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FR 8801740
CEA-CONF-- 9127

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Communication présentée à : Conference on nuclear fuel reprocessing and waste management (RECOD 87)
Paris (France)
23-28 Aug 1987

CONFERENCE "RETRAITEMENT DU COMBUSTIBLE NUCLEAIRE ET LA GESTION DES DECHETS"
- PARIS 24-28 aout 1987 -

PROLIXE - PROTOTYPE REPROCESSING UNIT FOR IRRADIATING WASTES CONTAMINATED WITH ALPHA EMITTERS

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Abstract:

A large number of hot cells are employed by the Process Research Service in the Radiochemistry building of the Fontenay-aux-Roses Nuclear Research Centre, for research on nuclear fuel reprocessing and the production of isotope of transuranium elements. These activities generate solid wastes highly contaminated with alpha, beta, gamma emitters. To comply with the new recommandations concerning radioactive waste management. The Prolixe hot cell was built in order to : 1/ reprocess the solid wastes contaminated with alpha, beta, gamma emitters produced in the Radiochemistry building ; 2/ produce package wastes storable in shallow-ground disposal sites (< 0,1 Ci.t-1 in alpha emitters) ; 3/ develop a process sufficiently flexible to make it applicable to waste produced in other installations. The process to be applied in Prolixe is based on the technique of waste leaching after grinding. Depending on the type of wastes to be processed, the leaching reactant will have a diferent composition 1/ nitric acid solution for cellulose wastes : 2/ nitric solutions containing Ag(II) for other material. The complete process should achieve : 1/ a high waste volume reduction factor : 2/ the production of immobilized waste packages storage in shallow-ground disposal sites : 3/ the recycling of transuranium elements by chemical treatment of the concentrated leachates : 4/ the generation of a minimal volume of effluents. This process can be considered as an alternative process to incineration for the reprocessing of solid wastes highly contaminated with alpha, beta, gamma emitters.

I - INTRODUCTION

The "Service d'Etudes de Procédés" operates twelve hot cells in the radiochemistry building of the Fontenay-aux-Roses Nuclear Research Center. The main programs running in these hot cells deal with :

1/ The studies of reprocessing of nuclear fuels : light water reactor fuels (conventionnal and MOX fuels) and fast breeder reactor fuels.

2/ The production of transuranium isotopes carried out - by processing radioactive effluents produced in other facilities (^{241}Am) - by processing irradiated targets (^{243}Am, ^{244}Cm) - by treating old stocks of PuO_2 (for ^{241}Am production).

Essentially all the chemistry is based on hydrometallurgical operation units : dissolution of fuels or targets in aqueous solutions, liquid-liquid extraction, extraction chromatography, etc... During these studies and productions, solid wastes produced are contaminated by radioactive isotopes : fission products (beta, gamma), transuranium isotopes (alpha emitters).

Regulations concerning the management of the (alpha, beta, gamma) contaminated solid wastes have changed in the recent past, thus to solve the alpha, beta, gamma waste problem it was decided to build a facility with the objective of producing decontaminated wastes suitable for final surface disposal (i.e. the final alpha contamination will be ≤ 0.1 Ci.t^{-1}), and that of

recovering transuranium isotopes for reutilization. The new facility named PROLIXE (Prototype of Lixiviation and Embedding) actually built will run a process based on the leaching of the wastes with specific leachants. Laboratory studies proved that an overall decontamination factor ranging between 20 to 30 will be attainable for alpha emitters.

The present paper will describe briefly the PROLIXE hot cell and the leaching process which can be considered as an alternative to the incineration process. This process will be applicable to the management of other solid wastes produced in other facilities.

II - ALPHA, BETA, GAMMA ACTIVE SOLID WASTES

The alpha, beta, gamma active solid wastes consist in 70 % (in mass) of organic materials and 30 % of metals and glasses. The organic materials comprise : polyethylene, polyvinyl chloride, cellulosic materials, rubber and elastomers. These materials are contaminated by various radioactive nuclides : fission products (main isotopes ^{147}Pm ; $^{144}Ce/^{144}Pr$; ^{141}Ce ; ^{134}Cs-^{137}Cs ; ^{125}Sb ; $^{106}Ru/^{106}Rh$; $^{95}Zr/^{95}Nb$; $^{90}Sr/^{90}Y$; ^{155}Eu), and transuranium isotopes : ^{237}Np, ^{238} to ^{242}Pu, ^{241}Am, ^{243}Am, ^{244}Cm, ^{252}Cf.

The level of alpha contamination of these wastes has been estimated at 1 $Cixt \cdot l^{-1}$ while the fission products provoke a gamma dose rate of 21 $Radxh \cdot l^{-1}$ (mean value) for a typical "La Calhène" bin loaded with the active wastes. Approximatively 600 "La Calhène" bins loaded with alpha, beta, gamma active solid wastes are produced each year in the radiochemistry building.

III - PROLIXE HOT CELL FACILITY

III-1 GOALS TO ACHIEVE

The main goals to achieve in operating the PROLIXE HOT CELL FACILITY are :

1/ To reduce the volume of the waste by a factor as large as possible.
2/ To decontaminate the alpha, beta, gamma active solid wastes produced in the radiochemistry building.
3/ To obtain conditionned wastes in a form of a drum suitable for transportation and final surface disposal (i.e. alpha contamination will be $< 0.1\ Cixt \cdot l^{-1}$).
4/ To recycle the recovered isotopes especially for transuranium elements.
5/ To generate a minimum volume of effluents.
6/ To develop a leaching and embedding process sufficiently versatile to be able to process active solid wastes produced in other facilities.

III-2 PROLIXE : GENERAL DESCRIPTION

The PROLIXE hot cell comprises six different zones : four zones with alpha containment and two zones for work under irradiation. The alpha zones are protected by a 15 cm lead shielding and the two gamma zones are protected by a 10 cm thick steel shielding. The different zones have lead glass windows and nine working stations are equipped with master slave manipulators (MA 11/80 and MT 200 "La Calhène" types). The PROLIXE hot cell can receive alpha, beta, gamma active solid wastes contained in "La Calhène" bins directly from the PETRUS hot-cell via an underground tunnel and from the other hot-cells with the aid of shielded transfert casks. The first alpha containment serves for the reception of the wastes. In the second alpha containment the wastes are classified according to their nature : metallics, glasses, organic materials (cellulosics, other organics), then the organic materials are crushed.

The third alpha containment serves for the leaching and drying of the crushed wastes. After drying, these decontaminated wastes are transferred into the fourth alpha containment to be embedded in a large drum. The drum containing the embedded wastes is then closed in the first gamma zone, then in the second gamma zone final irradiation and contamination controls are operated before the drum is taken out of the hot cell.

III-3 PROCESS DESCRIPTION

CLASSIFYING - After being introduced into the hot cell the wastes are sorted out according to their nature, form, ability to be crushed, ability to be leached by nitric acid. For example plastics, glass, cellulosic materials can be crushed and washed with nitric acid solutions ; large plastic or stainless steel pieces can be washed by nitric acid but cannot be crushed. Other wastes cannot be washed by nitric acid solutions : for example for electric cables and other metallic pieces other aqueous solutions must be used to remove contamination. The larger part of the wastes can be crushed : it represents more than 35 % of the total mass of the wastes.

CRUSHING - The crushable wastes (organics, glass, electric cables) are crushed with or without the "La Calhène" polyethylene bins in the ROTAC 44136 crusher made by AUREC S.A. (FONTAINEBLEAU, FRANCE). The crusher is equipped with two disc-blades mounted on two parallel driving shafts. The rotation speed is 66 r.p.m. The driving shafts are drawn by a 7,5 kw electric motor. A stainless steel basket containing a pocket filter is placed under the crusher to receive the crushed wastes. A volume reduction factor of ten can be obtained and the crushed particles are only a few millimeters long.

LEACHING - The pocket filter containing about 22 liters of crushed wastes is introduced in the leaching reactor, then the reactor is closed and the wastes are leached with 50 liters of nitric acid. For the major part of the wastes, the leachant also contains silver nitrate and it circulates in a loop passing through an electrolyser to generate Ag(II) ions in order to dissolve refractory plutonium dioxide traces contained in the wastes.

During the leaching period the wastes are agitated in the basket to improve the leaching efficiency. Laboratory results demonstrate that decontamination factors of 20 to 30 are obtained for alpha emitters : the efficiency of the decontamination for beta gamma emitters is higher. At the end of the leaching step, the nitric acid solutions loaded with radioactive isotopes are separated from the wastes and stored in special tanks before performing their concentration by distillation. The distillate can be reused for another decontamination experiment while concentrated solutions (5 liters) will be transferred towards the PETRUS hot-cell for transuranium isotopes recoveries and purification.

In the case of non crushed wastes, decontamination will be obtained under the action of nitric acid solutions or high pressure water streams.

SCRUBBING - After the leaching step the decontaminated wastes are then subjected to scrubbing in order to remove traces of active solutions and to neutralize the nitric acid in the wet wastes. After scrubbing with a slightly alcaline solution, the wastes are rinsed with water until a pH close to 7 is obtained. These scrubbing solutions can be reused for several treatments, then they will be concentrated by distillation.

DRYING - The wet wastes are subjected to a drying operation under the action of warm air, water is then condensed by passing the air through a condenser.

EMBEDDING - The decontaminated dry wastes are introduced in a large container of $1 m^3$ of internal volume then a mixture of CIBA GEIGY epoxy resin and sand is introduced in the container to immobilize the waste. Other sorts of matrix will be usable : cements or mixed matrix made of cement-epoxy resin.

In the case of the use of epoxy resin the temperature during the polymerization will not reach 100°C if the neutralization step of the wastes is correctly carried out. After filling-up the container a layer of pure epoxy resin will be prepared in order to maintain the top of the container free of contamination.

CONTROLS - The filled container is then submitted to contamination and irradiation control before being conveyed from the hot cell to the surface site disposal. Figure 1 represents all the operations performed in a treatment campain.

III-4 EXPECTED PERFORMANCES

On the basis of laboratory and cold test results, one can expect the following performances :

1/ Total volume reduction : an overall factor of 8 to 9 is expected.

2/ Overall decontamination factor : a D.F. of 20 to 30 is expected for alpha emitters ; for beta, gamma emitters the D.F. will be higher.

3/ The conditionned wastes will meet the requirements for surface site disposal (0.1 Cixt^{-1} in alpha emitters).

4/ 10 liters of active solution is expected to be produced for the treatment of 234 liters of alpha, beta, gamma active wastes. The major part of the solutions will be reused.

IV - CONCLUSIONS

A new hot cell facility named PROLIXE was recently built in the radiochemistry building of CEN-FAR Nuclear Research Center in order to process alpha, beta and gamma active solid wastes produced while reprocessing studies were being made and transuranium isotopes produced. To treat these active wastes a leaching process was designed ; it is based in particular on the use of nitric acid leachant with Ag(II) ion electrogeneration. The expected performances of the process can compete with those of the more popular incineration process. Actually the construction of the hot-cell is finished and cold tests are under way. Active runs will be started in few months.

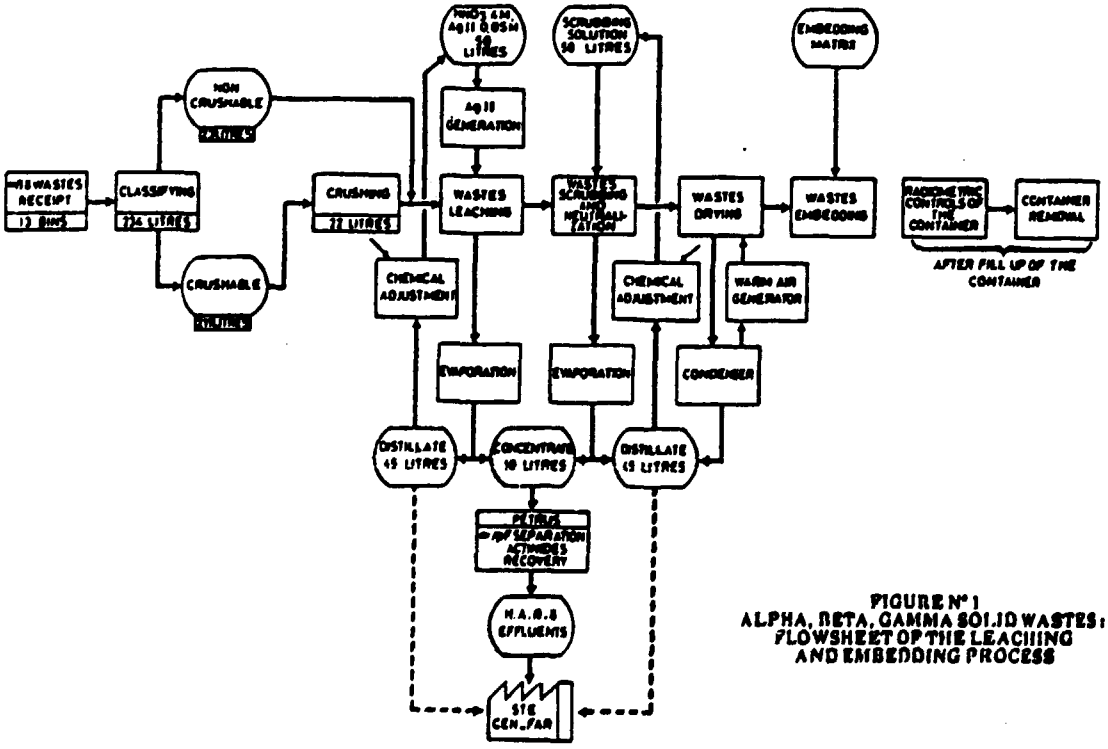

FIGURE N° 1
ALPHA, BETA, GAMMA SOLID WASTES : FLOWSHEET OF THE LEACHING AND EMBEDDING PROCESS