

Babylon: R&D program for French Bituminized Waste Products acceptability in deep geological repository

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General context

The reprocessing of nuclear spent fuel yields some aqueous nuclear waste streams that are processed by using inorganic reagents to co-precipitate radionuclides, leading to a co-precipitation sludge. In France, such as Belgium or Japan, bitumen is chosen for the conditioning of this co-precipitation sludge (Figure 1), producing some Bituminized Waste Products (BWPs). During this conditioning step, bitumen and sludge are co-extruded at a temperature close to 160°C, leading to the evaporation of the water initially present in the sludge, to the precipitation within the bitumen matrix of soluble inorganic salts and to the embedding of insoluble inorganic salts. The resulting mixture, composed of a mix of inorganic salts and of bitumen in a weight ratio ranging from 20/80 up to 40/60, is then poured into a steel drum of about 220 L, where it freely cools.

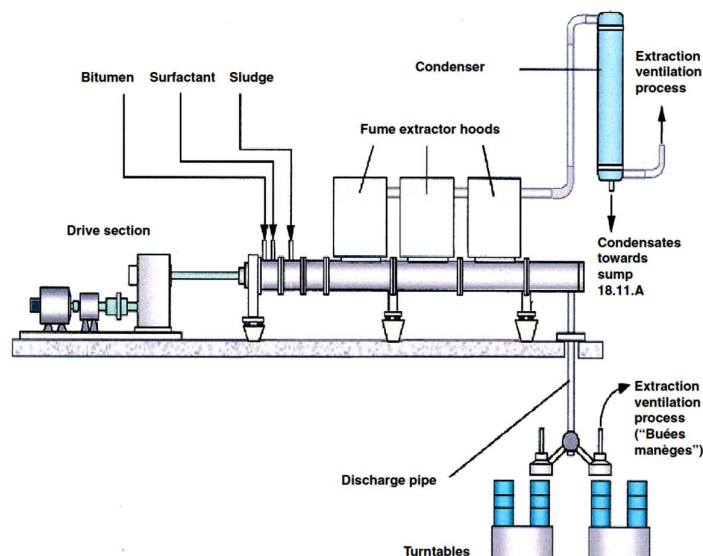


Figure 1: Example of a bituminization industrial-scale process in an extruder [1]

Since the 1960s in France, around 75,000 drums of Bituminized Waste Products (BWPs) have been produced. About 80 % of them are stored at Marcoule, and the remaining 20 % at La Hague. A few dozen and around a hundred of drums are currently produced per year at both sites respectively. Currently, among this inventory, all the drums stored at La Hague and almost half of those stored at Marcoule are intermediate-level and long-lived (ILW-LL) categorized, while the other half are low-level long-

lived waste (LLW-LL). ILW-LL are intended to be disposed in the Cigéo facility, which is the reference disposal solution for ILW-LL in France. In the scenario described in the Cigéo safety options file and as far as BWPs are concerned, 220 Litres drums will be packaged in CS4 concrete container (Figure 2). The resulting packages will then be brought and disposed in the Cigéo deep geological disposal.

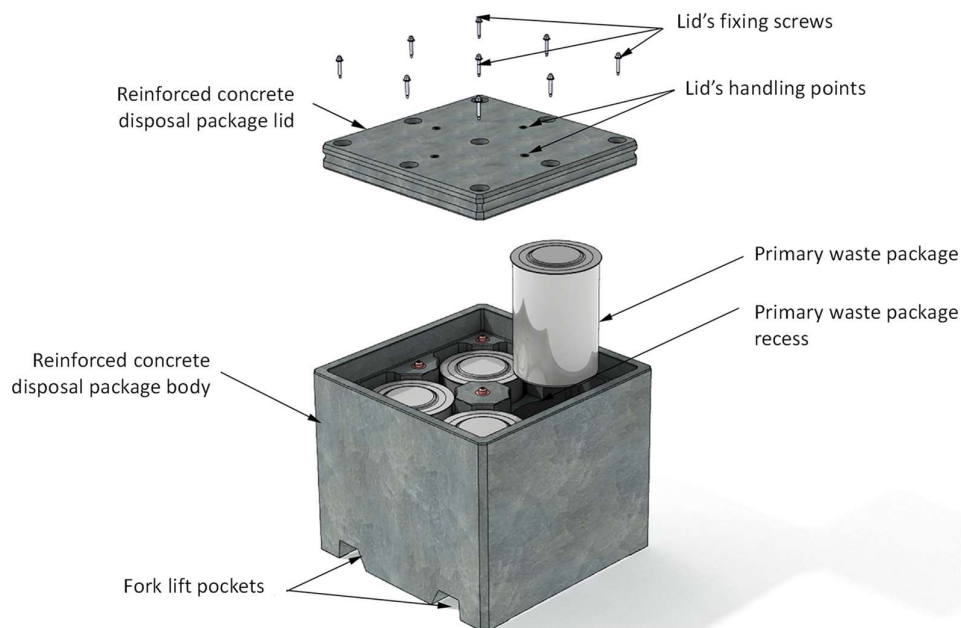


Figure 2: CS4 concrete package of four 220 L drums for direct disposal in CIGEO

Knowledge concerning the description and the simulation of BWPs package behaviour in such a surrounding, and concerning consequences on this surrounding, is a key point for ILW-LL BWPs acceptance in Cigéo. Related issues can be divided into three main topics.

The first one deals with **the intrinsic evolution over time of BWPs**, especially resulting from their **self-irradiation**.

Interaction between ionizing radiations from immobilized radionuclides initially present in co-precipitation sludge and the bitumen matrix leads to an organic bond breakage. The macroscopic consequences of this phenomenon are di-hydrogen gas production and an increasing in BWPs viscosity, both due to radiolytic products recombination. Bitumen and BWPs can be described as a fluid showing a high viscosity at ambient temperature. H_2 gas bubbles that are produced during BWPs irradiation are then trapped within the drums, leading to a swelling which depends on bubbles evacuation rates. Such a phenomenon was described and modelled, in order to simulate BWPs drums behaviour during their self-irradiation (H_2 source-term and release, matrix radiolytic ageing) [2] [3]. It has to be noted that BWPs produced at La Hague incorporate in their formulation a cobalt sulfur getter that limits, even annihilates such a swelling phenomenon.

The second one is associated with the **thermal behaviour of BWPs**, especially if a thermal stress due to a fire outside the container occurs.

From the beginning of BWPs production and depending on the reagents used during the coprecipitation step, six composition domains were identified: 4 for BWPs produced at Marcoule and 2 for BWPs produced at La Hague. When applying a thermal stress to BWPs, exothermic reactions can occur between incorporated salts and even between the bitumen matrix itself and salts such as sodium nitrate. Such an exothermic process was quantified over the whole BWPs composition domains, by measuring the energy resulting from these reactions and the associated temperature ranges. From these experiments, it was reported that nitrates are the main contributors to the exothermic reactions and that a

solicitation temperature lower than 150°C should not lead to significant energy release. Previous gram scale experiments were further supported by kilogram scale experiments. During latter experiments, 2 kg of BWPs mean compositions were heated up 300°C during 6 hours with neither BWPs measured overheating nor inflammation. Finally, at the concrete container scale, fire test showed that the concrete container kept its mechanical integrity, allowing its handling by using travelling crane. Moreover the design of concrete container was improved after fire test results in order to guarantee a temperature under 100 °C inner the container in external fire situation.

The latter is related to the **BWPs long-term behaviour** in presence of water within disposal cells. Once water will resaturate disposal cells, a water uptake phenomenon by BWPs will occur, leading to their swelling and to the leaching out of their salts. As far as salts leaching out is concerned, analyses demonstrated that no consequence regarding safety should be expected. The other issue deals with the over-damage that would occur in the near-field of disposal cells, due to BWPs swelling. Very few data on French BWPs water uptake-induced swelling were available at the time impact analysis were carried out. ANDRA developed therefore a conservative approach consisting in applying the maximum theoretical pressure value on the internal of disposal cells. It came out from the analysis that the resulting damage should be limited, however highlighting the interest for a model simulating the mechanical behaviour of BWPs in these conditions.

To tackle these issues, Andra and the three nuclear licensees, CEA, EDF, Orano, led to the setting up of a R&D program, called Babylon, started in 2020 and will end in 2026. The objectives of this program are described below.

Objectives of Babylon R&D program and related adopted approach

To reinforce the safety demonstration and to establish margins in Cigéo design and exploitation, the ongoing Babylon R&D program aims at completing acquired knowledge by pursuing the development of modeling tools to simulate BWPs behaviour, taking into account the influence of their self-irradiation. The Babylon program is thus articulated into three main parts, described in the following.

BWPs thermal reactivity

Based on the previous BWPs classification into six families, the thermal reactivity among each composition domain for each family was assessed by relating BWPs composition at each point of the composition domain and thermal quantities such as energy related to exothermal reactions or reactions onset temperature, measured by using microcalorimetry. The first part of the program aims at completing such a description of BWPs thermal reactivity. To do so, the first step consists in assessing uncertainties associated to thermal quantities measurements, from the making of the sludge up to the interpretation of calorimetric measurements. The second step consists in defining envelope curves of chemical reactivity within each BWPs composition domains. The completion of the approach will be then pursued by studying particular chemical compositions that were not taken into account in previous studies and that resulted from the industrial treatment of specific effluents and from the incorporation of degraded organic products. Moreover, data acquisition of thermal reactivity on radioactive BWPs retrieval will support the previously described approach made on non-radioactive samples.

BWPs thermal behaviour

The second part of the program aims at simulating BWPs thermal behaviour under thermal stress, taking into account radiolytic ageing. The present model has been actually developed and compared to experimental results on non-irradiated BWPs. In the Babylon program, non radioactive BWPs will be externally irradiated at the kg scale in order to evaluate firstly the influence of the presence of H₂ gas bubbles, and secondly the influence of the radiolytic rheological ageing, on the BWPs thermal behaviour. Elaborated samples will be thermally stressed and the experimental collected data will be compared to numerical simulations in order to evaluate their accuracy. Such an accuracy evaluation will be supported by a numerical sensitivity study, taking into account refined BWPs physical characteristics.

Finally, drum scale tests with non-radioactive BWPs are planned. Drums will be heated up to about 180°C with temperature data acquisition. These experiments will enable to validate at real scale the model of BWPs thermal behaviour.

Long-term behaviour of BWPs in disposal cells

The third part of the program aims at quantifying margins arising from the comparison between the former conservative approach as developed previously and a more realistic approach taking into account BWPs behaviour during their leaching and its consequence on disposal cells.

To do so, the first step consists in collecting experimental data related to the leaching behaviour of BWPs in volume-constrained and in stress-constrained conditions. Based on these experimental results, on the previously collected data in free leaching conditions, and on the resulting phenomenological description, a hydro-mechanical model is under construction to simulate BWPs behaviour in such conditions, especially concerning the relation between the build-up pressure development and the corresponding strain. Such a model and the resulting simulations will be then taken into account to evaluate the consequences of BWPs swelling on the over-damage of the near-field disposal cells.

Conclusion

CEA, Orano, EDF and Andra have launched in 2020 a quadripartite R&D program for a period of six years. It is based on both an experimental and modelling approach. It aims at improving the robustness of the safety assessment of BWPs disposal. Indeed, the program will consolidate knowledge of the behaviour of BWPs in order to confirm the design and operation of the disposal cells.

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