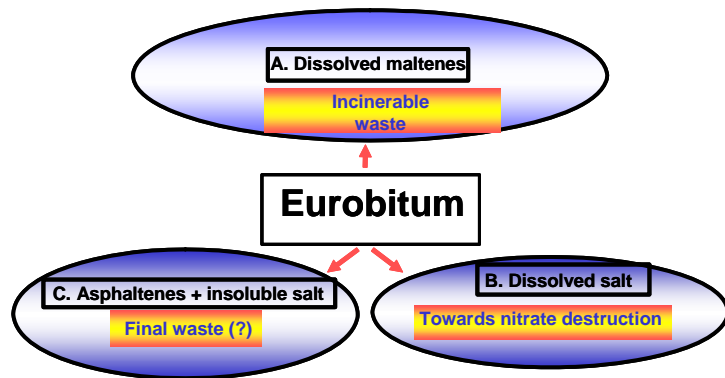


Background

Whereas in many countries the reference concept for the long-term management of intermediate-level bituminised radioactive waste is final disposal in a deep underground repository in a stable geological formation, the international interest in alternative solutions is growing. At SCK•CEN, a research project has been finalised on the possible alternatives to re-treat so-called 'homogeneous' bituminised waste such as Eurobitum. Eurobitum waste contains mixtures of nuclear fuel decladding slurries and waste concentrates from EUROCHEMIC, the former European research reactor fuel processing factory at Dessel, Belgium. It contains about 30% of NaNO₃. The waste is mixed with blown bitumen (type R85/40) in a 40:60 ratio. This type of bitumen contains about 80% of maltenes and 20% of asphaltenes.

One way to re-treat this type of waste would be plasma-incineration. Preliminary results showed that a very stable final vitrified waste can be obtained comparable to the stability of R7-T7 reference waste glass, and that the waste volume would be reduced to 75% of the original volume. The major disadvantages of this re-treatment technique is the high-tech and high cost plasma installation needed and the safety aspects related to the higher radioactivity content of this waste type.

The technique proposed in this paper is based on the dissolution of the bitumen in an organic solvent and the subsequent extraction of nitrates in water leading to the separation of (1) an organic effluent containing the maltenes, (2) an aqueous effluent containing the nitrates and (3) the final waste containing the asphaltene fraction and water insoluble salts including most of the radionuclides. This separation scheme together with the possible treatment of the secondary effluents is visualised on the right.



Room temperature separation technique applied to Eurobitum. An organic (A) and an aqueous (B) fraction could be further treated at Belgoprocess, whereas the fraction C is the final stable waste to be pressed and placed in a drum for final storage

Objectives

This paper describes the lab-scale results of a room temperature separation technique applied to real radioactive Eurobitum samples, sampled from a drum that was produced in 1981.

Principal results

We applied the separation technique described in the main reference. The maltenes and water soluble salts are separated from the Eurobitum successively. The maltenes can be easily removed from Eurobitum taking into account the difference in solubility between the maltenes and the asphaltenes using organic solvents with a flash point above 60°C. Once the maltenes are removed from the Eurobitum waste, the water soluble salts can be removed using water. By performing the separations in this order, the best separation results are obtained without risk of emulsification. This resulted in 3 waste fractions (A, B and C on the scheme above). Mass balance checks were not performed with the radioactive samples but this was done with non-radioactive simulants (in main reference), with satisfactory results.

Fraction A is an organic solvent mixture containing the maltenes with a flash point above 60°C. Four tests with different solvent compositions have

Gamma activity in Bq/g				
Maltene effluent number	Co-60	Cs-137	Am-241	Eu-154
1b	18	1308	81	7,1
2b	4	213	15	<1,3
3b	7	410	29	2,7
4b	5	189	15	1,6

γ activity of the organic effluent A

been performed. The results are in the table above and below. The uncertainty on the analytical results is 30%. Experiments 2, 3 and 4 show that the alpha and gamma activities are below the limits of the operating

licences of CILVA at Belgoprocess. The amount of this effluent produced would be between 8 and 24 g per gram Eurobitum re-treated. Purification prior to incineration could be optimised.

Alfa activity in Bq/mL			
Maltene effluent number	Pu-239+Pu-240	Pu-238+Am-241	Cm-243+Cm-244
1b	15	80	2
2b	6	23	0
3b	6	31	1
4b	6	25	1

α activity of the organic effluents(A)

Fraction B, the aqueous effluent, has also been analysed (table below). These effluents contain high concentrations of NaNO₃, but the activities of Co-60 and Cs-137 are acceptable under current water purification exploitation licenses if small amounts (some tens of m³) would be produced, and the alpha activity is below the detection limits. Per gram of Eurobitum re-treated, about 24 grams of water are used. If large amounts of Eurobitum were to be re-treated, this aqueous effluent should be purified prior to the normal aqueous waste re-treatment methods. In any case, a nitrate destruction method should be applied to convert the nitrate into N₂. Various reviews on nitrate destruction applied to radioactive waste are available in international literature. These could serve as a basis to select and test the most appropriate technique.

Gamma activity in Bq/g			Alfa activity in Bq/g	
Aqueous effluent nr.	Co-60 (Bq/g)	Cs-137 (Bq/g)	Aqueous effluent nr.	total alfa
1a	138	17,3	1a	< 0.4
2a	100	31,1	2a	< 0.2
3a	59,2	15,1	3a	< 0.5
4a	132	17,3	4a	< 0.3

Activity of the aqueous effluents (B)

Fraction C contains the asphaltenes and the insoluble salts. Fraction C represents 30% of the original mass of Eurobitum waste and can be easily pressed. Several lab tests simulating an industrial 2000-ton press showed that the volume of the waste was reduced with a factor 4 (without taking into account the emptied drums and the re-treatment installation). Due to some practical reasons, only one preliminary standard leach test has been performed, and therefore, only the order of magnitude of the result is relevant. The leach velocity of the final pressed waste is (only) about a factor 10 higher than the R7-T7 reference waste glass.

The results are generally in line with the expectations based on the experiments with non-radioactive Eurobitum simulates. Differences can be explained by the different compositions and synthesis methods of real Eurobitum samples and non-radioactive simulates.

The fractionation of the waste at room temperature prior to final waste re-treatment enhances the safety of the complete re-treatment. In this way, the maltenes can be safely incinerated, free from large amounts of inorganic substances and with reduced radioactivity. Also the separation of the nitrate salts into a α-poor aqueous fraction enhances the safety of the nitrate destruction. Furthermore, the final waste is almost free of leachable compounds, and is made of an asphaltene matrix containing the final radioactive waste. It has been shown in literature that the asphaltenes have affinity to complexate ionic substances. Furthermore, their poly-aromatic structure and high C:H ratio makes them quite resistant to radiolysis. Therefore, it is expected that the asphaltenes would serve as a good matrix for final disposal.

Future work

The robustness of the technique could be tested using other types of bituminised waste, such as other Eurobitum batches, or bituminised waste from France (AREVA - COGEMA) or Japan (JAEA). Other topics to be studied are the coupling with the existing radioactive effluent treatment methods, the selection and testing of a nitrate destruction technique, the gradual up-scaling, the nuclearisation, the secondary waste, the eventual re-use of the steel drums, a detailed and complete cost estimation, the public acceptance and comparison with other options.

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