CONDITIONING EXPERIMENTS OF LIQUID SCINTILLATION COCKTAILS USED FOR TRITIUM ANALYSES

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ABSTRACT

This paper describes the results of a liquid scintillation cocktails conditioning method. This radioactive waste type is generated by the radio-chemical analysis lab of a CANDU nuclear power plant.

Liquid scintillation cocktails are converted into a stable monolithic form, which minimizes the probability to release radionuclides in the environment during their interim storage, transportation and final disposal.

INTRODUCTION

Conditioning of liquid scintillation cocktail is carried out by cementation using aluminium stearate additive. Portland cement is the most common type of hydraulic cement and it is the original agent utilized for the solidification of low-level radioactive waste [1].

EXPERIMENTAL PROCEDURE

Liquid scintillation cocktails characterization

Liquid scintillation cocktails are Ultima Gold type. They are solutions containing:

- solvents: di-isopropyl naphthalene (DIN); ethoxylated alkylphenol; mono/di-phosphate ester; sodium di-octylsulphosuccinate;
- scintillators: 2,5-diphenyloxazole (PPO); 1,4-bis(2-methylstyryl)benzene(bis-MSB).

This waste contains varying amounts of tritium with activity below 1.00E+08Bq/l and small quantities of Nb-95, Zr-95, Sb-124, Cr-51.

Characterization of conditioning matrix components

Portland cement 42.5R

- chemical characteristics: SO_3 - 2.95%;
  Cl^- - 0.0071%;
- physical characteristics: setting of concrete time - 10800 s;
  stability - $5\cdot10^{-4}$ m;
- mechanical resistance: after 2 days - $25.5\cdot10^{6}$ N/m^2.

Aluminium stearate

- composition: stearic acid aluminium salt;
- physical and chemical properties:
  form - solid;
  colour - white;
  odour - odourless;
  melting temperature - $(120\ldots130)^0$C;
  solubility in ethanol at 20^0C - soluble.

Mechanical resistance verification of conditioning matrix

The verification is performed on the prism-test specimens, having $40\cdot10^{-3}$ m length, $40\cdot10^{-3}$ m width, $160\cdot10^{-3}$ m height, after 28 days from the preparing of the test specimens. During this period, the test specimens are preserved in plastic bags. An uniaxial compressive load is applied along the axis of the prism-test specimen until it is crushed. The maximum load is divided by the cross-sectional area of the specimen to determine its compressive strength.

Two prism-test specimens were tested with the composition and mechanical resistances shown in Table I.

Table I. Composition and Mechanical Resistance of the Samples

Code sample	Composition, wt.%				Mechanical Resistance [N/m^2]
	Waste	Aluminium Stearate	Cement 42.5R	Water	
A	7.54	4.71	63.10	24.65	$97\cdot10^3$
B	8.43	4.67	62.60	24.30	$88\cdot10^3$

The two values of mechanical resistance are higher than the required Waste Acceptance Criteria of disposal site (Baita Bihor National Repository), which is of $50\cdot10^3$ N/m^2 for the minimal disposal value.

A mechanical resistance higher value offers security for the final disposal of conditioned waste.

The water/cement ratio is a very important factor that affects the porosity of the hardened cement paste. The water/cement optimal ratio is of 0.39.

Long-term leach testing of conditioning matrix

Leachability is a parameter that refers to the release rate of radioactive species from the waste form as a result of interaction with water. The release rate and mechanism involved are highly dependent upon environmental conditioning including temperature, as well as pH of the leachant.

The test method requires the waste form specimen to be cylindrical, and the value of the ratio leachant volume/exposed surface area of the specimen to be of (0.1…0.2) m. The method uses demineralized water as a leachant. The test is performed at a temperature of 20^{0}C. The leachant is sampled and completely replaced with fresh leachant on the following schedule:

- initially, after 1,4,7, and 10 days;
- then once in fourth week;
- once per month during the following three months;
- then twice per year for as long as is considered necessary.

The result of the test is expressed as Eq.1, after the leachant sample is analyzed to measure tritium activity content.

$$R_n = \frac{a_n V}{A_0 S t_n} \qquad \text{(Eq.1)}$$

where:

R_n = release rate of radioactive species [cm/day];
a_n = activity of a given radionuclide released during leaching interval n, [Bq];
V = volume of the specimen, [cm^3];
A_0 = total activity of the radionuclide in the specimen at the beginning of the leach test, [Bq];
S = exposed surface area of the specimen, [cm^2];
t_n = duration of the leachant renewal period, [days].

Two cylindrical-test specimens were tested, having the composition shown in Table I. The leachant sample was analyzed by a Liquid Scintillation Counter to measure tritium activity content.

The leach rate of tritium from the two cylindrical-test specimens is presented in Fig.1.

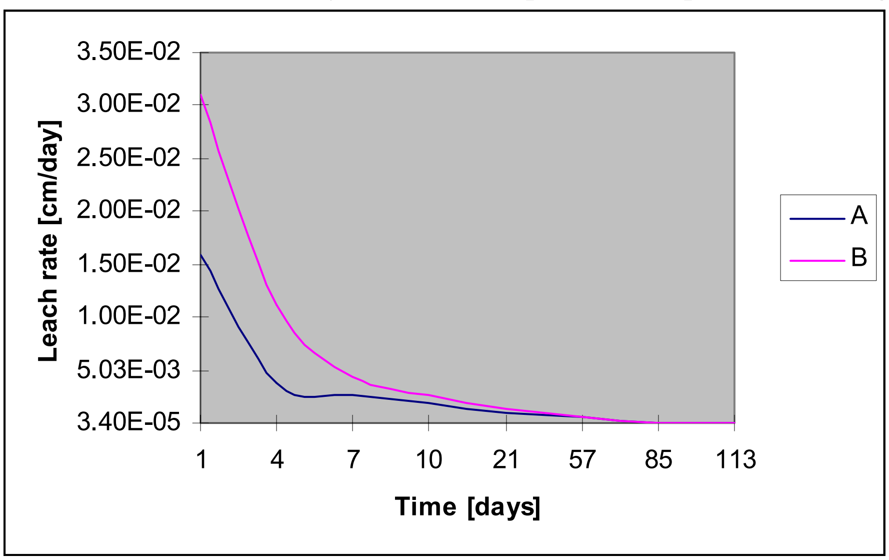

Fig. 1 The leach rate of tritium from the two cylindrical-test specimens.

The value of the leach rate after 113 days has a magnitude order of 0.034·10^{-3} cm/day, for A matrix, and 0.050·10^{-3} cm/day for B matrix, representing a low rate of tritium release from the waste form as a result of interaction with water. These values are much smaller than the maximal value required by Baita Bihor National Repository, which is 10^{-3} cm/day after one year.

CONCLUSIONS

Conditioning of liquid scintillation cocktails contaminated with tritium into a cement and aluminium stearate matrix may be carried out using the follow conditioning matrix, wt%:

- waste	8.43;
- aluminium stearate	4.67;
- water	24.30;
- cement	62.60.

This method is simple, with low cost of working materials. The operator exposure is negligible because there is not any vapor problem. The higher values of mechanical resistance and the low rate of tritium released from the waste form offer security for transportation and final disposal.

REFERENCE

1 "Conditioning of low and intermediate level radioactive waste", Technical Reports Series no. 222, International Atomic Energy Agency, Vienna, 1994.