
**ENVIRONMENTAL ASSESSMENT OF THE
PROCESSING OF REACTOR RECYCLE
MATERIALS CONTAINING TRANSURANIUM
ELEMENTS NOVEMBER 11, 1975**

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Special

ENVIRONMENTAL ASSESSMENT OF THE PROCESSING
OF REACTOR RECYCLE MATERIALS
CONTAINING TRANSURANIUM ELEMENTS

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United States Energy Research & Development Administration

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Contents

<u>Section</u>		<u>Page</u>
1.0	Introduction.	1
2.0	Proposed Action	1
3.0	Environment Affected.	1
4.0	Anticipated Benefits.	2
5.0	Production Processes.	2
6.0	Waste Treatment	5
7.0	Probable Fate of Neptunium and Plutonium.	7
8.0	Evaluation of Potential Environmental Impact	10
9.0	Conflicts	14
10.0	Alternatives.	14

Figures

<u>No.</u>	<u>Title</u>	<u>Page</u>
1	Area Map.	3
2	Environmental Features.	6

Tables

<u>No.</u>	<u>Title</u>	<u>Page</u>
1	Transuranium Alpha Activity In Paducah Residues	12

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1.0 INTRODUCTION

In a nuclear reactor, the main fission reaction is accompanied by the formation of transuranium elements through neutron capture by thorium or uranium atoms. During reprocessing to recover the unused portion of the fuel, most of the transuranics are removed. However, significant traces of neptunium and plutonium accompany the recovered thorium and uranium returned to production processes of the Energy Research and Development Administration (ERDA).

The ERDA Feed Materials Production Center (FMPC) located at Fernald, Ohio, will process various materials which originated as recovered thorium or uranium. Therefore, FMPC processes may introduce slight amounts of neptunium and plutonium into the environment.

2.0 PROPOSED ACTION

The proposed action considered in this assessment is the processing of reactor recycle materials containing transuranium elements.

Most of the reactor recycle material to be processed will be in the form of uranium nitrate solutions, various uranium compounds and mixtures of compounds, and miscellaneous uranium scrap and residues. The uranium materials will not exceed 10% ^{235}U and most will be under 2% ^{235}U . A small amount of thorium nitrate is also expected.

3.0 ENVIRONMENT AFFECTED

The FMPC site occupies 1,050 acres in Hamilton and Butler counties. Production activities are confined to a 136-acre area in the center of the site. A waste storage area of about 20 acres lies between the production area and the west site boundary.

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Land around the FMPC site is gently rolling to hilly. Farms and wooded lots predominate. Several small rural communities with a combined population of 4,000 are located within three miles of the site boundary: Ross, New Haven, Shandon, New Baltimore, and Fernald. Hamilton (pop. 68,000) is within an 11-mile radius and the northwest edge of Cincinnati (pop. 500,000) is 8.5 miles away. Most of these locations are shown in Figure 1.

Liquid effluent from the FMPC is discharged via a buried pipeline to the Great Miami River. The effluent is a mixture of treated sewage plant effluent, precipitation run-off and ground water drainage, and the supernate or filtrate from treated production wastes. Because of municipal and industrial contaminants added upstream, there is little recreational use of the river and there are no known water supplies drawn from the river. The aquifer in the buried river valley is used as a water supply and part of the aquifer recharge is supplied by the river. About 24 miles below the FMPC outfall, the Great Miami enters the Ohio River.

4.0 ANTICIPATED BENEFITS

Processing of recycle materials will return uranium and thorium to the reactor fuel cycle. Recycle uranium may be slightly enriched and the return of such materials to the gaseous diffusion plants lessens the power needs as compared with the power required to convert normal uranium to the target enrichment.

Processing of these materials also reduces the problem of long-term storage. Metal storage containers, such as 30-gallon or 55-gallon drums, are subject to internal corrosion from the contained material and to external corrosion from moisture. Eventual failure of these containers would require repackaging to avoid the spread of contamination and to conserve the valuable contents.

5.0 PRODUCTION PROCESSES

Uranium processing methods will depend upon the purity and ^{235}U content of the recycle material and the desired end product.

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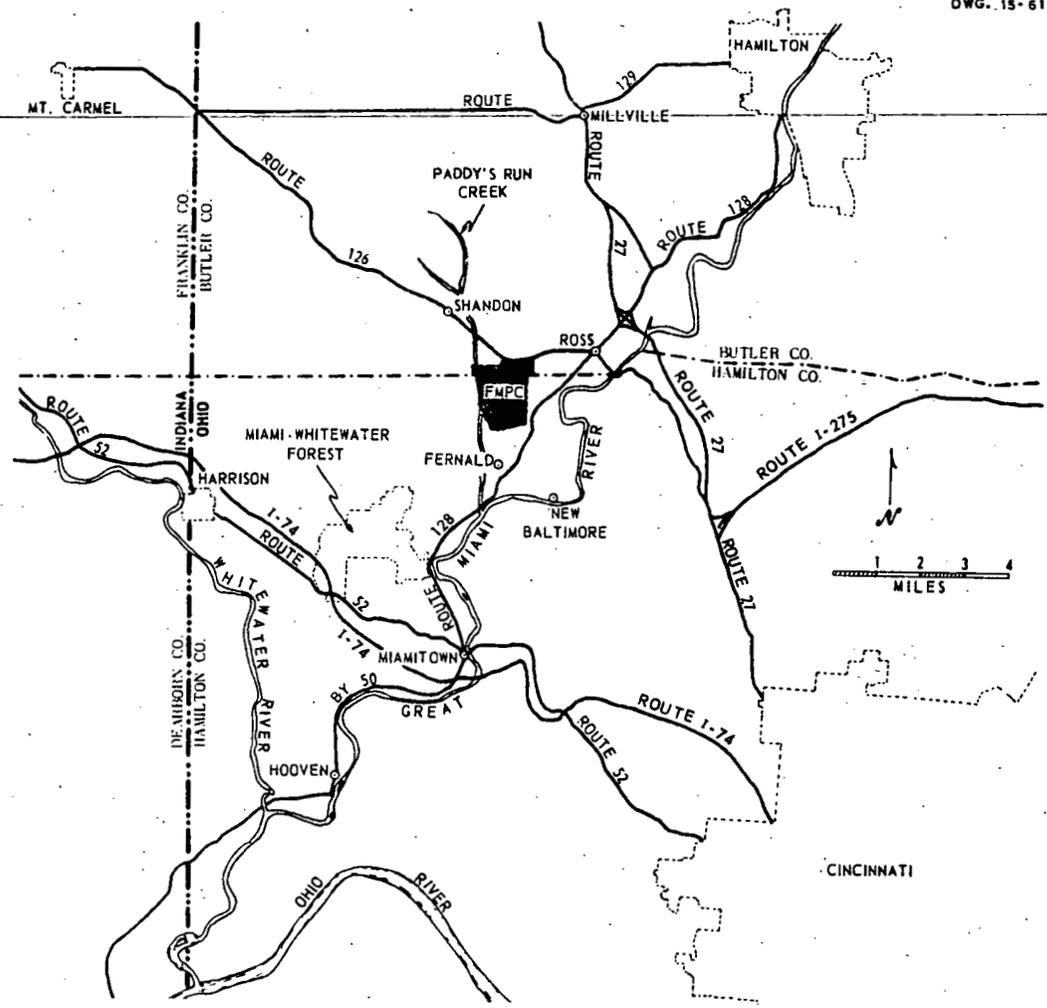


FIGURE 1 Area Map

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Processing which only includes a furnacing or calcining operation will not be considered in detail since there is no separation of impurities. Any transuranium isotopes in the starting material will end up in the product, except for small quantities which become air-borne and are not removed by air-cleaning equipment.

Normal uranium and uranium with a ^{235}U content no greater than 2% will be processed through the Refinery extraction system if purification is required. In this system, feeds are dissolved in nitric acid and the uranium is extracted with tributyl-phosphate in kerosene. Uranium is then re-extracted into deionized water, producing a purified uranyl nitrate solution. The uranium is then thermally decomposed to uranium trioxide (UO_3) powder which may be held for further on-site processing or may be shipped to a gaseous diffusion plant.

Ordinarily, UO_3 with a ^{235}U content significantly greater than normal, will undergo, at the FMPC, further processing to uranium tetrafluoride (UF_4) and uranium metal. In UF_4 production, UO_3 is first reduced to uranium dioxide (UO_2) with hydrogen and then converted to UF_4 by reaction with anhydrous hydrogen fluoride. Uranium metal is produced by reacting UF_4 and magnesium metal in a steel reduction pot lined with magnesium fluoride (MgF_2). This primary uranium metal is remelted, usually with assorted scrap uranium metal, to yield an ingot which is sent off-site to be extruded. Extruded tubes of enriched uranium are sent to an ERDA reactor site for canning and assembly into reactor fuel elements.

Systems are available at the FMPC for some limited processing of enrichments up to 10% ^{235}U . A safe geometry digestion system may be used for selected materials containing up to 10% ^{235}U . A safe geometry digestion system may be used for selected materials containing up to 10% ^{235}U . Uranyl nitrate solutions containing up to 10% ^{235}U may be converted to U_3O_8 in a safe geometry denitration system. Furnacing of material containing up to 10% ^{235}U is also possible.

In addition to normal and enriched processing, depleted uranium metal is also produced at the FMPC. The use of separate facilities and campaign scheduling effectively maintain a separation between the depleted, normal, and enriched materials.

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Recycle thorium will probably be received as a nitrate solution for purification in the Pilot Plant solvent extraction system. In this system, thorium is extracted from a nitrate solution into a diamylamyl phosphonate-solvent mixture. Thorium is re-extracted from the organic phase into a dilute, slightly acidic water solution. The resulting purified solution of thorium nitrate may be shipped or it may be held for conversion to a thorium compound or to metal.

6.0 WASTE TREATMENT

Waste treatment at the FMPC is designed to recover valuable nuclear and non-nuclear materials, to effectively remove or reduce contaminants in water and air, and to immobilize contaminated solid wastes for trouble-free long-term on-site retention.

6.1 Treatment of Liquid Process Wastes

Each of the individual FMPC production plants has sumps and equipment for the collection and primary treatment of process waste water. Uranium and thorium may be recovered as part of the treatment.

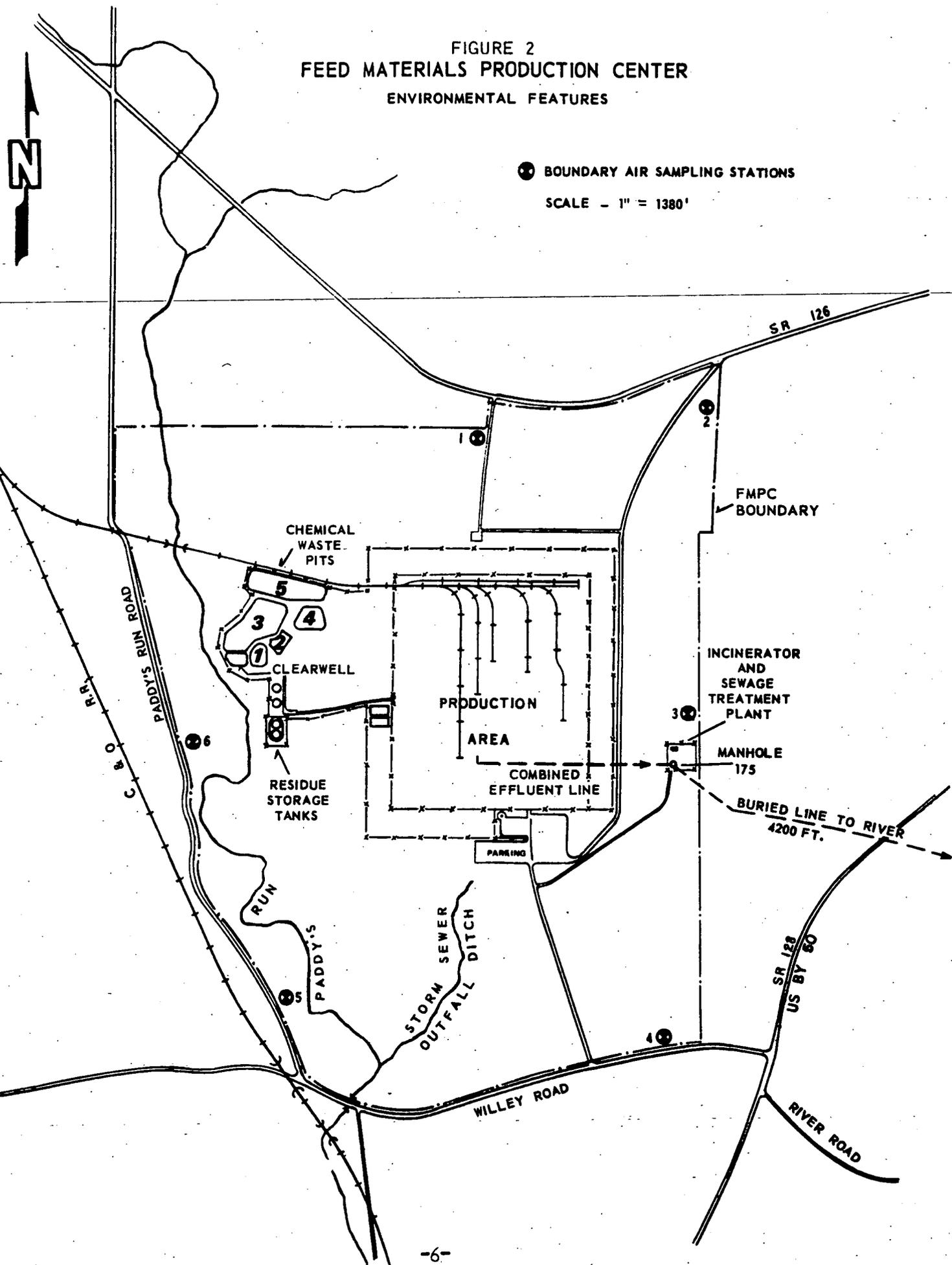
Effluents from the individual plants are collected at a central facility, called the General Sump, for additional treatment. After treatment at the General Sump, the waste is either filtered or pumped to a large settling basin, Pit 5. No effort is made to segregate wastes from depleted, normal, or enriched operations after the wastes have been approved for treatment and discard. In the waste pit, the solids settle and the clear supernatant liquid is pumped to a clearwell. If the General Sump treated waste is filtered, the collected solids are transferred to an on-site storage pit (Pit 3 or Pit 4). Both the filtrate and clearwell water are eventually combined with other liquid streams such as sewage plant effluent and storm sewer water. This combined stream flows by gravity in a buried pipeline to the Great Miami River. See Figure 2.

The combined waste stream is sampled continuously at the final on-site access point, Manhole-175. Daily 24-hour samples are analyzed for various radioactive and non-radioactive contaminants. Results are reported to ERDA, the U. S. EPA, the State of Ohio EPA, and the Miami Conservancy District. Results are summarized in an annual

FIGURE 2 FEED MATERIALS PRODUCTION CENTER ENVIRONMENTAL FEATURES

● BOUNDARY AIR SAMPLING STATIONS

SCALE - 1" = 1380'



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Environmental Monitoring Report which is distributed to all of these agencies and several other State, regional, and local regulatory groups.

6.2 Collection of Airborne Materials

Conversion of impure uranium and thorium compounds to reactor feed material involves operations which generate radioactive dusts, nuisance dusts, and corrosive mists or reaction products. ~~Ventilated enclosures are used to confine these materials and prevent employee exposures.~~ Efficient air cleaning systems are used to remove the contaminants, including valuable material which is returned to the production stream. Sampling of dust collector stacks is maintained on a continuous schedule to determine the operating condition of these air cleaning systems.

6.3 Disposal of Contaminated Solid Wastes

Contaminated solid wastes resulting from production activities are placed in an on-site storage pit (Pit 4) originally built to receive dry materials. Some water is present in the pit because of direct rainfall, drainage from the banks and adjacent concrete pad, and the occasional disposal of a chemical slurry. The water is kept well above pH 7 by the addition of lime. Periodically, this water is pumped to Pit 5 from where it flows to the clearwell and eventual discharge to the river.

Materials sent to Pit 4 are those wastes with a uranium or thorium content too low to warrant recovery. If recovery is warranted, the solids undergo treatment which may include incineration, furnacing or acid leaching. Pit 4 also receives a considerable amount of depleted uranium scrap and residues.

7.0 PROBABLE FATE OF NEPTUNIUM AND PLUTONIUM

In the FMPC production processes, separation of impurities from uranium may occur chemically or physically. Chemical separation occurs chiefly in the Refinery solvent extraction system, while physical separation may occur in the metal forming steps of reduction and remelt.

Because of the great difference in the total amounts handled, thorium processing at the FMPC will introduce to the on-site and off-site environment a minor amount of

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transuranic isotopes compared with that introduced by uranium processing. Therefore, only uranium processing is considered in this section.

Radioactivity from americium and higher elements will not be considered in this assessment. Although these transplutonium elements may be present in recycle uranium, the amount is normally insignificant compared with the radioactivity levels of neptunium and plutonium. In actual practice, the transuranic alpha activity is usually obtained by taking the sum of the alpha activity due to neptunium and plutonium only.

7.1. Extraction

Neptunium and plutonium have several stable valence states and the degree of separation in the Refinery extraction system will vary with the valence conditions. Acid concentration, salting-out effects, and complexing ions will also produce variations in the degree of separation.

Literature information indicates that a separation of neptunium and plutonium from uranium should occur in the Refinery. For neptunium, extraction from nitric acid solutions into tributyl-phosphate is in the order Np (VI) > Np (IV) >> Np (V) and Np (III). For plutonium, the order is Pu (IV) > Pu (VI) >> Pu (III). From these orders, it can be seen that co-extraction of Np and Pu with uranium is favored when the transuranics are in the higher valence states. However, unless strong oxidizing agents are added to the nitric acid feed solution, the lower valence states are favored and Np and Pu will be only partially extracted. No such oxidizing agents are added in the extraction system. Other factors of acid concentration and salting-out effects from the presence of uranyl nitrate act to additionally suppress the extraction of these transuranics.

A small amount of FMPC data from Refinery operations show that about 80% of the Np and Pu present in the feed solution remains in the aqueous raffinate.

After extraction, the barren raffinate solution is adjusted to pH 9-10 with lime and the resultant slurry is filtered or sent to Pit 5 for settling. In all stable valence states, Np and Pu are coprecipitated with insoluble hydroxides and will, therefore, be discarded with the raffinate filter cake or settled solids. Information obtained from the Paducah Gaseous Diffusion Plant shows that over

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99.95% of Np and Pu in waste solutions was precipitated by adjusting to pH 11-12 with sodium hydroxide.

As described in section 2.2, both the filtrate and Pit 5 effluent are combined with other liquid streams and flow to the Great Miami River.

7.2 Metal Production

Several studies have been made at the FMPC to determine the fate of Np and Pu in production processes. Data have been obtained from lab tests and by sampling products and residues from plant operations during the processing of recycle uranium containing Np and Pu. These studies have clearly shown a separation of the transuranics from uranium in the reduction and remelt steps.

During reduction, Np and Pu separate from molten uranium and enter the MgF_2 pot liner and the freshly formed MgF_2 reaction side product. In a lab reduction test, the starting UF_4 contained 7.9 ppm Np and 0.076 ppm Pu (U basis). The resulting derby metal contained 2.9 ppm Np and 0.041 ppm Pu (U basis). In this one test, the Np and Pu separations were 63% and 46%, respectively. No material balance was made but high concentrations of both transuranics were found in the MgF_2 .

In actual production, a Pu separation of 20% was observed for the reduction step. No separation figure was obtained for Np but analysis of the starting UF_4 and the resulting MgF_2 side product indicated that a significant separation did occur.

During the vacuum remelting of uranium metal, Np and Pu separate from the molten metal and enter the remelt residue and the walls of the graphite crucible and mold. In a lab test, Np and Pu separations were 64% and 5%, respectively. A Pu separation of 40% was observed in an actual production remelt operation. Evidence of Np separation was also seen in the production remelt.

MgF_2 from reduction is ground and part is reused as reduction pot liner. Excess MgF_2 containing normal or enriched uranium is leached with nitric acid to recover the uranium. It is expected that most of the transuranics in MgF_2 would be solubilized. The acid leach is processed through the Refinery solvent extraction operation and a UO_3 product is obtained. As noted in section 7.1, most of the transuranics would enter the treated raffinate solids.

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The barren MgF_2 tails after leaching are pumped to Pit 5 following reslurrying with water and adjustment of pH with lime. Remelt residues containing normal or enriched uranium are also processed through the Refinery system.

8.0. EVALUATION OF POTENTIAL ENVIRONMENTAL IMPACT

Transuranium radionuclides introduced in the FMPC production processes will be partitioned between the final products and the production wastes. The final products are sent to other ERDA sites where the handling of materials containing transuranium isotopes is a routine operation. Most of the transuranics in liquid wastes will be precipitated with a great mass of residue and be discarded in on-site storage pits along with residues that are transuranic-free. Soluble trace quantities of transuranics, well under the ERDAM-0524 concentration guides for uncontrolled areas, will be mixed in the plant effluent and discharged to the Great Miami River.

Solid wastes containing transuranium isotopes will be placed in on-site storage pits along with other contaminated solids. Traces of transuranics in dust collector discharges will not result in significant ambient air concentrations.

8.1. Transuranics in Liquid and Solid Wastes

In all stable valence states, Np and Pu in solution will coprecipitate with insoluble hydroxides. In the absence of complexing agents, the coprecipitation is complete. No interferences with complete coprecipitation are anticipated in the treatment of Refinery raffinates. Based on the observed low extractability of Np and Pu in the Refinery system, it is expected that at least 80% of these transuranics in extraction feed solutions will be found in the raffinate. According to Paducah precipitation data, less than 0.05% of Np and Pu present in the raffinate will remain in solution after waste treatment.

The concentration of Np and Pu to be expected in raffinate solids and filtrate may be estimated from Refinery data and an assumed concentration of Np and Pu in extraction feed solutions. Data from recent processing of uranium concentrates show that for each gram of uranium introduced in Refinery feed, there will be generated 0.9 g of wet filter cake (0.3 g of dry cake). These data also show one liter of filtrate generated for each 210 g of uranium in the Refinery feed solution. Mixing of the filtrate with other plant streams will reduce the concentration of

Np and Pu in the final site effluent. Records show that the daily average volume of final effluent is 3.3 times greater than the daily average volume of filtrate from the raffinate treatment.

Estimations of possible Np and Pu concentrations in Refinery feed solutions are speculative. Blending of materials will occur in preliminary steps such as furnacing or leaching and in the preparation of the Refinery feed. For the purpose of these calculations, the concentration of Np and Pu in the feed will be assumed to be 1500 d/m/g U. This concentration, which is equivalent to 1 ppm Np or 10 ppb Pu, is the limit for transuranium alpha activity in FMPC product returned to the Portsmouth Gaseous Diffusion Plant. Although the concentration of Np and Pu in recycle uranium received at the FMPC will be greater than this level, anticipated blending of various feeds is likely to bring the concentration under 1500 d/m/g U.

In summary, the data and assumptions to be used are:

- 1) Np and Pu in extraction feed: 1500 d/m/g U.
- 2) Fraction of Np and Pu not extracted: 0.8.
- 3) Amount of filter cake produced: 0.9 g/g U in feed.
- 4) Amount of filtrate produced: 1 liter for each 210 g U in feed.
- 5) Fraction of Np and Pu not coprecipitated with hydroxides: 0.0005.
- 6) Filtrate dilution factor: 3.3.

The resulting activity in filter cake will be:

$$\frac{1500 \text{ d/m/g U } (0.8)(0.99)}{0.9 \text{ g cake/g U}} = 1320 \text{ d/m/g wet cake}$$

Np and Pu alpha activity in the filtrate from raffinate treatment will be:

$$(1500 \text{ d/m/g U})(210 \text{ g U/liter})(0.8)(0.0005) = 126 \text{ d/m/l.}$$

Finally, after dilution with other plant streams, the activity in the combined effluent discharged to the Miami River will be:

$$\frac{126}{3.3} = 38 \text{ d/m/l}$$

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The ERDAM-0524 Concentration Guide (CG) for unidentified mixtures of Np and Pu in water in uncontrolled areas is 6600 d/m/l. The 38 d/m/l calculated above is only 0.6% of this limit. Average flows in the Miami River would reduce this value to less than 0.0006% of the CG.

As described in section 6.1, solids from the neutralization of raffinate are placed in on-site storage pits which have either a clay liner or a liner of synthetic rubber. Long-term sampling and analysis of water from test wells around these pits has shown that insoluble hydroxides do not migrate and no migration of Np and Pu would be expected. It is not probable that migration from the pits will cause the CG of 6600 d/m/l in ground water to be exceeded. Because of the pit linings, the insolubility of the hydroxides, and the large mass of transuranium-free solids already in the pits, it is highly unlikely that transuranium alpha activity in ground water will ever reach 1% of the CG.

If a waste is produced which has a transuranium alpha activity greater than 22,000 d/m/g, the waste must be packaged and stored in a manner that permits the packages to be readily retrieved in an intact, contamination-free condition for 20 years. This requirement, contained in ERDAM-0511, would prevent high concentrations of transuranium isotopes from being discarded in the open pits. With an extraction separation of 80% and a uranium-to-solids ratio of 1-to-0.9, the special packaging of raffinate solids would be required if the extraction feed had a transuranium alpha activity in excess of 26,000 d/m/g U.

Paducah residues which may be received at the FMPC are known to contain Np and Pu activities above 1500 d/m/g U. Information regarding these recycle materials are given in Table 1.

Table 1. Transuranium Alpha Activity In Paducah Residues

Material	Np ppm	Pu ppb	d/m/g		
			Np	Pu	Total
Feed plant ash	0.89	35.2	1335	5880	6615
Vacuum dust	0.33	21.1	495	3165	3669
Filter cake	4.8*	48*	7200	7200	14400

*Paducah reported that 30 g Np and 0.3 g Pu represents the maximum quantity of Np and Pu which might be in the filter cake. The data were obtained from estimates of the total quantities of transuranics fed to the recovery precipitation system at Paducah. The total quantity of filter cake is 6.25×10^6 g.

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The Np and Pu values given in Table 1 are not on a U basis but are on a material basis. Average uranium content of the filter cake is only 13% and on a U basis the Np and Pu alpha activity is 1.1×10^5 d/m/g U. Obviously, if this material were processed without blending, the raffinate solids would have to be packaged for 20-year retrieval and the transuranic alpha activity in the plant effluent would exceed the CG for uncontrolled areas. Blending would be desired to avoid these two problems. In addition, the Refinery UO_3 product would exceed the specification for transuranic activity in material returned to the Portsmouth Gaseous Diffusion Plant. Blending would, therefore, be necessary to meet product specifications.

Specific plans for future residue processing cannot be given now but must be drawn for each particular residue type. Processing steps will depend upon such factors as impurity levels, uranium concentration and ^{235}U content, availability of other materials for blending, and the desired end product. However, based on past experience, the Paducah filter cake will probably be blended for calcining and the calcined product will be blended with transuranic-free uranium concentrates for extraction. All recycle residues will be blended or otherwise handled in a manner to insure that Np and Pu activity in the final effluent does not exceed the ERDAM-0524 CG.

8.2 Airborne Discharges

Processing of recycle uranium at the FMPC is not expected to produce significant concentrations of airborne transuranium nuclides in the environment. This conclusion is based on the present levels of airborne uranium and the average uranium-transuranium activity ratios expected in the future.

During 1974, the average concentration of airborne uranium found at the six plant boundary stations was 0.7×10^{-14} $\mu Ci/m\ell$, or 0.3% of the ERDAM-0524 Concentration Guide (CG) for uncontrolled areas. If the uranium-transuranium activity ratio throughout FMPC operations, including airborne exhausts, is 1000-to-1, the concentration of airborne transuranium activity at the boundary would be 0.7×10^{-17} $\mu Ci/m\ell$. This concentration is 0.1% of the ERDAM-0524 CG for Pu in uncontrolled areas, and 0.5% of the CG for Np.

The assumed activity ratio of 1000-to-1 is a conservative estimate. It is equivalent to a transuranium content of 1500 d/m/g U. While some FMPC Refinery feeds, notably the Paducah residues, may contain a higher concentration, most

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of the uranium passing through the FMPC production steps will contain much lower concentrations or will be transuranic-free.

Activities around the disposal pits do not add significant amounts of uranium to the offsite air because of the nature of the cake and the application of clean cover. Wet filter cake remains damp for several years due to the movement of moisture to the cake surface. As pit areas are filled they are covered over with fly ash or soil, or both, which effectively ends any potential dusting problem. Data from the closest boundary air sampling do not show any higher concentrations than those found at more distant stations.

9.0. CONFLICTS

In the past, processing of uranium contain transuranium radionuclides has been carried out at the FMPC without the occurrence of conflicts with State, regional, or local plans or programs. No such conflicts are presently known to exist or are anticipated.

10.0. ALTERNATIVES

If reactor recycle materials are not processed at the FMPC, they may be stored, processed elsewhere, or discarded.

Processing at another location would not significantly alter the environmental impact. The FMPC is equipped to do the processing in a manner which would produce an environmental effect no greater than the minimal effect of routine uranium operations. In regard to the Paducah residues, transuranic-free uranium materials currently available at the FMPC are sufficient to provide a blending capacity which would eliminate the waste storage and effluent discharge problems discussed in section 8.1.

Disposal or long-term storage of reactor recycle material would seriously limit the uranium available for power reactors. Full utilization of all uranium material will be needed to avoid a serious uranium shortage. Other disadvantages of long-term storage are discussed in section 4.0.