Setting an optimal procedure for transfering intermediate radioactive effluents : contribution of laboratory experiments – 15234

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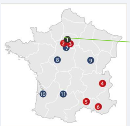
Introduction



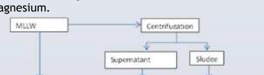
The CEA Saclay site, research center, includes a Liquid Waste Treatment Plant named facility INB 35.
In this plant, historical Medium Level Liquid Wastes (MLLW) are stored in tanks. This storage tanks were no longer in conformity.
Furthermore, salts precipitated into the tank over time, and so made the transfer difficult.
The objective of the project is to transfert these MLLW to a new suitable storage, or at least the source term present.

Preliminary studies : characterize the complex MLLW

Objectives of the Laboratory :

1. Radiologically and chemically characterize the complex MLLW with a heavy salts content ;
2. Carry out tests to help plan the MLLW transfer operations.

The radiological characterization compaign results showed that the source term, particularly α activity, was mainly present in the solid phase effluent (Fig. 1). The chemically characterization of the sludge showed that it was constituted by salts of phosphate, calcium and magnesium.



Gross Alpha activity	100%	1%	99%
Gross Beta activity	100%	35%	65%

Fig.1: Distributions of the gross alpha and beta activities

Further to this results in order to retrieve as much as possible of the source term, and given the issues related to the liquid waste particle extraction from the tanks (due to their sedimentation), it appeared that dissolution of MLLW solid phase salts by acidification would facilitate the operations. The operation rules permit the addition of HNO_3 acid at a maximum concentration of 6N to the tanks, while maintaining a pH ≥ 2.

Dissolution studies were carried out, adding variable quantities of 6N nitric acid to MLLW. The surpernatant pH, activitiy and composition in major elements were monitored :

- Addition HNO_3 acid 6N (initial pH of MLLW ≈ 7) enabled the percentage of unsolvables (sludge) to decrease by a factor 2 as from pH ≈ 2 (Fig. 2) with an increase of the gross alpha activity in the supernatant of the same order of magnitude ;
- The sludge dissolution implied an increase in the concentrations of magnesium, calcium and phosphate in the supernatant (Fig. 3) ;
- In order to check for the lack of particles > 1 mm, filtration test on 1 mm qrid was carried out on MLLW. No clogging was observed.

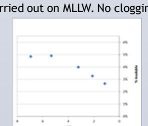


Fig.2: Unsolvables (sludge) changing percentage depending on pH

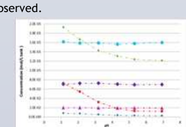


Fig.3: Supernatant composition evolution depending on pH

On the basis of this information it was decides that before any emptying operation, a treatment of MLLW would be carried out under stirring with HNO_3 6N at a pH ≈ 2, to obtain a maximum dissolution of unsolvables and thus be able to remove the source term by pumping.

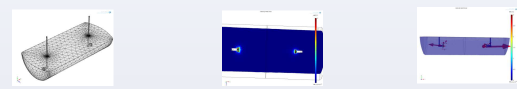
Initial state of the tanks

5 horizontal tanks in a semi-underground pit
200 m³ of MLLW to transfer
3 m diameter and 7,5 m length, 2 manholes
A compressed air stirring system no longer worked and emptying rods were clogged
pH>2 for the tank leaktightness
Sedimentation of the radioactive liquid waste

Chemical engineering

Understand and optimise :

- The homogenization of the effluent
- The pumping through a strainer wich would limit the diameter of particles (salts) transferred to the tanker



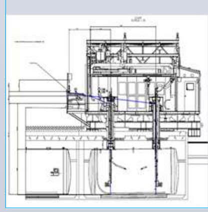
Results of the studies :

- There was a localized homogeneization of the liquid around the periphery of the stirrers, because of the diameter of the submerged stirrer action zones ;
- A particle agglomeration phenomenon occured on the strainer when high-speed stirring was running.

A homogenization scenario was defined in order to lift as much sludge as possible, to enable its dissolution by the nitric acid (duration of mixing, place of the stirrers, …).

Transfer MLLW in a new storage workshop

The mobile workshop



A mobile workshop which incorporates containement features, with two barriers associated with a suitable ventilation system.
Its fonctions :

- Tank stirring with submerged devices
- Sample taking
- Treatment to dissolve the salts
- Removal into a tanker.

The MLLW analyses showed that as the nitric acid 6N was injected and the pH decreased, the concentrations of phosphate, calcium and magnesium increased (Fig. 4) in conformity with what had been recorded in the laboratory. The total alpha and beta indexes also increased, whereas the gamma activity (mainly ^{137}Cs) remained constant (Fig. 5). This data was also in agreement with the information obtained from the laboratory.

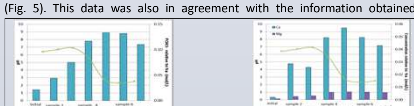


Fig.4: Evolution of the phosphate, magnesium and calcium concentrations as the acid was injected

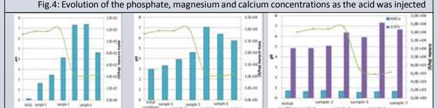


Fig.5: Evolution of the radiological activity as the acid was injected

The radiological and chemical characterizations carried out during the MLLW tank treatments with HNO_3 6N before emptying showed that the procedures implemented to solubilize the solids present in the concentrates performed well.

The process developed for the retrieval of MLLW in these storage tanks enabled most of the term source present. The material remaining after the retrieval operations represents about 2.5 % of the total initial effluent volume. At the end, 10 to 15 cm of sludge and a few liters of liquid were still present in the tank. Concerning the source term, the treatment implemented enabled the retrieval of 95 % of the beta-gamma activity, and 75 % of the alpha activity.

The collaboration between the analytical laboratory and the dismantling project was a real advantage.
Thanks to this tight collaboration, the duration of the transfer was reduced by half, compared to the first ones.
Calling on assistance from chemical engineering modeling enabled the phenomena related to the circulation of liquid within the tank to be understood. From this, the practises required for the optimization of stirring and pumping conditions were able to be deduced.
The MLLW retrieved, once treated chemically, will be immobilized in a cementary matrix. Consequently, a compromise must be found between the amounts of liquid added compared to the source term removed, as well as the direct immobilization of the sludge material during the clean-up phase.

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A special thank to the SPIE team which allowed the realization of these operations.