorium fuels is not a part of this study.

MONAZITE

Currently, thorium-bearing monazite sand is the only commercial source of ThO_2 . Monazite is an anhydrous orthophosphate of the cerium earths which may or may not contain ThO_2 in significant quantity. A typical chemical analysis of domestic thorium-bearing monazite is shown in Table 2.⁽⁶⁾ Monazite sand has the following physical characteristics:⁽⁴⁾

- grain size less than 0.02-in. diameter,
- heavy (specific gravity 5.0 to 5.2),
- paramagnetic,
- nonconductive (electrical),
- radioactive,
- hard (5-5.5 on Mohs' scale), and
- brittle.

Monazite generally forms small grains which occur as minor accessory minerals in plutonic rocks. Subsequently natural mechanical concentration of this mineral occurs, forming placer deposits. There are numerous heavy-mineral placer deposits containing monazite within the U.S.^(4,8) Monazite has been mined from fluvial placers in North Carolina, South Carolina and Idaho, and from beach placers in Florida and Georgia. Currently, the only domestic production of monazite is as a byproduct from two heavy-mineral placer mining operations located at Green Cove Springs, Florida; and between Boulogne, Florida and Folkston, Georgia. The heavy-mineral content of these

ancient beach placers is about 3%. The monazite content of the heavy-mineral concentrate is about 1%, or about 0.03% of the placer.^(a)

TABLE 2. Analyses of Domestic Thorium-Bearing Monazite Sand⁽⁶⁾

<u>Mineral</u>	<u>Typical %</u>	<u>Range %</u>
ThO ₂	3.4	1.2-7.0
Ce ₂ O ₃	19.5	19.5-37.3
P ₂ O ₅	20.5	18.4-29.3
SiO ₂	8.5	0.3-8.5
ZrO ₂	0.1-1.0	0-3.2
TiO ₂	2.1	0-4.7
Fe ₂ O ₃	4.5	0-7.8
Al ₂ O ₃	12.5	0-12.5
RE ₂ O ₃	40.1	0-40.1
U ₃ O ₈	0.15	0-0.15

OTHER MINERALS

There are twenty major thorium-bearing minerals other than monazite distributed throughout the world. Three of these mineral types (see Table 1) occur in identified vein deposits at 13 widely scattered areas in the U.S.,⁽⁷⁾ however, to date these resources have not been developed. There have also been recent domestic discoveries of thorium as a result of exploration for other commodities. Thorian uranite and thorogummite were reported in an abandoned serpentine-talc quarry located in Northampton County, Pennsylvania. Buttes Gas & Oil Company began a study of a large deposit of titanium in southwestern Colorado believed to contain a significant amount of thorium.⁽³⁾ Small quantities of thorite have been produced domestically, but the commercial importance of this ore has been limited in comparison with that of monazite.^(6,9)

(a) Information obtained from mine operators.

RARE-EARTH BYPRODUCT

In addition to domestic naturally occurring thorium-bearing minerals, there is also considerable thorium impounded in the form of crude thorium hydroxide at the only rare-earth processing facility in the U.S., which is at Chattanooga, Tennessee. The rare-earth processing plant has not maintained an inventory of ThO_2 discharged to the impoundment; however, management at this plant estimates the ThO_2 content at about 3×10^6 lb (1.5×10^3 short tons). ThO_2 is currently discharged to the holding pond at a rate of about 250 ton/year. Domestic monazite accounts for about 19% of the plant throughput, whereas the remaining 81% is from imported monazite. The Federal government maintains a surplus of monazite sand to build stocks of rare-earth materials.

HEALTH HAZARDS

The health hazards associated with the processing of thorium-bearing compounds are discussed extensively in the open literature.^(5,6) The following is intended to highlight these discussions as they pertain to the production of reactor-grade ThO₂ from thorium-bearing minerals.

Radiological toxicity is the controlling hazard in processing thorium, as the chemical toxicity has been found to be low. Because of slow clearance of thorium from the human body, it is a potentially hazardous material. Elements of major concern and their nonoccupational maximum permissible concentrations are shown in Table 3. The radioactive decay of thorium is shown in Figures 1 and 2.

TABLE 3. Nonoccupational Maximum Permissible Concentrations for Various Thorium Isotopes and Members of the Th²³² Decay Chain⁽⁵⁾

<u>Isotope</u>	<u>MPC in air, μc/ml</u>	<u>MPC in water, μc/ml</u>
Natural thorium		
Insoluble	10 ⁻¹²	10 ⁻⁵
Soluble	10 ⁻¹²	10 ⁻⁶
Th ²³²		
Insoluble	10 ⁻¹²	4 x 10 ⁻⁵
Soluble	10 ⁻¹²	2 x 10 ⁻⁶
Th ²²⁸		
Insoluble	2 x 10 ⁻¹³	10 ⁻⁵
Soluble	3 x 10 ⁻¹³	7 x 10 ⁻⁶
Th ²³⁰		
Insoluble	3 x 10 ⁻¹³	3 x 10 ⁻⁵
Soluble	8 x 10 ⁻¹⁴	2 x 10 ⁻⁶
Ra ²²⁸		
Insoluble	2 x 10 ⁻¹²	3 x 10 ⁻⁸
Soluble	2 x 10 ⁻¹²	3 x 10 ⁻⁵
Ra ²²⁴		
Insoluble	2 x 10 ⁻¹¹	5 x 10 ⁻⁶
Soluble	2 x 10 ⁻¹⁰	2 x 10 ⁻⁶

FIGURE 1. Thorium Decay Series⁽⁶⁾

FIGURE 2. Radioactive Decay of Th^{232} Plus $\text{Th}^{228(6)}$

The thorium decay series differs considerably from the uranium decay series in that half-lives are all relatively short. Considering all factors (specific activities, total energies, and percent of equilibrium attained), it has been concluded⁽⁶⁾ that thorium deposited in the body is probably a greater radiological hazard than an equal quantity of uranium. A comparison of the decay series for thorium and uranium is shown in Table 4.

The radiological hazards in processing thorium vary with the degree of separation, the concentration of the toxic members of the chain, and the time interval between the processing steps. The nonthorium fraction poses the more serious handling, storage, and disposal problems.

Because of the short half-lives, any separation into thorium and nonthorium fractions results in a rapid change in the composition of the fractions soon after separation. The nonthorium fraction contains Ra^{228} , which has the strongly active Th^{228} as one of its decay products. The content of Th^{228} can be controlled by separating thorium from nonthorium at proper intervals. The Ra^{224} in the nonthorium fraction decays to Ra^{220} , a short-lived radioactive gas that may build up high levels of radioactivity in confined areas.

After separation of the thorium and nonthorium fractions, the thorium fraction has only slight alpha (α) activity, no beta (β) radiation and only slight gamma (γ) radiation. However, activity of the Th^{228} side of the chain is quickly re-established. The first equilibrium state is reached in about 36 days, as shown in Figure 2.

The control of fugitive dust is also a major item of importance. Most of the exposure to thorium of people in close proximity to thorium processing operations comes from inhaling dusts. It is believed that the lung retains a large part of the inhaled material and, while no chemical toxicity results, the lung tissue is irradiated by the material deposited there. Some of the inhaled material eventually becomes fixed in the bone, which may result in radiation damage to blood-forming tissue.⁽⁶⁾

TABLE 4. Decay Series of Thorium and Uranium⁽¹⁰⁾

Element of Name	Symbol(s)	Half-Life	Energy of Radiation (MeV)		
			α	β	γ
THORIUM SERIES					
Thorium	$^{90}\text{Th}^{232}$	1.34×10^{10} years	4.20	--	--
Mesothorium 1	$^{88}\text{Ra}^{228}$ (MsTh ₁)	6.7 years	--	0.053	--
Mesothorium 2	$^{89}\text{Ac}^{228}$ (MsTh ₂)	6.13 hours	4.5	1.55	--
Radiothorium	$^{90}\text{Th}^{228}$ (RdTh)	1.90 years	5.42	--	γ
Thorium X	$^{88}\text{Ra}^{224}$ (ThX)	3.64 days	5.68	--	--
Thoron	$^{86}\text{Rn}^{220}$ (Tn)	54.5 seconds	6.28	--	--
Thorium A	$^{84}\text{Po}^{216}$ (ThA)	0.158 seconds	6.77	β	--
Thorium B	$^{82}\text{Pb}^{212}$ (ThB)	10.6 hours	--	0.36	--
Thorium C	$^{83}\text{Bi}^{212}$ (ThC)	60.5 minutes	6.05	2.20	γ
Thorium C'	$^{84}\text{Po}^{212}$ (ThC')	3×10^{-7} second	8.77	--	--
Thorium C''	$^{81}\text{Ti}^{208}$ (ThC'')	3.1 minutes	--	1.82	2.62
Thorium D	$^{82}\text{Pb}^{208}$ (ThD)	Stable	--	--	--
URANIUM SERIES					
Uranium	$^{92}\text{U}^{238}$ (U1)	4.55×10^9 years	4.21	--	--
Thorium	$^{90}\text{Th}^{234}$ (UX ₁)	24.1 days	--	0.13	0.09
Protactinium	$^{91}\text{Pa}^{234}$ (UX ₂)	1.14 minutes	--	2.32	0.80
Uranium	$^{92}\text{U}^{234}$ (U11)	2.69×10^5 years	4.75	--	--
Thorium	$^{90}\text{Th}^{230}$ (Io)	8.22×10^4 years	4.66	--	γ
Radium	$^{88}\text{Ra}^{226}$	1600 years	4.79	--	0.19
Radon	$^{86}\text{Rn}^{222}$	3.825 days	5.49	--	--
Polonium	$^{84}\text{Po}^{218}$ (RaA)	3.05 minutes	5.99	β	--
Lead	$^{82}\text{Pb}^{214}$ (RaB)	26.8 minutes	--	0.65	γ
Bismuth	$^{83}\text{Bi}^{214}$ (RaC)	19.7 minutes	5.50	3.15	1.8
Polonium	$^{84}\text{Po}^{214}$ (RaC')	1.5×10^{-4} second	7.68	--	--
Thallium	$^{81}\text{Ti}^{210}$ (RaC'')	1.32 minutes	--	1.80	--
Lead	$^{82}\text{Pb}^{210}$ (RaD)	22.2 years	--	0.025	0.047
Bismuth	$^{83}\text{Bi}^{210}$ (RaE)	4.97 days	--	1.17	--
Polonium	$^{84}\text{Po}^{210}$ (RaF)	139 days	5.30	--	γ
Lead	$^{82}\text{Pb}^{206}$ (RaC)	Stable	--	--	--

In addition to the radiological health hazards of producing ThO_2 , dusts generated during the processing of thorium-bearing ores may also contain a high free silica content, which is also considered a health hazard.

PROCESS DESCRIPTION

Three major processes are currently employed to produce reactor-grade ThO_2 from domestic thorium-bearing monazite sand. Those processes--mining and beneficiation, extraction, and purification--are discussed in the following sections. Major process steps are described, associated wastes and effluents are identified, and the production process is compared with the production of U_3O_8 (yellow cake).

MINING AND BENEFICIATION OF MONAZITE SAND

The monazite deposits currently being mined are ancient beach placers which contain 3% heavy minerals, of which 1% is monazite sand containing about 4% ThO_2 . In some cases the sand grains are cemented with a tar-like organic substance (humate) forming a hardpan resembling soft sandstone. These deposits are normally about 19 ft thick, flat laying, in areas of shallow water table, and are covered with from 10 in. to 3 ft of topsoil. The terrain is flat with a vegetative cover of pine timber and undergrowth. The surrounding area is sparsely populated. The two currently active mining operations are about 35 mi from Jacksonville, Florida, the nearest major population center.

The following mining and beneficiation technique, developed in 1944, has enabled domestic beach placer mining operations to compete with low-cost, post-World-War-II foreign imports. The primary steps in this operation are land clearing and reclamation, dredging, concentrating, product separation, and effluent treatment. This operation is conducted over a 22-hour day, 7 days/week. The process flow is shown in Figure 3.

Land Clearing and Reclamation

Prior to mining, the pine timber is harvested for paper manufacturing, the undergrowth is removed and burned, and the topsoil is removed and stockpiled. As an area becomes mined out, the spoils (barren sand) are contoured as required, covered with topsoil, planted with grass, and fertilized. Some small lakes are left.

FIGURE 3. Simplified Schematic Diagrams of Mining and Beneficiation of Heavy Minerals

Dredging

The placer deposits are mined using an electric-powered, floating suction dredge with a rotating cutter head (Figures 4 and 5). The heart of the hydraulic dredge is the main centrifugal pump with its suction and discharge lines. The suction line is carried in a live boom called the ladder. Its position and the working movements of the hull are controlled by winches and spuds on the hull and by anchors on the bottom or on shore. The dredge does its hard digging while swinging toward starboard, with cutter blades slicing up into the bank. The swing back to port is normally made with the cutter at the same level, cleaning up loosened material. The material loosened by the cutter head is drawn into the suction line and passes through the pump and is discharged to the wet mill (concentrator) via a floating discharge line. A more detailed description of the configuration and the operating characteristics of hydraulic dredges is presented by Nichols.⁽¹¹⁾

The dredge at the larger of the two mines at Green Cove Springs, has a 22-in. diameter discharge and moves about 600 ton/hour, which is about 50% of design capacity. Dredge output is limited by the capacity of the wet mill. At this digging rate, about 15 acres per month are mined. The connected electrical load is about 800 kW @ 2300 V. The personnel requirements are two operators and two maintenance personnel per shift.

The dredge at the smaller operation, at Boulogne, Florida, has a 16-in. diameter discharge and moves about 550 tons/hour.

The cut-off grade for dredge operation is 2% or less heavy mineral content in the sand.

Ore Concentration

The ore is concentrated using wet gravity separation at the wet mill (Figure 6) which may be floated behind the dredge or located on land next to the dredge pond. The wet mill includes a surge bin, hammer mill, classifier screens, and two parallel separation circuits. These circuits consist of a series of sluices (Figure 7) and/or Humphrey spirals (Figure 8), pumps, overboard tailing discharge lines (Figure 9), and concentrate discharge line.

FIGURE 4. Suction Dredge

FIGURE 5. Rotating Cutterhead on a Suction Dredge

FIGURE 6. Floating Wet Mill

FIGURE 7. Sluice Circuits on Wet Mill

FIGURE 8. Humphrey Spirals on Wet Mill

FIGURE 9. Tailings Discharge Lines on Wet Mill

All output from the dredge is input to the wet mill. The wet mill produces a 75% heavy-mineral concentrate at a rate of 18 to 20 tons/hour, which is pumped or trucked to the dry mill for product separation, depending on the mine. If necessary, the concentrate is washed with caustic solution to remove about 80% of the humate from the sand grains prior to discharging the concentrate from the wet mill. This wash solution, containing the humate together with all other material (about 97% of the dredge output) is discharged to the dredge pond.

The concentrate is dewatered either at the wet mill or at the dry mill, depending on how it is to be transported. The overflow water is returned to the dredge pond. The wet concentrate containing 20% moisture is stock-piled and allowed to dry to a maximum moisture content of about 8%.

Personnel requirements for the wet mill are five operators and five maintenance personnel per shift.

Product Separation

At the dry mill the dewatered concentrate is heated to 220°F in a dryer fired with #6 fuel oil to remove the remaining moisture. The dry mill uses a combination of electrostatic, magnetic, and wet gravity techniques to separate the various heavy-mineral products (Zircon, ilmenite, rutile, leucoxene, and monazite) from the gangue minerals and from each other. The distribution of the dry mill throughput is shown in Table 5.

The recovery efficiency of monazite is low at one of the mines because of the low efficiency of the equipment used in the final magnetic separation step. Therefore, about 60% of the monazite is discharged to tails. The mine is currently replacing this equipment and hopes to achieve a recovery efficiency of 80 to 90% in the near future. The other mine currently recovers 90% of the monazite fraction.

The thorium-bearing product at each mine is a 95% monazite sand. At one mine the monazite product is shipped in 55-gal drums with 1200 lb of sand per drum. The drums are marked "RADIOACTIVE--LOW SPECIFIC ACTIVITY." At the other mine all the product is packaged in heavy paper bags with 50 lb/bag.

TABLE 5. Distribution of Dry Mill Throughput^(a)

	<u>% of Mill Feed</u>	<u>% Recovery</u>
Input 75% HM Concentrate	100.0	--
95% Monazite Product Shipped	0.3-0.7	30-90
Other HM Product Shipped	66.8	90
Discharge to Dry Mill Tailings Pile	32.5-32.9	--

(a) Based on data furnished by mine operators.

Both mines ship the monazite product to the rare-earth plant at Chattanooga by truck at about 6- to 8-week intervals. The mine with the higher monazite recovery rate ships 600 to 700 ton/year of monazite sand product, whereas the other mine ships 300 to 400 ton/year.

The personnel requirements for each dry mill are four operators and four maintenance personnel per shift.

Effluents and Wastes Generated

The major effluents and wastes generated by a typical heavy-mineral placer mining operation are shown in Table 6. The primary contaminants and current control methods are also identified. From this data it is evident that the dry mill tailings, which contain a significant amount of monazite and the 95% monazite product, have relatively high radioactivity. No data was available on the level of radionuclides in the mine discharge water. There was also no data available pertaining to air quality adjacent to the mine property.

TABLE 6. Effluents and Wastes Generated by a Typical Heavy-Mineral Placer Mining Operation(a)

Effluent/Waste	Source	Rate of Discharge	Type of Contaminant	Concentration of Contaminant	Current Control Method	Discharge Point
Mine Water	Dredge Pond and Concentrate Dewatering	700-3000 gpm	Suspended Mineral and Organic Fines	$18 \times 10^3 - 20 \times 10^3$ mg/s	Add Alum to Flaculate and Settle Fines in Holding Ponds	Flow to River and Evaporation to Atmosphere
			Organic Acids Na^+	$\text{pH} \leq 5$ U(b)	Add Caustic to Adjust pH None	
Mill Tailings	Wet Mill	511 ton/hr	Fines	U	Reclaim Land	Dredge Pond
	Dry Mill	6 ton/hr	Organic Debris Monazite	U $0.3\% - 2.1\%$	Recycle Tailings to Remove Monazite (in the future)	Dry Mill Tailings Pile
Radionuclides	Ore		Th-232 Ra-226	0.96 ± 0.02 pc/g 1.7 ± 0.1 pc/g	None	Atmosphere
	Wet Mill Tailings		Th-232 Ra-226	0.17 ± 0.01 pc/g 0.32 ± 0.05 pc/g		
	Ore Concentrate		Th-232 Ra-226	30 ± 1 pc/g 51 ± 3 pc/g		
	95% Monazite Product		Th-232 Ra-226	6800 ± 100 pc/g 4800 ± 500 pc/g		
	Dry Mill Tailings		Th-232 Ra-226	93 ± 1 pc/g 90 ± 6 pc/g		
Fugitive Dust	Dry Mill	25 lb/day	Free Silica Organic Matter Other Minerals	U	General Ventilation with Dilution Used at One Mill and Induced Draft With Cyclone Used at the Other Mill	Atmosphere
Gasses and Smoke	Ore	U	H_2S	Noticeable Odor at Dredge Pond	None	Atmosphere
	Dry Mill	U	#6 Fuel Oil Products of Combustion	U	None	Atmosphere
	Burning Brush	15 acre/mo	Smoke	U	None	Atmosphere

(a) Based on data furnished by mine operators and by the State of Florida Department of Health and Rehabilitative Services.

(b) U = Unknown.

Comparison of Mining Monazite Sand with Mining U_3O_8 Ore

There is a vast difference between the mining method required to produce mill-grade monazite sand and mill-grade U_3O_8 ore. This vast difference is dictated by the difference in the nature of the ore body and the ore grade.

The U_3O_8 mineralization occurs as a mineral coating on individual sand grains in a well-cemented (hard) sandstone. The ThO_2 is chemically bound within the individual monazite sand grains which are unconsolidated or lightly cemented. Moreover, the U_3O_8 ore occurs in pods or lenses which may be located close to the surface or at depth; whereas, the monazite sand occurs in low concentration in placer deposits located at or very close to the surface. The content of U_3O_8 in ore ranges from about 0.08% to about 2.5% with an average mill blend of 0.15% to 0.22% grade. Uranium is mined by open pit or underground methods. The content of ThO_2 in the placer sand is about 0.0012%. Monazite sand is mined using hydraulic dredging together with wet gravity concentration and electrostatic and magnetic separation techniques. Thorium is produced as a byproduct and does not bear the cost of the mining operation, whereas the U_3O_8 ore bears the full cost of the mining operation.

Another major difference is that considerably more land is disturbed in producing ThO_2 from monazite sand than in producing an equal quantity of U_3O_8 .

EXTRACTION OF THORIUM

There are currently four distinct processes known for opening monazite: alkaline attack, normally with caustic soda; sulfuric acid attack; chlorination in the presence of carbon; and high temperature reduction with carbon. Of these, only the caustic soda attack is currently in commercial use. There are presently two rare-earth plants in operation which process monazite sand: one plant in France and the other operated by the W. R. Grace Company at Chattanooga, Tennessee (Figure 10).

Process Description

A simplified schematic of the process is shown in Figure 11. The monazite sand is ground to pass a 300 mesh sieve and is reacted with hot caustic soda to give sodium phosphate and a mixed thorium-lanthanum hydroxide. Sodium

FIGURE 10. U.S. Monazite Processing Facility

FIGURE 11. Simplified Schematic of Thorium Extraction Process

phosphate and excess caustic soda are removed from the reaction by washing with water. The mother liquor, after making up to strength with fresh caustic, is recycled. The thorium-lanthanum product is separated into its components by making use of the difference in basicity between thorium and lanthanum. A detailed description of this process is given by Cuthbert⁽⁶⁾ and by Callow.⁽¹²⁾

Over 95% of the original thorium, together with gangue minerals, is impounded in the thorium pond (Figure 10). The ThO_2 content of the plant discharge to this pond averages 250 ton/year. It has been estimated by the plant management that the ThO_2 content in this pond is on the order of 1500 short tons. An additional unknown quantity of ThO_2 is contained in an abandoned pond area that has been dewatered.

According to management, this mill has the capacity for increasing its current throughput by 100% without a need for major capital expense.

Effluents and Wastes Generated

The major effluents and wastes generated at the rare-earth plant and discharged from the plant property are shown in Table 7. All other solid residue is impounded on the plant property in settling ponds, as shown in Figures 10 and 11. At the present production rate, the existing thorium pond capacity is sufficient until 1990, when a new pond will be required.

There is no data available on air quality adjacent to the plant property.

Comparison of Thorium Extraction with U_3O_8 Extraction

The composition of the effluents and wastes generated by thorium extraction is different from the composition of those generated in milling U_3O_8 . Moreover, the total throughput of the rare-earth mill is considerably less than for a typical U_3O_8 mill. Also, since crude thorium hydroxide is a byproduct, it does not bear the cost of milling, whereas U_3O_8 does.

PURIFICATION OF THORIUM DIOXIDE

The only plant in the U.S. that produces reactor-grade ThO_2 from crude thorium sludge is Tennessee Nuclear Specialties, located at Jonesboro, Tennessee.

TABLE 7. Effluents and Wastes Generated by a Typical Rare-Earth Processing Plant^(a)

Effluent/Waste	Source	Rate of Discharge	Type of Contaminant	Concentration of Contaminant	Current Control Method	Discharge Point
Mill Water Discharge	Main Settling Pond	70 gpm Average	Suspended Solids	0.15 g/l Average	Solids settled out and pH adjusted	City Sewer
			Soluble α and β Activity	0.011 $\mu\text{Ci} \times 10^{-4}/\text{ml}$ Average		
			Insoluble α and β Activity	0.002 $\mu\text{Ci} \times 10^{-4}/\text{ml}$ Average		
Phosphate Pond Water	Caustic Recovery Process	U ^(b)	Suspended β	19 pc/l	None	Evaporation to Atmosphere and Seepage to Ground Water and Recycle
			Dissolved β Phosphate	3 pc/l		
Thorium Pond Water	Monazite Dissolution Process	U	Suspended α	2000 pc/l	None	Evaporation to Atmosphere and Seepage to Ground Water and Recycle
			Dissolved α	74 pc/l		
			Suspended β	900 pc/l		
			Dissolved β	39 pc/l		
Fugitive Dust	Entire Plant	U	Radionuclides	0.06 $\times 10^{-11}$ $\mu\text{Ci}/\text{ml}$ at NE Corner of Property and 0.095 $\times 10^{-11}$ $\mu\text{Ci}/\text{ml}$ at NW Corner of Property with NE Wind at 5 mph/hr Sample at 23 cfm on Watman #40 Filter	None	Atmosphere
		U	Suspended Particles	U		
	Ball Mill	U	Radionuclides	1.9 $\times 10^{-11}$ $\mu\text{Ci}/\text{ml}$	Dust Collector	Atmosphere
		U	Suspended Particles	U		
Radioactivity	Sand Storage Building		Gamma (γ)	15 mR/hr	Restricted Area	Atmosphere
	Abandoned Thorium Pond		Gamma (γ)	8-10 mR/hr at 3 ft Above Surface	Restricted Area	Atmosphere
	Active Thorium Pond		Gamma (γ)	0.5 mR/hr	Restricted Area	Atmosphere
	Soil-Settling Pond Dam		Radionuclides	0.4 $\times 10^{-4}$ $\mu\text{Ci}/\text{gm}$	Restricted Area	
	Soil-Field Between Phosphate Pond and Lab		Radionuclides	16 $\times 10^{-4}$ $\mu\text{Ci}/\text{gm}$	None	
Chemical Fumes	Caustic Attack and Dissolution	U	Caustic and Acid Fumes	U	Scrubber	Atmosphere

(a) Data furnished by Tennessee Department of Public Health.

(b) U = Unknown.

Process Description

Three methods of purifying thorium concentrate are possible: solvent extraction, fractional crystallization, and selective precipitation. The solvent-extraction method is currently used in the U.S.

Solvent extraction is performed in three steps:

1. Thorium and uranium are extracted from the aqueous feed solution into the organic-solvent phase; most impurities remain in the aqueous phase. The organic solvent used is tributyl phosphate (TBP).
2. Any contained rare earths and impurities are scrubbed from the thorium-rich organic phase with fresh aqueous solution.
3. Thorium is stripped from the solvent into an aqueous stream.

The thorium-nitrate product thus produced is then precipitated as the oxalate, and calcined to produce 99+% pure ThO_2 . Recovery of thorium is in excess of 95%. A detailed description of this process is given by Cuthbert⁽⁶⁾ and by Callow.⁽¹²⁾

According to plant management, the capacity of the plant is about 700 lb of ThO_2 /day. Currently the plant produces 4 batches/year at 5000 lb/batch (20,000 lb/year). The plant operates the thorium circuit for two 5-day weeks for each batch.

Effluents and Wastes Generated

Lime is added to the effluent, and the solids are removed by filtering and are buried. The water is discharged to a holding pond where it is evaporated. About 3.6×10^6 gal/year are evaporated. No water is discharged from the pond.

No data was obtained on air quality adjacent to the plant.

Comparison of Purification of ThO_2 with U_3O_8 Extraction

Except for the oxalate precipitation and calcination steps, the extraction process is quite similar for both substances.

ANALYSIS OF ThO₂ PRODUCTION CAPACITY
AND POWER PRODUCTION POTENTIAL

The current and maximum ThO₂ production capacities for each of the major processes (mining and beneficiation, extraction, and purification) are shown in Table 8.

TABLE 8. ThO₂ Production Capacity

Process	ThO ₂ Production, short ton/year	
	current	maximum
Domestic mining operations	40	63
Thorium extraction	250	500
ThO ₂ purification	10	65-100

The combined output of the two domestic monazite-producing placer operations is about 40 ton ThO₂/year. This output will increase to about 63 ton/year due to increased monazite recovery which will occur when one of the mines replaces a magnetic separation unit with a more efficient unit in the monazite circuit. The wet mills and dry mills are currently operating at near 100% capacity.

The rare-earth processing plant is currently operating at about 50% capacity to match the current demand for rare earths.

The purification circuit is operated only 8 week/year to match the current demand for ThO₂ (10 ton/year). The output of this plant would increase to about 65 ton/year if the plant were operated 52 week/year on a 5-day/week basis, which is about equal to the maximum output from domestic mining. This output would increase to about 100 ton/year if operation were on a 7-day/week basis, assuming 80% plant availability and additional mill-feed-stock availability.

The estimated quantities of associated products, solid wastes, and waste water resulting per ton of ThO_2 produced from domestic placer deposits are shown in Table 9. The radioactivity levels resulting at various steps in the production process are shown in Tables 6 and 7. Radioactivity data pertaining to the purification process have not as yet been obtained.

TABLE 9. Estimated Average Output Resulting from the Production of One Short Ton of Reactor-Grade ThO_2 from Domestic Placer Deposits

<u>Item</u>	<u>Quantity</u>
ThO_2 Product	1 ton
95% Monazite Sand Product	23.6 ton
Other Heavy-Mineral Product	2596 ton
Rare-Earth Product	11.7 ton
Exposed Mine Spoils	1934 ton
Mill Tailings	10.9 ton
Disturbed Land	0.5 acres
Beach Placer Dredged	101.2×10^3 ton
Waste Water Treated	4.37×10^7 gal
Fugitive Dust	25 lb

The potential for producing power using the thorium-uranium fuel cycle is shown for various reactor concepts in Table 10. It is assumed that the thorium fuel will not be recycled. This table is predicated on the current capacity for producing reactor-grade ThO_2 , as shown in Table 8. Also shown in Table 10 is the estimated increased power production potential resulting from expanding the existing purification capacity (currently the limiting factor) to match the current ThO_2 output of the rare-earth plant. It must be realized that Table 10 is also predicated on a stable market for both heavy minerals and rare-earth products. The increased production potential, as a result of expanding the purification capability, must also rely on the availability of imported monazite to sustain the rare-earth plant at its current production level.

TABLE 10. Power Production Potential

Reactor Fuel Cycle	ThO ₂ ^(b) Reload ST/yr-1000 MWe	Reactor Units Supported ^(e)	
		Existing Purification Capacity ^(c)	Expanded Purification Capacity ^(d)
HTGR (Normal Cycle)	16.6	4-6	16
HTGR-U3/Th			
HTGR ^(a)	8.8	7-11	28
HTGR-U3 (DE)/U/Th-15%			
CANDU ^(a)	55.9	1-2	4
U3 (DE)/U/Th-20%			
CANDU ^(a)	58.1	1-2	4
U5 (DE)/U/Th-20%			
LWR ^(a)	24.2	2-3	10
PNR-U5 (DE)/U/Th			
GCFR ^(a)	22.0	3-4	11

(a) Non-proliferation fuel cycle.

(b) C. M. Heeb, "ThO₂ Requirements for Non-Proliferation Fuel Cycles," PNL, personal communication to W. I. Enderlin, January 4, 1978.

(c) Based on current maximum capacity of 65-100 ton/year ThO₂.

(d) Based on expanding purification capability to match current ThO₂ output of rare-earth processing facility (250 ton/year).

(e) Reactor unit = 1000 MWe-thorium fuel not recycled.

Information presented in Table 10 indicates that the current potential for sustaining the operation of a thorium-uranium fuel cycle with ThO_2 produced as a byproduct in the U.S. ranges from 1000 MWe to 11,000 MWe, depending on the mix of reactor types selected. This range can be increased to 4000 MWe to 28,000 MWe by expanding the ThO_2 purification capability as previously stated.

To extend the power production potential further, it would be necessary to process additional monazite, and to surplus or market the additional rare-earth products produced, or to develop other thorium-bearing resources.

If additional monazite is processed it would be necessary to further expand the purification capacity and to increase monazite imports and/or domestic placer mining activity. It would also be necessary to either market or surplus the additional heavy minerals produced. As previously stated, the existing rare-earth processing plant has the capacity for doubling current throughput.

If other thorium-bearing minerals are to become a source of additional ThO_2 , it would be necessary to develop these resources. Lead time is normally about 10 years. Moreover, extraction processes have not been developed for any of the other thorium-bearing minerals except for thorite. Thorite cannot be processed using the caustic soda process; hence, a new extraction facility using an acid leach-solvent extraction process would be required.^(13,14,15)

Of course, the cost of this additional production capacity would be borne by the nuclear power industry, less any credits for additional heavy minerals and rare-earth products that could be marketed.

CONSTRAINTS

The following constraints, which have a potential for impacting domestic large-scale production of reactor-grade ThO_2 , have been identified in the course of this study:

- There is uncertainty in the marketplace. The development of a market for this product is dependent on the successful development and application of the thorium-uranium fuel cycle. The current demand for ThO_2 in the U.S. is about 10 ton/year. In the past, it has been difficult for domestic thorium resources to compete with low-cost foreign monazite imports, which normally have a thorium content.
- There is limited production capacity. The current capacity for producing this product domestically is limited to 65 to 100 ton/year.
- There is limited supply of available thorium resources. Although there are considerable thorium resources available in the U.S., only 250 ton/year are readily available for conversion to reactor-grade ThO_2 . Of this supply, 81% is from imported ore. Moreover, the availability of this supply is tied to the market for heavy minerals and rare-earth products. It is quite possible that as the demand for thorium increases on the worldwide market, the availability of foreign monazite may decrease, as it has in the past.
- There is mounting public concern over the potential environmental stresses which may result from large-scale mining, extracting, and refining of reactor-grade ThO_2 . This is evidenced by the current indecision regarding public policy and licensing requirements pertaining to mining uranium and thorium ores. Moreover, local governments have expressed concern over current levels of radioactivity detected at various points in the thorium production process.
- There is a lack of incentive for industry to initiate development of domestic thorium resources or to expand ThO_2 production capacity because of the uncertainty of a future market.

These constraints should be investigated to determine the magnitude of their impact on the development of the thorium-uranium fuel cycle. Also, if necessary, strategies should be developed to minimize their impact.

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