FR8501792

Symposium on radioactive waste management 85
Tucson, AZ (USA) 24-28 Mar 1985
CEA-CONF--7814

NUCLEAR RICH ALPHA CELLULOSIC WASTE MANAGEMENT EXPERIMENTS BY ACID DIGESTION

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CEN-CADARACHE - FRANCE

ABSTRACT

At Cadarache, where the french plutonium fuel fabrication plant is located, the strategy used for the management of rich alpha waste (superior to accepted level for storage) consist in incinerating the wastes, crushed and washed by cryogenic crushing and soda-nitric solutions.
Although all "technological" wastes could be processed this way, the cellulosic are sorted and treated separately by the sulfuric acid digestion process.
This process has definite advantages, particularly since it is specific to cellulosis, which dissolves easily at low temperature, i-e under the boiling point of H_2SO_4. Except for this aspect, of great importance for the gaz treatment operations and the resistance of material to corrosion, the process is identical to the one given in the litterature : dehydration of cellulosis by H_2SO_4 72 % and carbon oxydation by HNO_3 13 N.
The apparatus used hold in a small volume (10 m^3) ; the gloves-box in which the dissolver and the filtration treatments (insoluble Pu sulfate for one part, and reaction gaz for the other) are placed is in stainless steel coated with corrosion proof paint; the equipments are made of glass (dissolver) teflon (flanges) PVDF (pipes) hastelloy (pompes).
A general balance is given for the recuperated nuclear materials, as well as for the mass and volumes of input and output cellulosic wastes.

INTRODUCTION

A description is given of the operational facility and process used to reprocess cellulosic waste material produced in the Cadreche plutonium fuel fabrication plant.

CELLULOSIC WASTES

This term covers tissues and cotton wools used to clean process equipment, especially inside glove boxes. They thus contain nuclear material in significant amounts in the form of oxides or salts (nitrates, oxalates) : their average plutonium content is on the order of 100 grams per 100 liter drum of waste material, i.e. four times higher than for other wastes to be processed. The cellulosic wastes are therefore segregates at the point of use in order to be submitted to specific treatment.

PROCESS SELECTION PROCEDURE

The process selection criteria included the following :
- efficient recovery of nuclear material,
- easy and reliable implementation inside radioactive containment barriers,
- minimal production of radioactive liquid wastes or further solid wastes.

Operating experience over several years with a leaching facility using nitric acid, detergents and sodium hydroxide was made difficult with poor efficiency attributable in particular to problem of filtering cellulosic sludge.

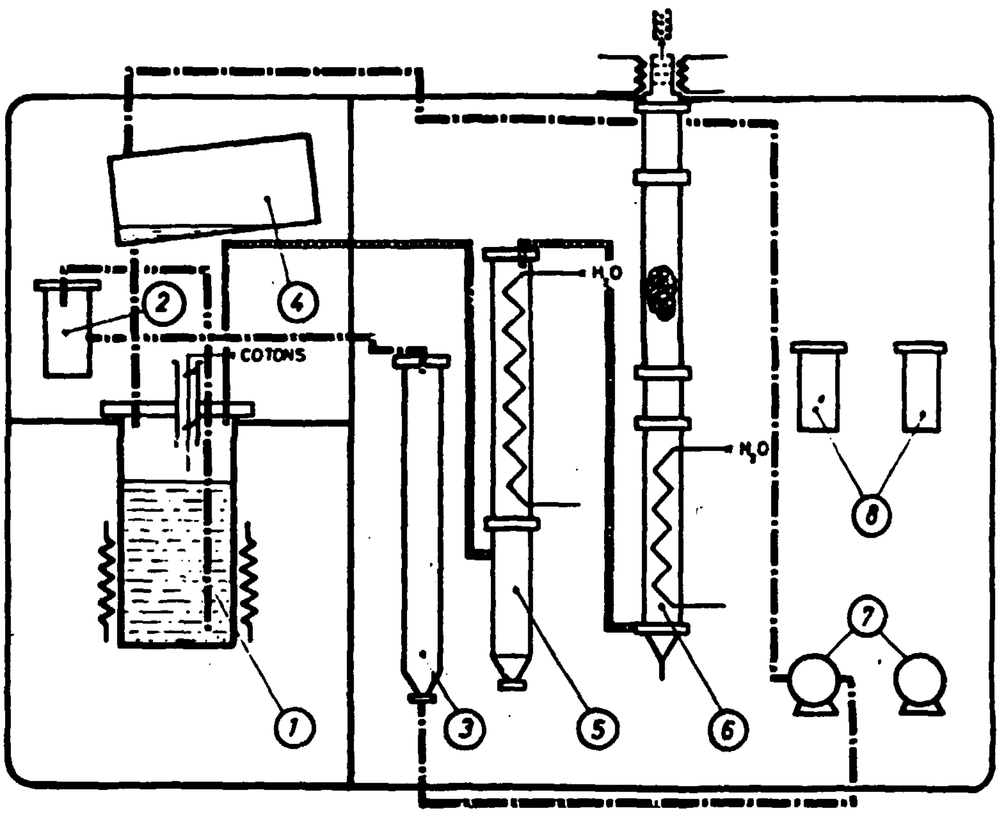
Incineration was eliminated from the outset because of its inherent drawbacks : generation of fumes rich in PuO_2 requiring reprocessing of exhaust filters, production of a highly refractory oxide, contaminating ash and tars.

However, a process based on sulfuric acid dehydration followed by nitric acid oxidation offers a number of advantages :

- virtually all of the plutonium is recoverable, since the plutonium sulfate formed by the process is insoluble in H_2SO_4, but is easily recovered in HNO_3 after filtration.
- The H_2SO_4 can be recycled to reduce consumption.
- The process is implemented at a temperature below the H_2SO_4 boiling point (unlike digestion process for other types of wastes).
- gaseous effluents are produced from a liquid medium, and therefore have low contamination levels.

Corrosion is therefore not a problem too difficult to solve, and a simple conventional gas filtration system can be installed.

The process thus presents numerous advantages and no major drawbacks.



SCHEMATIC OF ACID DIGESTION SYSTEM

active solutions
gazeous effluents

DESCRIPTION OF THE FACILITY AND PROCESS CYCLE

The facility currently in operation is not a true industrial unit (it handles only small batches introduced manually), but the throughout capacity is sufficient for all of the cellulosic wastes produced at the Cadarache plutonium fuel fabrication complex.

The entire facility is contained inside a three compartment stainless steel glove box, as shown in the following schematic view.

The wastes, removed from analyzed vinyl bags, are introduced into the dissolver (1) through an access lock. The dissolver is a 25 liter glass cylinder equipped with an immersion heater, a thermocouple and a compressed air stirring device, and contains 72 % sulfuric acid at 110° C. The carbonization reaction is instantaneous : a few liters of cotton are introduced in this way.

Then, using a metering pump, several 20 cc doses of 13 N nitric acid are introduced at 110° C. The carbon is eliminated and the nitrous fumes released are visible in the glass tube that carries them to the condensing column (5) and the neutralizing column (6).

Given the dissolver capacity, it was necessary to adjust the cellulose batch volume and the nitric acid doses because of the abundant foaming produced by nitric acid recovery of the finely divided carbon.

The third step is to eliminate the water produced in the reaction by raising the temperature to 170° C, leaving the initial sulfuric acid bath together with the plutonium sulfate which deposits out on the bottom of the dissolver.

These three steps are repeated as often as necessary, up to a maximum of 1 kg of plutonium for criticality and safety reasons.

The plutonium sulfate is extracted by vacuum removal of the solution from the dissolver into vessel (3) across PTFE tissue filter pockets (2). The filtered solution is recycled into the dissolver via the buffer tank (4).

The following process effluents are produced :

- <u>liquids</u> : sulfuric acid after maximum recycling, and activated water condensates. These wastes are discharged by means of electromagnetic pump (7) across 0,2 µm filters (8).
- <u>gazes</u> : CO_2, N_2, NO, H_2O. These gases are filtered across the standard glove box filtration system.

RECOVERY EFFICIENCY

Analytical uncertainties on the plutonium content of the initial waste material make it difficult to specify accurate figures : the Pu contents are indicated within ± 10 % by neutron counting and gamma spectrometry.

A typical process balance is shown below for one containing five bags coming from fabrication glove-boxes, with mixed-oxydes charges :

Volume of cotton processed : approximately 5 liters

- Pu mass taken into account	: 104 g
- Pu mass (non soluble) recovered in filters	: 87,2g
- Pu mass (soluble)in recycle H_2SO_4	: 4,4g
- Pu mass in condensats *	: ≤ 0,1g
- Balance + rétention	: 12,6g

* The volume of effluent of very low activity (washing of gaz) is about one hundred liters.