ESTIMATE OF THE SOURCES OF PLUTONIUM-CONTAINING WASTES GENERATED FROM MOX FUEL PRODUCTION IN RUSSIA

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ABSTRACT

In Russia, mixed oxide (MOX) fuel is produced in a pilot facility "Paket" at "MAYAK" Production Association. The Mining-Chemical Combine (MCC) has developed plans to design and build a dedicated industrial-scale plant to produce MOX fuel and fuel assemblies (FA) for VVER-1000 water reactors and the BN-600 fast-breeder reactor, which is pending an official Russian Federation (RF) site-selection decision. The design output of the plant is based on a production capacity of 2.75 tons of weapons plutonium per year to produce the resulting fuel assemblies: 1.25 tons for the BN-600 reactor FAs and the remaining 1.5 tons for VVER-1000 FAs. It is likely the quantity of BN-600 FAs will be reduced in actual practice.

The process of nuclear disarmament frees a significant amount of weapons plutonium for other uses, which, if unutilized, represents a constant general threat. In France, Great Britain, Belgium, Russia, and Japan, reactor-grade plutonium is used in MOX-fuel production. Making MOX-fuel for CANDU (Canada) and pressurized water reactors (PWR) (Europe) is under consideration in Russia. If this latter production is added, as many as 5 tons of Pu per year might be processed into new FAs in Russia.

Many years of work and experience are represented in the estimates of MOX fuel production wastes derived in this report. Prior engineering studies and sludge treatment investigations and comparisons have determined how best to treat Pu sludges and MOX fuel wastes. Based upon analyses of the production processes established by these efforts, we can estimate that there will be approximately 1200 kg of residual wastes subject to immobilization per MT of plutonium processed, of which approximately 6 to 7 kg is Pu in the residuals per MT of Pu processed.

The wastes are various and complicated in composition. Because organic wastes constitute both the major portion of total waste and of the Pu to be immobilized, the recommended treatment of MOX-fuel production waste is incineration or calcination, alkali sintering, and dissolution of sintered products in nitric acid. Insoluble residues are then mixed with vitrifying components and Pu sludges, vitrified, and sent for storage and disposal.

Implementation of the intergovernmental agreement between Russia and the United States (US) regarding the utilization of 34 tons of weapons plutonium will also require treatment of Pu-containing MOX fabrication wastes at the MCC radiochemical production plant. The joint treatment and immobilization of both MOX-fuel production wastes and radiochemical

production plant Pu-sludge wastes appears to be both feasible and cost effective. One proposed scenario combines these two types of wastes at the pre-flux stage of the glass matrix preparation in a single MCC integrated waste treatment facility. However, no matter what process is ultimately adopted in the RF, the combined efforts of our two countries to eliminate such nuclear weapons consequences of the arms race would allow us to enter the new millennium with greater optimism.

INTRODUCTION

The estimated sources of Pu-containing wastes presented in this paper were obtained during contract work performed to immobilize weapons Pu-bearing sludge wastes in matrices in order to prevent recovery for military uses. This contractual work was entered into by and between the MCC and Lawrence Livermore National Laboratory. This collaboration was possible due to the Intergovernmental Agreement between the RF and the US on Management of Plutonium that has Been Withdrawn from Nuclear Military Programs, and the agreement between the RF and the US Department of Energy (DOE) on the scope of the cooperative activities.

Since May, 1998, three contracts, all entitled "Immobilization of MCC's plutonium-bearing waste" have been signed. Each contract had a particular objective, as described below:

- Contract B347676 – Preparation of feasibility studies of comparison of two technologies to reprocess plutonium-bearing sludges with and without recovery of plutonium (Feasibility Study).
- Contract B506210 – Preparation of Declaration of Intent (DOI) for implementation of Pu-Bearing Sludges Vitrification Facility and Vitrified Waste Storage Facility at MCC. The Russian DOI is basically equivalent to portions of the US DOE ROD and NEPA process for nuclear projects and is required in Russia for new projects.
- Contract B506233 – Development of Justification of Investment (JOI) for implementation of Pilot Plutonium-Bearing Waste Vitrification Facility and Vitrified Waste Storage Facility. The Russian JOI is equivalent to developing a facility engineering design that is somewhere between a detailed conceptual design and a preliminary (or Title I) design that also includes developing life cycle cost estimates, preliminary environmental assessments, and preliminary safety analyses.

MCC, together with specialists from five RF institutes, All-Russian Design Research Institute of Complex Power Technology (VNIPIET), V.G. Khlopin Radium Institute (KRI-NPO RI), A. A. Bochvar All-Russia Scientific Research Institute of Inorganic Materials (VNIINM), Scientific Research and Design Institute of Installation Technology (VNIKIMT), and All-Russian Research and Design Institute of Production Engineering (VNIPIPT), dealt with the task of how to reliably process and immobilize Pu-bearing sludges generated from radiochemical activities at the processing facilities of MCC.

Preparatory activities, documentation of the work, and some applied research and development has been conducted. Experiments were conducted both in laboratories and in specially equipped "hot cells" that apply microwave ultra-high frequency (UHF) fields to process and vitrify products. Analysis of Pu-bearing sludge reprocessing feasibility studies shows that vitrification

of wastes in borosilicate glass matrices without Pu recovery appears to be more efficient than either vitrification in phosphate glass or plutonium recovery for reuse.

BACKGROUND

Comparison of Options for Reprocessing of Pu-bearing Wastes

A comparative assessment of the engineering and economical potential has been performed of MCC's two possible Pu-bearing sludge management technologies: (1) reprocessing sludge with plutonium recovery, and (2) direct waste immobilization into a glass matrix using UHF microwave power [1,2].

In the three feasibility and contract studies, three Pu-bearing sludge reprocessing options were examined:

- Option 1 - Sludge reprocessing with Pu recovery and purification as PuO_2, with cementation of insoluble waste residues (an existing reprocessing procedure at MCC);
- Option 2 - Microwave sludge solidification without Pu recovery and with borosilicate glass production (the V. Khlopin NPO RI option);
- Option 3 - Microwave sludge solidification without Pu recovery, and with phosphate glass production (the A. Bochvar VNIINM option).

Option 1 is considered to be the baseline option, and options 2 and 3 are compared to it. Figure 1 shows a schematic diagram for processing sludge as described and assessed in MCC feasibility studies. All three options use the same technique for sludge removal. Additional information is provided in paper 6, abstract 466 of this Conference [3].

The following method is in use at MCC for emptying a Pu-sludge storage tank containing radioactive waste. The MCC wastes are stored in large tanks set in excavations in rock located at process areas underground. Each storage tank is a reinforced concrete reservoir 3200 m^3 in volume, 30 m in height, and 12 m in diameter, lined inside with sheet stainless steel. Liquid medium-level alkali and nitrate wastes are clarified in the tanks. Then the clarified portion is decanted, and the solid phase, i.e., sludge, is stored in a tank. The heights of the stored residuals in the tanks varies from 4 to 22 m. The sludges differ in composition and density.

Approximately 6,000 m^3 of radioactive sludge-like waste has been accumulated in the tanks to date and approximately 600 kg of Pu is estimated to be in storage.

The MCC plans to decommission the storage tanks in a stage-by-stage manner. One of the difficult tasks is to wash out sediments and treat the recovered sludges in such a way that it would be possible to pump it out for further reprocessing. Special equipment has been developed, manufactured, and tested for sludge discharge at the MCC.

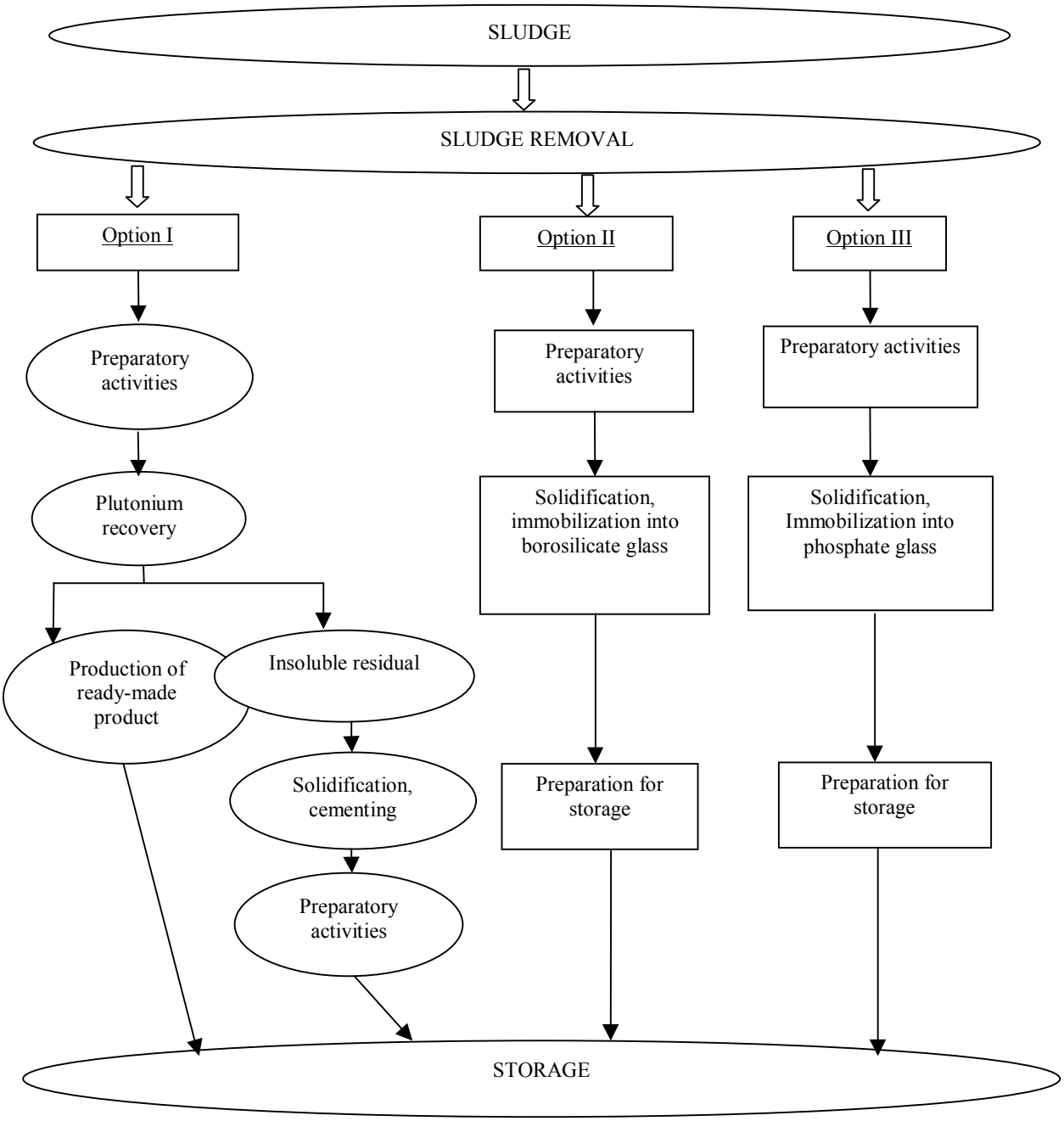

Fig. 1. Schematic diagram of Pu-sludge processing.

Tank AG-8301/3 has been used as a test facility and reservoir. Height of sludge accumulated in the tank varied from 4 to 4.5 m. To agitate and remove the sludge, special removal equipment was installed in risers of different diameters, which perforated into the tanks.

This tank serves as a large-scale laboratory to obtain reliable sludge composition and density data and to study and improve sludge agitation techniques. Because equipment to agitate and thus obtain a uniform sludge was not installed at the tank beforehand, the sludge composition is non-homogeneous in height. The solid phase concentration varies from 60 g/l in the upper layer

up to 800 g/l in the lower residual layer. Such sludge forms a spatial skeleton and contains almost no free liquid phase.
Owing to a successful combination of engineering decisions and techniques, sludge removal equipment has been continuing to operate in a removal mode. Figure 2 shows the equipment installed in the tank. After removal from the storage tank, the sludge is prepared for further processing. The type of preparation depends on the final reprocessing option.

For the basic option (option I), sludge is scrubbed by concentrating and dissolving out the soluble salts. Then, the solution is sent for Pu recovery. An insoluble sludge portion that contains no plutonium is solidified in containers 0.2 m^3 in volume, employing a cementing facility. The cement solid wastes are then sent to storage prior to disposal.

In option II, two-phase scrubbing is performed to decrease the sodium nitrate content; then the sludge is concentrated until its solid phase concentration reaches 120 g/l. The removal of sodium improves the quality of the resulting glass. The concentrated sludge then is mixed with glass-forming additives, and fed into the melter crucible. Finally, the sludge is immobilized into borosilicate glass using a microwave melter technology.

In option III, the sludge is subject only to concentration up to a concentration of 120 g/l before immobilization into a phosphate glass using microwave melting technology.

In both vitrification options, the immobilization of Pu-bearing waste into a glass-like matrix is performed in a capsule crucible and afterwards the crucible is installed in a multi-purpose storage canister. The solidified wastes are forwarded for storage in specially equipped storage facilities located in MCC's underground cavities.

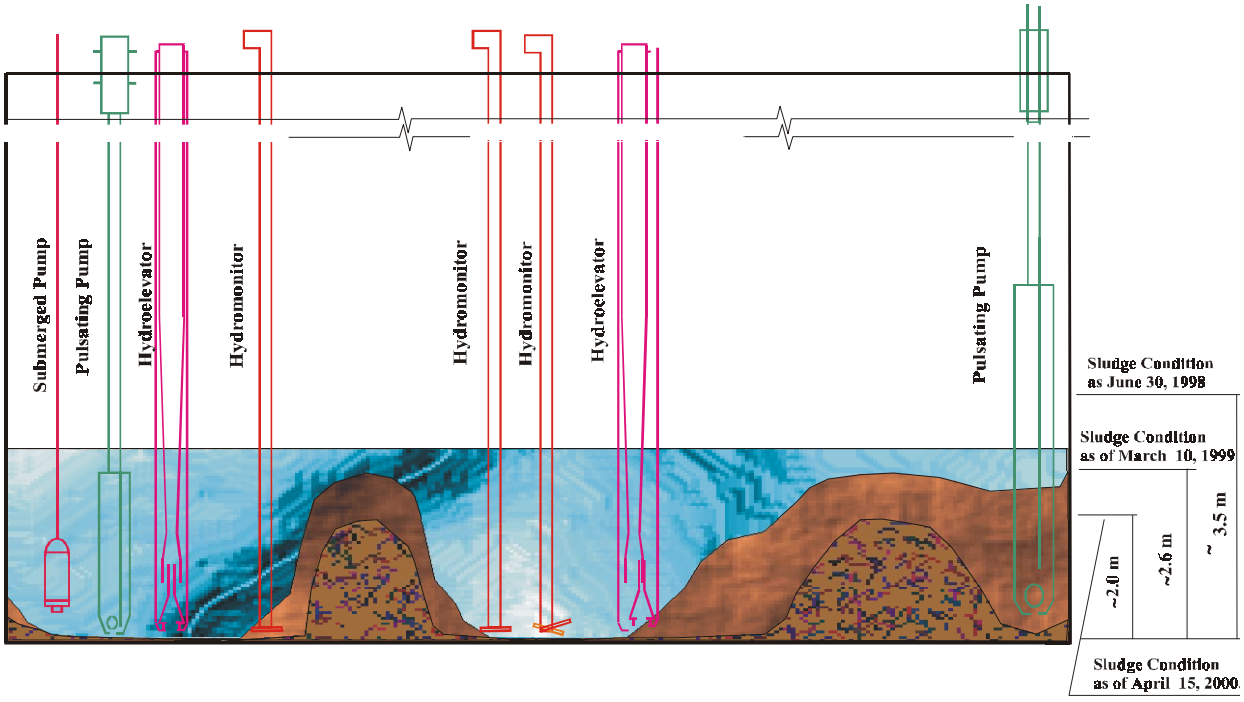

Fig. 2. Illustration of the condition of sludges in a 3200-m^3 tank as a function of time.

Comparisons of the three options, as estimated at the completion of the feasibility studies, show that the immobilization of Pu-bearing sludge into a glass-like matrix is more cost effective than the basic option of sludge processing with Pu recovery. Results from hot cell experiments in the MCC have persuaded specialists of the efficiency of the 5-kW microwave facility, and that the immobilization of waste into both borosilicate-glass and phosphate-glass matrixes is reproducible and that both vitrification techniques are a viable technology.

Long-term Work Plan for Pilot Waste Reprocessing and Storage Facilities at MCC

The last task in the first contract feasibility study, stage 1, was preparation of the long-term work plan diagrammed in Fig. 3. The plan ends with implementation of the pilot vitrification and storage facilities in stage 8. All activities listed in the work plan are to be performed over a 4.5-year period, provided that funds are allocated in the amount of $17.3.M US.

In the second DOI contract stage, a detailed plan was prepared over a six-month period to set out the objectives of specific investments and technical objectives to be implemented. These were agreed to by the RF Regional Administration, and approved by the RF Ministry of Atomic Energy. Only two options of waste reprocessing were considered, and both options envisaged immediate immobilization of Pu-bearing waste either into borosilicate or phosphate glass matrices. The option of reprocessing the Pu sludges to recover the Pu was not to be considered further based on the DOI.

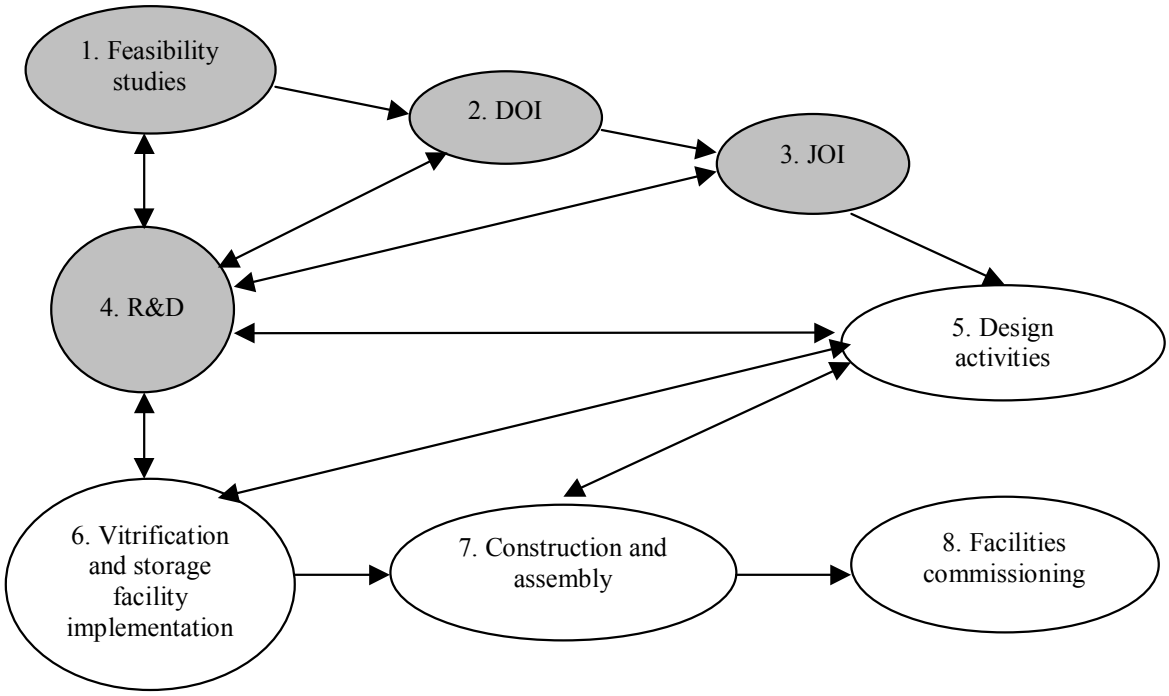

Fig. 3. Illustration of stages 1-8 of the long-term work plan for tasks in RF-US DOE Contracts B347676, B506210, and B506233 (shaded stages-work underway).

At the third stage, the product of the RF-US collaboration became a Task Order development for the JOI. After intensive discussion of all stages of work and minor modifications and corrections, the third contract, B506233, was signed. During 2000-2001, MCC together with the RF Institutes developed the JOI for construction of the pilot vitrification facility and the vitrified waste storage facility. The preparation of the JOI was a ladder-type stage, involving the following subtasks:

- Develop Justification of Investment documentation (that is an intensive item of work);
- Conduct hot cell experiments with the actual product;
- Improve particular units of the pilot facility and prepare engineering specifications for the latter in an independent document;
- Determine which one of two technologies to use for waste immobilization into the solid matrix;
- Prepare a future work plan.

The JOI is the final stage in choosing between the two waste vitrification technologies, according to the terms of the contract. Based on results from experiments, the borosilicate matrix was selected because this vitrification technique, which is based on a heating process with applied UHF-energy, proved to be more technologically effective. In addition, the economic potential determined during feasibility studies also favored the borosilicate glass option. It was shown that placement of the multi-purpose canisters (MPC) of solidified wastes for the borosilicate glass matrixes would require only three storage facilities (6800 MPC), while the canisters for phosphate glass matrixes would require five storage facilities (11860 MPC).

Pursuant to the Russian standards, the JOI must show economic necessity, engineering feasibility, and social expediency for the funds invested. The JOI calls for the submission of sufficient information to make the necessary agreements and prepare expert assessments. To meet these requirements, a 12-volume document was prepared that contains construction and technical decisions, primary requirements for non-standardized equipment, radiation and nuclear safety decisions, and the environmental impact assessment. Currently, the JOI is under review for agreement according to Russian policies and requirements.

Five applied research and development (R&D) reports have been completed that describe the large amount of vitrification work that has been accomplished. Many R&D issues have been resolved, including:

- Basic principles to control the vitrification process have been determined;
- Plasma formation in the air space of the melter has been mitigated, and the magnetron has been protected against plasma;
- Chemical stability of vitrified waste has been studied with a specially installed facility;
- Auxiliary ohmic crucible heating has been applied that makes it possible to obtain a glass block of proper quality and without crust formation;
- The actual sludge, which contains up to 450 mg of plutonium per 1 kg of glass and fragmentation radionuclides, has been immobilized into borosilicate glass and melted. According to results from tests on chemical stability and mechanical strength, the product meets Russian standards requirements.

If the positive RF decision to site a full MOX-fuel production industrial facility at MCC is made, the following technological changes to the MCC facilities are to be made:

- Reception, incoming inspection, temporary storage of input materials and accessory items;
- Preparation of items made of plutonium metal for utilization;
- Anodic dissolution of plutonium alloys;
- Extraction refinement of plutonium from solutions;
- Dissolution of a part of uranium dioxide powder;
- Production of master-mixture containing uranium dioxide and plutonium dioxide;
- Production of molding powders;
- Production of fuel bushes (pressing, grinding, sintering);
- Production, storage, and replenishment of many fuel rods;
- Fuel assembly production, storage, and replenishment of transportation containers;
- Utilization of technological production wastes.

ESTIMATED SOURCES OF WASTES FROM MOX FUEL PRODUCTION AT THE MCC

As part of these LLNL contracts, assessments were made as how to treat and immobilize the Pu-containing wastes generated from MOX fuel production (or fabrication) at the MCC. No wastes from MOX spent fuel reprocessing are considered in these assessments.

The intrinsic wastes from MOX-fuel production, according to the literature, previous work, and results from existing production analyses, are highly varying and complex in composition. However, this waste can be divided into three categories:

1. Defective primary products;
2. Inorganic waste;
3. Organic waste.

Table I provides data on the estimated contribution of each waste category to total waste and on plutonium content. From data in Tables I and II , it can be seen that organic waste from MOX-fuel production makes up the major portion of total waste and contains the majority of the plutonium. It is essential to point out that there are a variety of organic waste types (e.g., latex and chloropolymers, viscose and cotton materials, wood and sorbents). That is why special attention should be paid to the development of organic waste utilization technology for waste products from MOX-fuel production.

Figure 4 shows a block diagram for reprocessing the waste generated from MOX-fuel production. Insoluble waste after incineration or calcination, alkali sintering, and dissolution of sintered products in nitric acid are fluxed and then vitrified. We propose that this method be used to treat MOX fuel production wastes for storage and disposal in a vitrified form. An option to

add the MOX fuel fabrication wastes to the Pu-sludge vitrification processes was also explicitly assessed.

During the activities associated with JOI preparation, there was active discussion of the possible production quantities at MCC of heat-releasing fuel assemblies (FA) based on mixed uranium and plutonium fuel. If FA's are produced only for VVER-1000 and BN-600 reactors, 2.75 MT of weapons plutonium would be used annually. However, if deliveries of MOX-FA's for extra reactors such as Canadian and European are produced, up to 5 MT/y of weapons-grade plutonium would be used. Commissioning of this extra production would result in the generation of extra Pu-bearing wastes. Having studied the issues of vitifying Pu-sludge wastes and radiochemical production waste processing, it was timely and useful to consider the possibility of the joint immobilization of such types of waste. One of the scenarios proposed for consideration is combining these two types of wastes in certain ratios at the pre-flux stage. This option is shown with dotted lines in Fig. 4.

The costs to implement Pu-bearing waste reprocessing have been approximately estimated in the JOI and are summarized in Table III. It is evident that joint reprocessing and vitrification of the two waste types is cost effective. A cost difference of $3 million compared to $12-14 million is attractive and an earnest argument in favor of continuing this work. However, it must be clear that these values presented here are approximate. Therefore, a decision has been made to continue work on a feasibility study of options for processing the Pu-bearing waste generated from MOX-fuel production in order to develop a better cost estimate. This work has become an independent item in a plan to investigate promising trends in waste disposal options for the MCC.

Handling of radioactive wastes safely is another one of the many problems that are of importance in nuclear facilities. The MOX fuel wastes contain significant amounts of radioactive nuclides, including highly dangerous ones. MCC is cognizant of the task to utilize a technology that is as harmless as possible to the environment during the lifetime of all radioactive elements. Vitrification of these wastes appears to be a viable technology for waste treatment.

CONCLUSIONS

Actual sources and quantities of Pu-bearing wastes generated by the MOX fuel reprocessing scenarios described here are dependent upon actual production experience. This will only be attained after the RF review and siting decisions with construction and operation of the facility, and with continued US and RF funding.

The MCC has established all the conditions required to deploy MOX-fuel production at their site. Sufficient free underground mined cavities for installation of the necessary production equipment are an opportunity that encourages an RF decision to locate production at this site. In addition, MCC's proximity to the most promising site for a geological repository, the granitoids of the Nizhnekansky massif, help solve the waste disposal problem in an ecologically safe manner. This site would provide:

- Interim storage under permanent control;
- Geological disposal in a repository (solidified radioactive waste).

Table I. Waste sources and their plutonium content.

Waste category	Waste description	Insoluble residues, subject to immobilization, kg/MT Pu	Pu in residuals, subject to immobilization, kg/MT Pu
Defective primary products	Defective bushes (VVER-1000).	37.2	0.37
	Powder generated from disassembly of defective heat-releasing elements (BN-600)	1.1	0.53
	TOTAL:	38.3	0.9
Inorganic waste	Powder, resulted from grinding, cleaning, wipe.	8.5	0.08
	Items made of molybdenum (VVER-1000)	26.0	0.05
	Equipment manufactured of pyrolitic graphite (BN-600)	125	0.05
	Salt-electrolyte (BN-600)	60	0.36
	Fuel rod shells (wastage) (VVER-1000, BN-600)	100	0.1
	Glass fiber (VVER-1000, BN-600)	8	0.12
	Scrap metal (VVER-1000, BN-600)	13.3	0.33
	Glass (VVER-1000, BN-600)	0.46	0.01
	Alundum (VVER-1000)	8	0.8
	Residuals from clarification of the solutions, generated from equipment decontamination, and the like.	3.3	0.02
	Asbestos (VVER-1000)	1.3	0.02
	TOTAL:	353.86	1.94
Organic waste	Cleaning materials (VVER-1000, BN-600)	97.5	0.8
	Filter clothes (VVER-1000, BN-600)	375	2.5
	Rubber items (VVER-1000, BN-600)	195	0.26
	Wood wastes (VVER-1000, BN-600)	75	0.03
	Film materials (VVER-1000, BN-600)	15	0.03
	Oil for vacuum pumps (VVER-1000, BN-600)	3.9	$6.6 \cdot 10^{-5}$
	Used sorbents (VVER-1000, BN-600)	3.9	$3.3 \cdot 10^{-3}$
	TOTAL:	765.3	3.623
TOTAL WASTE:		1157.46	6.463

Table II. Waste categories as percent of total waste and the Pu content as percent of each waste category.

	Waste quantity as % of total waste	Pu as % of each waste category
Defective primary products	3.3	14.0
Inorganic waste	30.5	30.0
Organic waste	66.2	56.0

Table III. Estimated costs of reprocessing Pu-bearing waste.

Type of waste	Cost ($M) (US)
Radiochemical facility waste: Pu sludges only	18
MOX-fuel production waste only	12 – 14
Joint reprocessing of both waste types: MOX wastes and Pu sludges	21

The MCC prefers a technology for the reliable immobilization of plutonium-bearing waste. We believe that continuation of the RF-US collaboration to study these immobilization technologies will be to the benefit of both countries. It is essential to add that the nature of radioactive waste won't allow us to forget to monitor its status whether relations between politicians are good or bad.

ACKNOWLEDGEMENTS

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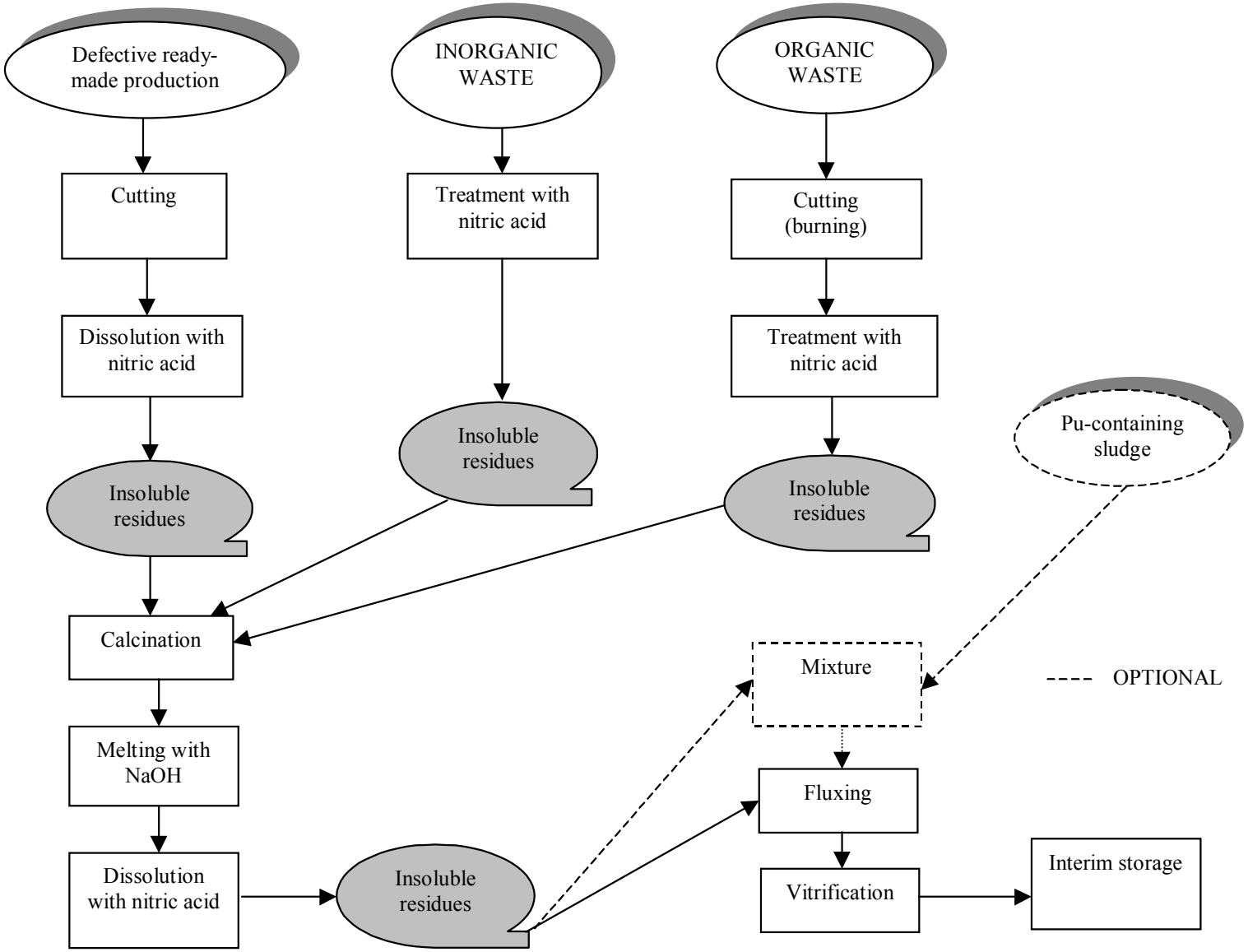

Fig. 4. Block diagram of reprocessing Pu-bearing waste generated from MOX fuel production.