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TREATMENT OF SOLID WASTE HIGHLY CONTAMINATED BY ALPHA EMITTERS: RECENT DEVELOPMENTS OF LEACHING PROCESS WITH CONTINUOUS ELECTROLYTE REGENERATION

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TREATMENT OF SOLID WASTE HIGHLY CONTAMINATED BY ALPHA EMITTERS : RECENT DEVELOPMENTS OF LEACHING PROCESS WITH CONTINUOUS ELECTROLYTE REGENERATION

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ABSTRACT

Development of processes for leaching solid waste contaminated by alpha or alpha/beta/gamma emitters has been pursued at the Nuclear Research Center in Fontenay-aux-Roses, France with the recent active commissioning of two pilot facilities: the Elise glove box system in February 1987 and the Prolixe shielded hot cell in March 1988. The Elise facility is designed to handle alpha waste and the Prolixe facility is designed to handle alpha/beta/gamma waste. The common goal of the studies conducted in these facilities is to define the operating conditions for declassification of solid waste, i.e. to ensure that the alpha concentration of this waste will be less than 3.7×10^6 Bq/kg after treatment, packaging and decay prior to storage in surface repositories. The leaching process developed is mainly based on the continuous electrolytic regeneration of an aggressive agent, AgII, which can induce the dissolution of PuO_2, the most difficult compound to remove from the solid waste. This paper summarizes recent achievements in the development of this process.

INTRODUCTION

The nuclear activities conducted in the radiochemistry building of the Nuclear Research Center at Fontenay-aux-Roses mainly pertain to the study of nuclear fuel reprocessing and the production of transuranic elements such as 241Am and 244Cm. These activities, which are carried out in glove boxes or inside shielded hot cells, generate solid waste that is contaminated by alpha-emitter radioisotopes in the case of the glove boxes and by alpha/beta/gamma-emitter radioisotopes in the case of the shielded hot cells.

To facilitate management of this solid waste, a program was established with the following objectives : 1) construction of two solid waste research and treatment facilities : Elise, a system consisting of three glove boxes for treatment of alpha waste; and Prolixe, a shielded hot cell for treatment of alpha/beta/gamma waste ; 2 study of leaching methods to be used to decontaminate the waste to ensure that their residual contamination is compatible with surface repository disposal of the packaged waste (residual alpha contamination $< 3.7 \times 10^6$ Bq/kg) ; 3) preparation of process flow sheets for waste treatment with enough versatility to accommodate waste from other facilities and thus enabling the design of industrial waste treatment systems. The leaching process developed is primarily based on electrolytic regeneration of a reagent, AgII, which can induce the dissolution of highly refractory plutonium compounds such as PuO_2 in a nitric acid medium at ambient temperature (1-10). The principle of this process and its main steps are reviewed below, together with a brief description of two pilot facilities. In addition, a report is provided on the status of process development in Elise, Prolixe and industrial facilities for PuO_2 oxidizing dissolution (UP3 reprocessing plant) and treatment of plutonium-containing ashes (UP1 reprocessing plant).

PROCESS PRINCIPLE AND MAIN STEPS

The solid waste decontamination process employed is based on the nitric acid waste leaching technique. To achieve alpha-emitter decontamination factors compatible with the waste declassification target, i.e. obtaining a residual alpha contamination of less than 3.7×10^6 Bq/kg for the decontaminated, packaged waste, the nitric acid leaching is performed with AgII, a highly oxidizing reagent (E_o (AgII/AgI) = 1.92 V/NHE in HNO_3 medium) capable of inducing the dissolution of refractory compounds of plutonium (the principal radioactive contaminant of solid waste), e.g. PuO_2.

Relatively large quantities of the AgII reagent are required because it reacts with some components of the waste, such as cellulose. the electrolytic regeneration technique was adopted for the AgII to minimize the quantity of reagent required for treatment. The operating principle for this technique is shown in Fig. 1.

The solid/liquid exchange area must be increased to improve the kinetics of radionuclide transfer from the solid waste to the solution. This is achieved by crushing the waste into fragments with maximum dimensions of $\leq 10^{-2}$ m, which also significantly reduces the apparent volume of the waste. Such operations are not feasible for waste consisting of massive metal parts which must therefore be carefully sorted before crushing the main waste feed; these metal parts will be treated during special leaching campaigns. Once most of the radioactive contaminants have been transferred to the leaching solution, this solution is concentrated to permit recycling of most of the liquid stream and subsequent recovery of radioactive contaminants such as Pu, Am and Cm, as well as of the silver contained in the concentrates. The main steps in this process are listed in Table I.

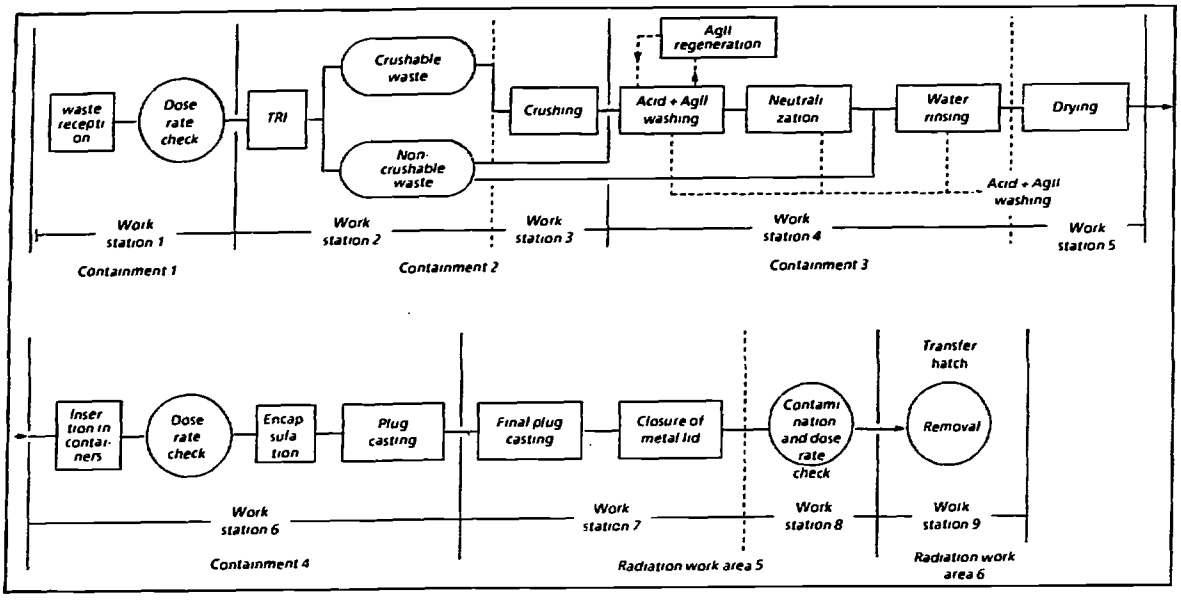


Fig. 1. Principle of Solid Waste Leaching With Electrolytically Regenerated AgII

TABLE I
Main Steps in Solid Waste Treatment Process

STEP	*TARGETS*
SORTING	- *Separation of crushable and non-crushable materials*
CRUSHING	- *G Increase of solid/liquid exchange area* - *Reduction of waste apparent volume*
LEACHING HNO_3 + AgII H_2O	- *Virtually quantitative transfer of radioactive contaminants from solid to leaching solution*
DRYING	- *Minimize residual water content to make decontaminated waste compatible with epoxy resin encapsulation* - *Enable easy transfer of decontaminated waste*
ENCAPSULATION	- *Obtain a package suitable for surface repository storage*
CONCENTRATION OF LEACHATES	- *Recycle main liquid stream to another treatment line* - *Minimize volume of treatment feed solutions to recover radioactive contaminants and silver*
CHEMICAL TREATMENT OF CONCENTRATES	- *Recover radioactive contaminants for reuse* - *Recover silver for recycling*

DESCRIPTION OF PILOT FACILITIES

Elise Facility

The Elise facility is a system consisting of three glove boxes used to treat solid alpha waste. This facility is shown in Fig. 2.

Glove box no. 1 is used to: 1) receive the waste initially contained in polyvinyl chloride (PVC) bags placed in 200 dm3 oil drums; 2) manually sort the waste after cutting open the PVC bags and removing the non-crushable metal parts; 3) crush the waste.

The following operations are performed in glove box no. 2 : 1) waste leaching in an AgII nitric acid solution ; 2) electrolytic regeneration of AgII ; 3) rinsing of waste with water ; 4) drying of waste with hot air ; 5) concentration of contaminated leachates. The waste leaching, rinsing and drying operations are all performed in the same reactor. Glove box no. 3 is used to encapsulate the decontaminated dry waste in an epoxy resin matrix. The waste crushed before and after leaching and drying is transferred from one glove box to another by suction devices.

Prolixe Facility

Prolixe is a shielded system used to treat alpha/beta/gamma waste. This facility is shown in Fig. 3.

This solid alpha/beta/gamma waste treatment facility consists of a shielded hot cell with nine work stations. Each work station has: 1) type MA II/80 and MT 200 master-slave manipulators made by La Calhene ; 2) a viewing system including a lead-glass window and an alpha containment window ; 3) process components. The first six work stations are arranged in four stainless steel alpha containments shielded by a 0.15 m lead wall ; the other three work stations are located in two radiation work areas with a simple dynamic containment shielded by a 0.1 m steel wall.

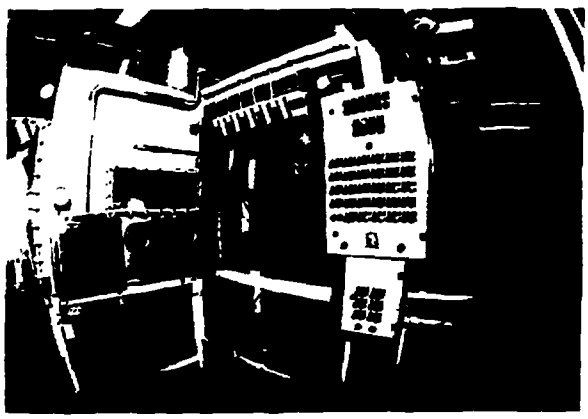


Fig. 2. The Elise Pilot Facility for Treatment of Solid Alpha Waste

Fig. 3. The Prolixe Shielded Pilot Facility for Treatment of Solid Alpha/beta/gamma Waste.

The first containment receives the alpha/beta/gamma solid waste packaged in type AA29 containers made by La Calhene on a mechanical conveyor from the nearby Petrus shielded hot cell, and from the other shielded hot cells in the building in type RD10 or RD15 shielded transport casks.

In the second containment the waste is inventoried, sorted and crushed according to type.

The third containment is used for leaching and drying the crushed waste. The dry, decontaminated waste is then transferred to the fourth containment for encapsulation and final packaging.

The encapsulated waste package is closed in the first gamma area.

The closed package is checked in the second gamma area before removal from the containment.

Figure 4 shows all the operations performed and their location in the shielded hot cell.

PROCESS DEVELOPMENT STATUS

Sorting and Crushing

After installation in the containments the AA29 containers (Prolixe) or PVC bags (Elise) are opened and the

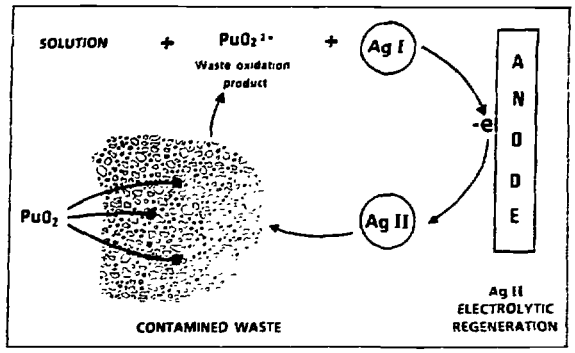


Fig. 4. Prolixe Facility Process Flow Sheet Indicating Arrangement of Work Stations.

waste is sorted according to crushability. Only the waste consisting of massive metal parts cannot be crushed ; these items represent an average of less than 15% of total waste. Table II indicates the average composition of the crushable waste.

In the Elise and Prolixe facilities the waste is crushed by an industrial unit with circular blades fitted on two parallel shafts. The shafts are driven by a 7.5 kW electric motor and rotate at a speed of 1.1 revolutions per second (rps).

The crushed waste is graded by a metal grate that retains the crushed materials larger than 10^{-2} m. If the feed hopper is overloaded or in case of excess demand, the crusher is automatically shut off and the rotating direction of the shafts is reversed. Following shutdown, crushing is resumed at a normal rate.

The average crusher throughput is 6.6×10^{-3} kg/s (24 kg/h) and is limited mainly by the calibrating grate.

Since the start of active operation, the following amounts of radioactive waste have been sorted and crushed, respectively :

Elise (active commissioning in February 1987) = 3600 kg or 120 drums of 0.200 m^3 capacity.

Prolixe (active commissioning on March 25, 1988) = 960 kg or 270 type AA29 La Calhene containers of 0.017 m^3 capacity.

The average volume reduction factor achieved after crushing is close to 2.5 for both facilities.

Most of the crushed waste is repackaged for shipping to an offsite treatment facility. Only the high-level alpha waste fraction (Pu for Elise waste and ^{241}Am and ^{244}Cm for Prolixe waste) is kept for further processing.

TABLE II
Average Composition of Crushable Waste.

MATERIAL	*% BY WEIGHT*	*WASTE TYPE*
*PE**	*20*	- *Sample vials* - *Cans 0.25 to 10 dm³*
*PVC***	*27*	- *Glove box sleeves* - *Strips* - *Armored hoses*
ELASTOMERS	*21*	- *Glove box gloves* - *Pre-gloves*
PLEXIGLAS	*14*	- *Chromatographic columns* - *Apparatus*
CELLULOSE	*14*	- *Cardboard* - *Paper* - *Cotton*
GLASS	*2*	- *Laboratory equipment* - *Vials, pipettes*
COPPER	*2*	- *Electric cables*

**PE = Polyethylene **PVC = Polyvinyl chloride*

Leaching

The leaching reactors installed in the Elise and Prolixe hot cells (one and two reactors, respectively) have an internal capacity of about 70 dm^3. The crushed waste of 20 kg or less is contained in a polypropylene filter bag (retention capacity of 5×10^{-5} to 3×10^{-4} m or 50 to 300 μm) placed in a perforated stainless steel or zirconium basket. The leaching solution of 50 dm^3 or less flows through the reactor in a loop configuration. The electrolysis unit for continuous Ag II regeneration is located outside the reactor. During leaching, the waste/leaching solution mixture is agitated by a turbine driven at a speed of 2 rps by an electric motor integral with the reactor lid. Figure 5 provides an internal view of the Elise leaching reactor.

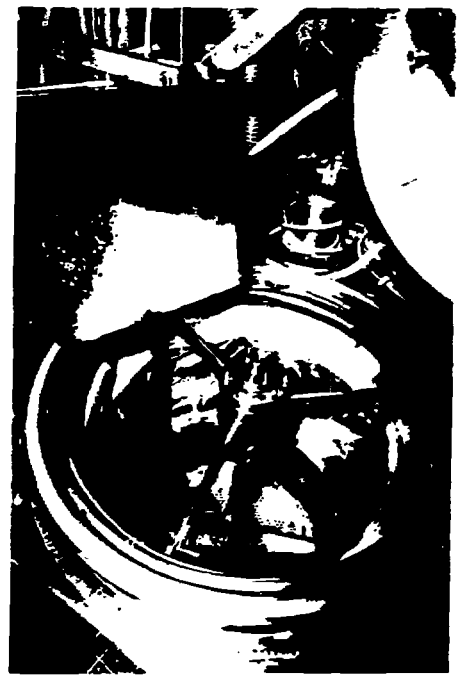


Fig. 5. Inside View of the Elise Leaching Reactor.

HNO_3 Leaching

Most of the leaching campaigns conducted to date were for treatment of crushed waste using nitric acid solutions with an acidity of 1 to 4 M at ambient temperature.

Two representative types of waste were selected for these studies : 1) waste contaminated by plutonium initially contained in an aqueous or organic (tributylphosphate) solution from a reprocessing research cell in the case of Elise ; 2) waste contaminated by ^{241}Am and ^{244}Cm in oxide form (AmO_2 and CmO_2) generated in a neutron source preparation cell (alpha/neutron reactions) in the case of Prolixe. Table III lists the overall results of the nitric acid leaching performed.

Successive operations were carried out using a new leaching solution batch for each step to determine the limits of the nitric acid leaching technique. The performance of the treated waste batch is determined by analysis of waste samples taken before and after treatment. The representativeness of each sample was confirmed by the absence of significant scattering of data (about 1% variation) for the analyses performed after destruction of the samples. Three to four samples were taken before and after treatment of a waste batch.

TABLE III
Results of Waste Leaching Campaigns Conducted in the Elise and Prolixe Facilities

	ELISE	*PROLIXE*
Total weight of treated waste (kg)	*19*	*168*
Number of leaching campaigns	*20*	*9*
Desorbed α activity *Bq* *(Ci)*	*5.14×10^{11}* *(13.9)*	*1.77×10^{13}* *(480)*
Principal leached radioisotopes	*^{238}Pu ; ^{239}Pu ; ^{240}Pu ; ^{241}Am*	*^{244}Cm ; ^{241}Am*

TABLE IV
Leaching of Plutonium-Containing Solid Waste in Elise.

Waste feed	weight (kg)	9,78													
	α activity [Bq (Ci) / Bq/kg (Ci/t)	4,70 10^{10} (1,272) 4,81 109 (130)													
Leaching no.		Main waste											Fines			
		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Leaching solution	HNO_3 (M)	1,72	1,72	1,4	1,3	1,5	1,58	1,24	1,115	1,37	1,45	0	1,19	1,19	1,19	0
	volume (dm^3)	48	37	43	39	42	39	44	39	41	38	39,5	23	18	19	20
Time (s)		2400											2400			
Desorbed α activity (Bq)		1,76 10^{10}	4,6 10^{9}	2,85 10^{9}	9,25 10^{8}	4,4 10^{8}	6,2 10^{8}	7,8 10^{8}	3,1 10^{8}	2,95 10^{8}	2,9 10^{8}	8,9 10^{7}	2,4 10^{9}	9,6 10^{8}	7,2 10^{8}	1,2 10^{8}
Total desorbed α activity Bq (Ci)		2,88 10^{10} (0,7784)											4,19 10^{9} (0,113)			
		3,3 10^{10} (0,892)														
Solutions balance	leaching [total (dm^3) ; actual (dm^3) / average recycling no.	470 ; 180 2,6														
	distillate (volume ; α activity ; PF**)	218 ℓ ; 4,4 10^{7} Bq ; 623														
	concentrate (volume ; α activity ; CF*)	3,3 ℓ ; 2,75 10^{10} Bq ; 72,6														
Decontaminated waste	weight (kg)	10,6														
	α activity [Bq (Ci) / Bq/kg (Ci/t)	4,8 10^{9} (0,130) 4,5 10^{8} (12,3)														
	DF***	10,5														
Slurry	weight (kg)	1,12														
	α activity [Bq (Ci) / Bq/kg (Ci/t)	9,2 10^{9} (0,250) 8,3 10^{9} (224)														
	DF***	1,45														

*CF = concentration factor - **PF = purification factor - ***DF = decontamination factor

TABLE V
Leaching of Alpha/Beta/Gamma Emitters

Waste feed	weight (kg)		20				
	α activity	Bq (Ci)	5,22 10^{11} (14,12)				
		Bq/kg (Ci/t)	2,61 10^{10} (706)				
	Irradiation	β, γ Gy/s (Rad/h)	2,8 10^{-6} to 3,6 10^{-6} (1 to 1,3)				
		neutrons Gy/s (mRad/h)	2,8 10^{-6} to 4,2 10^{-6} (10 to 15)				
Leaching no.			1	2	3	4	5
Leaching solution	HNO_3 (M)		2	2	2	4	0
	Volume (dm^3)		40	40	40	40	40
Time s (h)			10800 (3)	14400 (4)	10800 (3)	32400 (9)	25200 (7)
Desorbed α activity Bq (Ci)		^{244}Cm	2,1 10^{11} (5,7)	4,4 10^{10} (1,2)	1,1 10^{10} (0,3)	1,4 10^{9} (0,038)	3,7 10^{7} (0,001)
		^{241}Am	7,8 10^{10} (2,1)	2,4 10^{10} (0,66)	7,4 10^{9} (0,2)	1,1 10^{9} (0,03)	2,9 10^{8} (0,008)
		^{238}Pu	9,0 10^{10} (2,45)	2,8 10^{10} (0,77)	7,8 10^{10} (0,21)	1,1 10^{9} (0,03)	2,6 10^{8} (0,007)
		$^{239,240}Pu$	7,4 10^{9} (0,2)	2,3 10^{8} (0,063)	1,6 10^{9} (0,043)	3,3 10^{8} (0,009)	< 3,7 10^{8} (< 10 3)
		Total leaching	3,8 10^{11} (10,45)	9,9 10^{10} (2,693)	2,8 10^{10} (0,753)	3,9 10^{9} (0,107)	= 3,7 10^{8} (= 0,017)
		Total campaign	5,19 10^{11} (14,02)				
Decontaminated waste	weight after dewatering (kg)		17,5				
	α activity	Bq	3,8 10^{9} [1,5 10^{9} (^{238}Pu), 7,8 10^{8} ($^{239,240}Pu$), 8,9 10^{8} (^{241}Am), 6,3 10^{8} (^{244}Cm)				
		Bq/kg (Ci/t)	2,14 10^{8} (5,8)				
	DF		122 [77 (^{238}Pu), 14 ($^{239,240}Pu$), 110 (^{241}Am), 373 (^{244}Cm)				
	Irradiation	β, γ Gy/s (Rad/h)	2,8 10^{-8} to 5,6 10^{-8} (1 to 2.10^{-2})				
		neutrons Gy/s (mRad/h)	= 2,8 10^{-9} (= 1)				

Tables IV and V give the results of campaigns conducted in Elise and Prolixe, respectively, as examples of the data obtained with nitric acid leaching of waste.

Table V : Nitric acid leaching of a 20 kg batch of solid waste contaminated by alpha emitters (^{244}Cm, ^{241}Am, Pu) and beta/gamma emitters in Prolixe

Common observations can be made about the experiments conducted with the two types of waste: 1) after leaching, the leachates are not completely removed even after dewatering of the waste; about 4 to 5 dm^3 remain in the wet waste and further washing is necessary to obtain a high waste decontamination factor (DF) even in the case of quantitative transfer of the radioactive contaminant from the solid to the leaching solution. In this case $DF(\alpha) = 10^n$, where n = number of leaching + washing operations ; 2) a significant fraction (10 to 15%) is present in the form of fine particles, undoubtedly consisting of cellulose ; these particles are not retained by the filtering media of the washing reactor and accompany the leaching solution in the circulation loop. Before transferring the leachates to the concentration step, these "fines" must be recovered by high-efficiency filtering, which is a time-consuming operation.

Furthermore, in some ways the two types of waste reacted differently to the leaching. Although the kinetics of transfer of the alpha activity from the waste to the leaching solution is fairly rapid for the two types of waste, with equilibrium being reached after 600 s (10 mn) and 7200 s (2 h) for the plutonium-containing waste (Elise) and the waste contaminated by $^{244}CmO_2$ and $^{241}AmO_2$ (Prolixe), the transfer mechanism is surely different in each case. For the plutonium-containing waste of Elise, this mechanism is of the "liquid-liquid" extraction type due to the certain presence of TBP, which impregnates the waste. For the Prolixe waste, however, it may be considered that virtually all of the radioactive contaminant has been transferred to the leaching solution during the first leaching operation, since the following ones only desorb the fraction of radioactive contaminants retained in the wet waste. Significantly, $DF(\alpha)$ is close to 120 for the Prolixe waste and only 10 for the Elise plutonium-containing waste. It should nevertheless be recalled that :

- the waste was initially highly contaminated by alpha emitters (4.8×10^9 Bq/kg for Elise and 2.6×10^{10} Bq/kg for Prolixe) ;
- the residual contamination specification ($\leq 3.7 \times 10^6$ Bq/kg) applied to an encapsulated waste package ; a mass dilution factor of 6.6 is due to for encapsulation in epoxy resins ;
- the residual contamination specification applies to a package that has been aged for three centuries, so that the contamination by some radioactive contaminants such as ^{244}Cm (t1/2 = 18.1 years) or ^{238}Pu (t1/2 = 87.7 years) will have decreased substantially.

In view of these observations, it may be noted that the goal of declassifying the medium-level alpha or alpha/beta/gamma radioactive waste by nitric acid leaching is indeed feasible. This goal could be more easily achieved by oxidizing leaching with AgII.

AgII Oxidizing Leaching

Figure 6 shows a diagram of the AgII electrolysis unit currently installed in the Prolixe facility. A similar unit will be installed in the Elise facility in 1990.

This unit consists of a Zircaloy 2 vessel which contains:

- a Zircaloy heat removal coil ;
- a platinum anode ;
- a mechanical agitator ;
- a cathode compartment separated from the anode compartment by a sintered aluminum silicate diaphragm ;

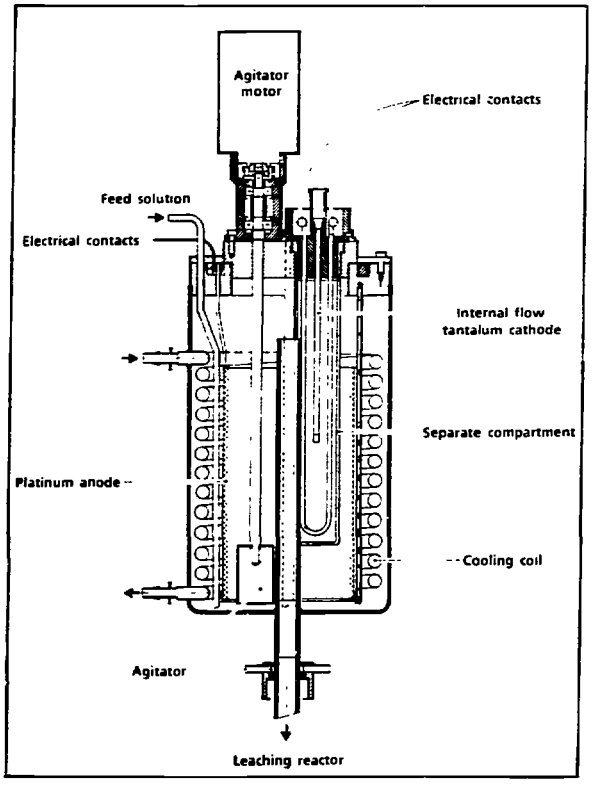


Fig. 6. Electrolysis Unit Employed to Generate AgII.

- a hollow tantalum cathode supplied with an internal flow cooling water.

The feed solution enters the electrolysis unit through a pipe integral with the lid and the AgII solution exits through an overflow pipe. Maximum steady-state production of AgII without waste is about 1.8 moles/h with a current of 100 A. Figure 7 gives several curves showing the production kinetics of AgII. An AgI-AgII conversion rate constantly less than 100 % is achieved because AgII is an unstable reagent that is reduced by water. A steady-state concentration of AgII close to 2.5×10^{-2} M is observed (for A_{gT} = 0.1 M, I = 100 A, θ = 296 K), which therefore corresponds to a conversion rate of 25 %.

The first waste leaching campaign using the AgII oxidizing method started in December 1989. This campaign involves treatment of a batch of metal waste mainly consisting of stainless steel highly contaminated by alpha/beta/gamma emitters from dismantling of a shielded hot cell formerly used for spent fuel examination (RM2 cell of the Nuclear Research Center at Fontenay-aux-Roses). The weight of the waste batch is about 24 kg; the required agitation of the leaching solution in contact with the waste is carried out by nitrogen bubbling through perforated nozzles placed in the reactor, unlike for the crushed waste. The results of this experiment were obtained in January 1990.

Excellent performance characteristics were expected, since the oxidizing technique for the metal waste already proved effective during development of a process for decontaminating stainless steel PuO_2 containers that will be used in the Melox mixed oxide (UO_2/PuO_2) fuel fabrication plant under construction on Cogema's Marcoule site in the Rhone Valley of France.

The stainless steel containers have the following dimensions : dia. = 0.085 m, h = 0.215 m ; the weight of their Z6CN18-10 austenitic steel is approximately 0.42 kg. The average plutonium quantity in each container is approximately 4×10^{-4} kg, which yields an alpha contamination level of about 2.2×10^{10} Bq/kg (600 Ci/t).

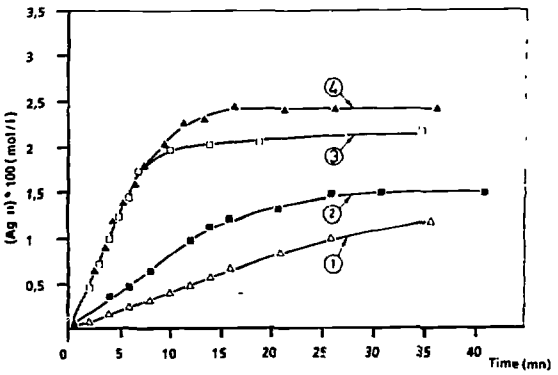
The oxidizing treatment using electrolytically generated AgII includes spraying the inside and outside surfaces of the containers with nitric acid jet solutions containing AgII. The AgII is generated in an electrolysis unit located outside the washing vessel.

Table VI gives the results obtained during treatment of seven containers. The decontamination factors ($DF\alpha$) achieved are close to or greater than 10^4, which gives a residual alpha activity of less than 6.7×10^5 Bq/kg (18 mCi/t) for the containers before packaging. These performance characteristics are especially high considering that the result is obtained over a short period of time (2700 s or 45 mn) and that the radioactive contaminant (Pu) is in a nitric acid medium and can thus be recovered by a simple process, e.g. TBP extraction or chromatography with ion exchange resins.

Drying

After leaching and washing, the waste is dried by circulation of hot air. This operation is performed inside the leaching reactor in Elise, whereas a separate reactor is used in Prolixe. The waste cannot be dried unless it is stirred; the drying of a 10 kg batch of waste therefore requires about 21,600 s (6 h).

The vapor contained in the wet air is condensed before heating of the air and recycling in the reactor. it is necessary to ensure that the water rinsing step has been completed before the waste drying operation is initiated. During two operating incidents in Elise, when the drying was carried out for waste impregnated with a nitric acid solution, the start of an exothermal reaction was observed along with partial "carbonization" of the waste and melting of the polypropylene bag. For obvious safety reasons, the waste washing and product effluent acidity verification step is mandatory.



I = 10 A (Curve 1), 20A (Curve 2), 100 A (Curve 4)
Anolyte = HNO_3 = 4M; $AgNO_3$ = 0.1 M; Vol = 12 dm^3; temperature = 23°C

Fig. 7. Kinetics of Electrolytic AgII Generation in a Nitric Acid Medium.

TABLE VI
Decontamination of Stainless Steel Containers Contaminated by PuO_2 Using Electrolytically Generated AgII

CONTAINER		OXIDIZING TREATMENT		RESULTS			
N°	Weight Pu 10^{-3} kg	Current (A)	Time s (h)	Weight loss 10^{-3} kg	Desorbed α activity Bq (mCi)	Final α activity Bq/kg(Ci/t)	D F (α)
1	0,205	30	7200 (2)	7,2	$2,0\ 10^9$ (54,05)	$6,6\ 10^5$ $(17,8.10^{-3})$	$7,2.10^3$
2	0,130	50	3 x 900 = 2700 (0,75)	5,7	$1,43\ 10^9$ (38,7)	$4,7\ 10^4$ $(1,28.10^{-3})$	$7,2.10^4$
3	0,155	50	2700 (0,75)	3,6	$1,56\ 10^9$ (42,34)	$1,7\ 10^5$ $(4,5.10^{-3})$	$2,24.10^4$
4	0,155	50	2700 (0,75)	3,5	$1,0\ 10^9$ (27,03)	$1,3\ 10^5$ $(3,43.10^{-3})$	$1,9.10^4$
5	0,225	50	2700 (0,75)	4,1	$1,53\ 10^9$ (41,4)	$4,2\ 10^5$ $(11,3.10^{-3})$	$8,74.10^3$
6	0,195	50	2700 (0,75)	2	$1,33\ 10^9$ (36,03)	$2,5\ 10^5$ $(6,86.10^{-3})$	$1,25.10^4$
7 *	0,255	30	25200 (7)	28,8	$3,1\ 10^9$ (83,79)	$1,5\ 10^5$ $(3,99.10^{-3})$	$5,3.10^4$

* Container used for preliminary design study to specify operating conditions.

Fig. 8. Cyclone Evaporator with a Capacity of 2.77×10^{-6} m^3/s (10 l/h).

Encapsulation

Waste encapsulation after decontamination will mainly be implemented using epoxy resins made by Ciba-Geigy. It will be carried out in containers with a total volume of $0.22Zm^3$ (Elise) or 1 m^3 (Prolixe) lined with a 2 to 3×10^{-2} m epoxy resin coating providing added protection for the encapsulated waste monolith. The package is obtained by preparing successive layers of encapsulated waste, a technique that enables suitable epoxy resin waste encapsulation while avoiding difficulties associated, for example, with the use of a mixing technique.

Inactive Elise-type waste packages with a 50 kg capacity have been produced for a total package weight of about 330 kg ; no active waste packages have been produced to date. An application for certification of Elise-type waste packages is being reviewed by the organization in charge of nuclear waste management in France, the French National Radioactive Waste Management Agency (Andra).

Leachate Recycling

After leaching, most of the solution must be recycled for subsequent treatment of another batch of waste. a special device was designed for this purpose using the thermosiphon phenomenon and the cyclone effect. This was necessary because only such a device proved suitable for concentrating effluents that had become highly reactive due to intense reactions between HNO3 and dissolved reducing organic substances originating from the waste, which produced nitrous vapors (NO_x) and generated large quantities of foam.

This highly compact device, which has a total heating power of 8.2 kW provided by five electric heating elements, has an evaporation capacity of 2.77×10^{-6} m^3/s (10 l/h). The purification factors for the distillates produced, even during concentration of particularly reactive foam-generating effluents, are approximately 2×10^5.

OTHER INDUSTRIAL APPLICATIONS

The process described for decontamination of waste prior to permanent storage enables the recovery of fissile material from by-products or waste containing valuable materials. Two facilities have been designed for this purpose to meet the requirements of Cogema. Their operating results will supplement previous experience and will also contribute to future advances in the treatment of alpha waste.

Dissolution unit in the T4 facility at the La Hague UP3 plant.

This unit has been included in the process flow design of Cogema's new reprocessing plant. It is used to dissolve batches of non-conforming plutonium dioxide, analytical samples and dust recovered from filters, in order to recycle plutonium.

The unit was designed specifically for the treatment of powders. It was put into active service at the end of 1989.

Prototype ash treatment unit at the Marcoule UP1 plant.

In this installation the leaching process is used to recover plutonium contained in ashes from an excess air incinerator that treats various types of solid waste from the reprocessing plant.

The unit is a full-scale prototype capable of meeting all reprocessing plant requirements. Initial testing has been completed and active operation is scheduled for early 1990.

CONCLUSIONS

The oxidizing dissolution technique has undergone spectacular development in France since the initiation of studies in 1981. It has been routinely employed for six years at Fontenay-aux-Roses with the production of ^{241}Am, which requires dissolution of aged PuO_2 inventories, and is deployed on an industrial scale at Cogema's UP3 reprocessing plant. In addition, a plutonium-containing ash treatment facility using oxidizing dissolution with electrolytically generated AgII will be put into active service at Cogema's UP1 reprocessing plant in early 1990. For the first time it has been demonstrated during treatment of stainless steel PuO_2

containers that waste could be declassified using a simple process, offering good promise for the future performance of the Elise and Prolixe facilities.

In recent years, since active commissioning of these cells, further expertise has been gained in many process phases. Examples of this progress include :

- crushing of 3600 kg of plutonium-containing waste (Elise) and 960 kg of alpha/beta/gamma waste (Prolixe) ;
- performance of about 30 leaching campaigns that enable transfer of 1.85×10^{13} Bq (about 500 Ci) of solid waste alpha emitters to the leaching solutions ;
- ample demonstration of leachate recyclability through the use of cyclone concentrators.

The goal of declassifying waste by electrolytically generated AgII in Prolixe and Elise should be achieved in 1990. An effort will also be made in 1990 to determine the chemical methods required to recover actinides from concentrated leachates and recycle silver.

Most of the information collected during development of the waste leaching process in Elise and Prolixe was helpful for designing an industrial facility for treatment of solid waste contaminated by alpha/beta/gamma emitters. This facility, the LEDA shielded hot cell installed in the Atalante building of the Rhone Valley Nuclear Research Center in France, is scheduled for active service before the end of 1990.

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