



## 10.2 Belgian Research on Fusion Beryllium Waste

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Future fusion power plants will generate important quantities of neutron irradiated beryllium. Although recycling is the preferred management option for this waste, this may not be technically feasible for all of the beryllium, because of its radiological characteristics. Therefore, at SCK•CEN, we initiated a research programme aimed at studying aspects of the disposal of fusion beryllium, including waste characterisation, waste acceptance criteria, conditioning methods, and performance assessment. One of the main issues to be resolved is the development of fusion-specific waste acceptance criteria for surface or deep geological disposal, in particular with regard to the tritium content. In case disposal is the only solution, critical nuclides can be immobilised by conditioning the waste. As a first approach to immobilising beryllium waste, we investigated the vitrification of beryllium. Corrosion tests were performed on both metallic and vitrified beryllium to provide source data for performance assessment. Finally, a first step in performance assessment was undertaken.

### 1. INTRODUCTION

The time schedule for the development and construction of future fusion reactors foresees the construction of the first generation of commercial power plants based on nuclear fusion technology for the large-scale production of electricity towards the end of the 2040's. The development path towards commercial fusion power plants (FPP) will likely include the next-step device ITER, a technology demonstration reactor DEMO, and a commercial prototype reactor PROTO. An important factor in the public acceptance of fusion power will be the environmental and safety impact of these new plants, and in particular of commercial FPP. In future reactor models, beryllium is often considered as a plasma-facing component in the vacuum vessel and/or neutron multiplier in the breeding blanket. Recently, the European Power Plant Conceptual Study PPCS investigated the safety and environmental issues of a generic FPP based on a helium-cooled pebble bed (HCPB) blanket and a helium-cooled divertor [1]. Although in any recycling of the irradiated beryllium

should be the primary waste management option, not all of the beryllium can be recycled and reused after an interim storage of up to 100 years. For this waste, a solution could be final disposal in a surface repository or in a deep geological repository. At SCK•CEN, we started a research programme focusing on fusion beryllium waste and all aspects of its possible disposal, including waste acceptance criteria for surface and deep geological disposal, performance assessment calculations, conditioning methods, and beryllium corrosion studies in disposal environments. This paper presents an overview of the work performed until now.

### 2. WASTE ACCEPTANCE CRITERIA

#### 2.1. Fusion waste inventory

For the beryllium waste inventory issuing from a FPP we consider an interim storage period of 50-100 years, during which the radioactive elements decay to lower activities. As the basis for our analysis of waste acceptance criteria, we used waste inventories reported for the SEAFP-2 plant models [2]. These plant

models are all based on DT fusion in a steady state tokamak. The main features of the three plant models include a plasma major radius of 9.4 metres, a fusion power of 3000 MW, and an average neutron wall load of 2.1 MW/m<sup>2</sup>. The divertors are assumed to be replaced after

20 full power months of operation, and the blankets after 5 full power years. In their in-vessel features, the three power plant models differ only in the definition of the blankets, whose main features are summarised in Table 1.

Table 1. Main characteristics of the SEAFP-2 Plant Models.

<i>Plant Model</i>	<i>FW/Blanket structure</i>	<i>Tritium-generating material</i>	<i>Neutron multiplier</i>	<i>FW/blanket coolant</i>
1	Vanadium alloy*	Li <sub>2</sub> O ceramic pebble bed	None	Helium
2	Low activation martensitic steel**	Liquid Li <sub>17</sub> Pb <sub>83</sub>	Li <sub>17</sub> Pb <sub>83</sub>	Water
3	Low activation martensitic steel**	Li <sub>4</sub> SiO <sub>4</sub> ceramic pebble bed	Beryllium	Helium

\* The vanadium alloy is V-4Cr-4Ti. \*\* The low activation martensitic steel has a nominal composition 0.1C-9Cr-2W-0.2V-0.07Ta.

P. Rocco investigated the possibilities of recycling for the SEAFP-2 models, taking into account the contact dose and the decay heat after a decay time of 50 years [3]. Two categories of recyclable material are envisaged. The 'complex recycling material' has an upper contact dose rate of 20 mSv/h and requires remote handling recycling. 'Simple recycle material' has an upper contact dose rate limit of 2 mSv/h and may be recycled by hands-on operations. Waste with a contact dose rate higher than 20 mSv/h or a decay heat of more than 10 mW/m<sup>2</sup> cannot be recycled and is classified as permanent disposal waste. Rocco's study yielded 64 ton of beryllium waste for Plant Models 1 and 2, and 4930 ton per reactor for Plant Model 3 [3]. For each Plant Model, 22 ton of beryllium cannot be recycled and needs permanent disposal, either surface or geological disposal.

We based our beryllium waste inventory on SEAFP-2 plant model 2, which does not use beryllium as a neutron multiplier. The purpose of this exercise was to apply existing waste acceptance criteria (WAC) to verify whether the fusion waste would, under the existing, fission-related legislation, be acceptable for

near-surface disposal. To this purpose, we used the WAC for the El Cabril surface disposal site in Spain [4]. Without taking into account the chemotoxicity of beryllium, approximately 13.75 ton of first wall beryllium per reactor would be unacceptable for surface disposal. The main reason for this is the high amount of tritium present in the irradiated beryllium. Furthermore, a high level of <sup>10</sup>Be is present for which no maximum specific activity is mentioned in the El Cabril WAC. The conclusions to be drawn from these calculations are:

- the option of detritiation is in any case to be pursued to limit the amount of beryllium for deep geological disposal
- We need specific waste acceptance criteria for fusion waste, taking into account those radionuclides abundant in fusion waste and taking into account typical release rates for these nuclides.

## 2.2. Towards fusion-specific waste acceptance criteria: the case of tritium

Based on existing WAC, the tritium level in beryllium from fusion reactors would be too

high to be accepted for surface disposal. However, the existing WAC are derived for tritium in fission waste, which is mostly present in tritiated liquids. In the case of fusion waste however, the tritium is incorporated in solid metals. Therefore, the fission-specific WAC are not as such applicable to fusion waste. As a first step towards a thorough performance analysis yielding waste acceptance criteria for the surface disposal of irradiated beryllium components from fusion reactors, we considered the beryllium tiles from the first wall armour of the SEAFP-2 plant models. We will briefly describe the model for tritium release. Details can be found elsewhere [4].

The conceptual model for tritium release is based on a simplified corrosion model for beryllium in an alkaline environment. Assuming tritium is uniformly embedded in the lattice structure of beryllium, the molar ratio of tritium and beryllium can be estimated. Further assuming that the diffusion of tritium from the bulk of the beryllium metal to the pore water is negligible, and that no porous networks exist in the beryllium, the beryllium and tritium release will be based on a simple two-step corrosion model. Based on this corrosion model, the maximum beryllium and tritium flux from the conditioned waste to the disposal vault and finally towards an underlying aquifer can be assessed. For the purpose of demonstrating the general performance assessment methodology, we considered the generic Belgian concept for surface disposal of low level waste, as schematically shown in Figure 1. The result of our simplified calculations is that, in a first approach, the annual dose rates due to tritium in groundwater as a result of the surface disposal of fusion beryllium waste are negligible [4].

### 3. CONDITIONING METHODS

Currently, in Belgium, the management option for beryllium waste from materials test-

ing reactors is intermediate storage without prior conditioning. The beryllium is packed in stainless steel containers together with dry clean quartz sand. The steel canisters are stored in a ventilated storage building until a suitable conditioning method is found. In the framework of fusion waste too, conditioning methods could be used to immobilise radionuclides in irradiated beryllium and retard their release into the biosphere. As a contribution to the discussion on the treatment of beryllium waste from fusion reactors, we compared cementation, bituminisation, vitrification, and phosphatisation [6]. The criteria used were the long-term durability of the immobilising matrix, the technical feasibility (including existing industrial know how), and cost. As a reference point we also present the case of direct disposal of beryllium waste without prior conditioning.

#### 3.1. Cementation

Cementation is a proven technique for the conditioning of diverse waste streams [7]. It consists of immobilising waste in a cement matrix and has been applied during the last decades to low- and intermediate-level radioactive waste. The basis of many types of cement used for waste conditioning is ordinary Portland cement (OPC), but often additives such as fly ash and blast furnace slag, are incorporated in the cement matrix to improve its characteristics. Cementation is a simple, relatively inexpensive technique, resulting in a non-flammable matrix with good durability. The main drawback in the case of beryllium however, is the high pH (12,5) of the cement matrix, which will give rise to high corrosion rates. The associated production of H<sub>2</sub> could lead to gas-driven transport of radionuclides through the clay layer (considering geological disposal in a deep clay layer) [8].

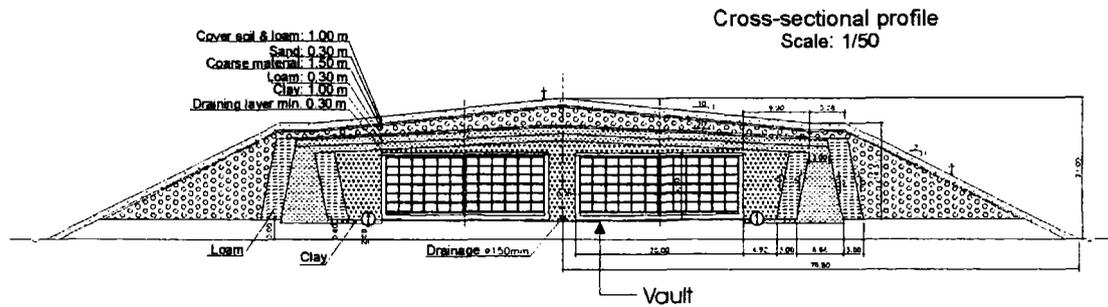


Figure 1. Cross-sectional view of generic concept for surface disposal of LLW in Belgium. One vault is 25m wide and 27m long and contains 936 container. (Source: NIRAS/ONDRAF [5])

### 3.2. Bituminisation

This process has been applied to the conditioning of low-level and intermediate level sludge, evaporator concentrates and ion-exchange resins [8], but never to beryllium. The bituminisation process is technically complex. The encapsulation of beryllium in bitumen could present problems. The beryllium from fusion reactors will probably be in the form of large metal pieces, and hot bitumen will rapidly harden at contact with the cold metallic pieces, preventing bitumen from completely filling the voids between the beryllium parts. As beryllium remains in its metallic form, corrosion and associated gas formation cannot be excluded. However, since bitumen does not impose extreme pH values, the corrosion rate will be lower than in cement, provided it is not disposed of in a cementitious backfill material.

### 3.3. Vitrification

Vitrification is currently applied for the conditioning of highly active fission product residues resulting from the reprocessing of spent fuel from the Belgian nuclear power plants [10]. One of the reference processes used today, consists of two steps, starting with the isolated fission products from the reprocessing plant, which are in the form of concentrated nitric acid solutions. First, the nitric solution of fission products is fed continuously to a rotating calciner, in which the solution is evaporated, and the residue dried and partially calcinated. The calcinated residue is then mixed with other glass constituents to reach the desired composition. It is subsequently heated, melted, and the liquid glass is then

poured into a canister [10]. A vitrification process for the beryllium waste from fusion reactors could be based on this type of process and consist of dissolving the beryllium in nitric acid, calcining, and mixing the thus obtained beryllium oxide with molten glass.

The use of glass as an immobilising matrix for the deep geological disposal of high-level radioactive waste in clay layers has been extensively investigated at SCK·CEN [11], including the characterisation of the vitrified waste forms [12]. The long-term behaviour of the beryllium in the glass matrix is very favourable. As beryllium is fully oxidised during the vitrification process, corrosion of beryllium and the associated hydrogen production will be suppressed during geological disposal. The vitrified waste is very resistant to leaching, with corrosion rates as low as 1  $\mu\text{m}/\text{year}$  [13].

### 3.4. Phosphatisation

Phosphate ceramics are currently developed at IPN Orsay (France), for the conditioning of uranium, plutonium, and neptunium. It is a wet process in which the dissolved species are mixed with phosphoric acid, evaporated, and sintered at 300-800 MPa and 1250°C [14]. These matrices, which integrate the radioelements in a mineral solid solution, show particularly low corrosion rates, about 0.003  $\mu\text{m}/\text{year}$  [14]. Beryllium might be a suitable material to be conditioned in phosphate. Indeed, Be may substitute for calcium on the  $\text{Ca}^{++}$  sites in Apatite ( $\text{Ca}_5(\text{PO}_4)_3(\text{F}, \text{Cl}, \text{OH})$ ). As beryllium and calcium are both alkaline earth metals, the synthesis of Be phosphates might then be possible.

### 3.5. Direct disposal

It is possible to consider the storage of beryllium without a conditioning matrix. In Belgium, beryllium was put in sealed containers with dry sand to fill the voids and an outside container to provide shielding. The technique used at SCK·CEN is described elsewhere [15]. The canisters are stored in a ventilated room to prevent tritium build-up by diffusion through the canister. This is probably the cheapest method, and it offers the advantage of easy recovery of the beryllium. The disadvantages of this method are the same as for encapsulation in cement or bitumen: gas generation through corrosion, and the toxicity of the beryllium metal.

### 4. VITRIFICATION OF BERYLLIUM

As a first step towards the demonstration of a conditioning method for beryllium, we incorporated a batch of beryllium pebbles in a glass matrix. The vitrification method consisted of three steps:

- (i) oxidation of beryllium powder at 900°C in air, yielding beryllium oxide powder
- (ii) primary glass melting: mixing of beryllium oxide powder with glass constituents and melting at 1250°C during three hours
- (iii) annealing: after cooling down, the beryllium glass is finally annealed at 500°C during three hours.

For the melt, we used the ingredients for a modified borosilicate glass, to which sodium and aluminium oxide were added. The result was a beryllium glass with a beryllium load of approximately 2 wt.%. Figure 2 shows a photograph of the beryllium glass. The next step in our research programme will be to investigate the microstructure of the produced beryllium glass. We cut the glass cylinders into thin disks that will be polished and investigated with Electron Micro Probe Analysis to map the distribution of elements (and in particular beryllium). Additionally, beryllium glass powder was produced to perform leach tests in water and clay water (see also section 5).

## 5. CORROSION TESTS

In order to predict the long-term behaviour of beryllium in a disposal environment, data are needed on its corrosion characteristics. In principle, for an exhaustive comparison between the various conditioning methods reviewed in section 3, corrosion tests should be performed on each waste form in its appropriate disposal environment. In a first phase however, we will restrict our corrosion research to metallic beryllium and vitrified beryllium.

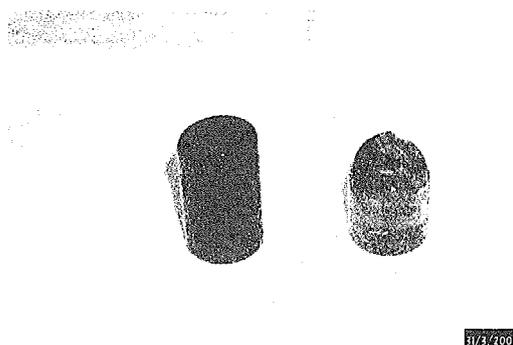
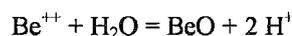


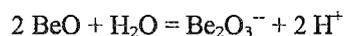
Figure 2. Vitrified beryllium.

### 5.1. Corrosion of metallic beryllium

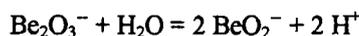
Pure metallic beryllium is highly corrosion-resistant in high-purity water at low temperatures. Typical corrosion rates are less than 25 µm/year [16]. Because metallic beryllium, due to its high affinity for oxygen, is almost always covered with an oxide or hydroxide layer, the behaviour of this film will determine the corrosion characteristics of beryllium [17]. At low pH, the solubility of the BeO film is determined by the reaction



In the lower pH range then, the solubility of BeO increases rapidly with acidity. In the high pH range, the solubility of the BeO film is determined by the reaction



In the higher pH range, the solubility of BeO increases with pH. At even higher pH values, beryllate ions are formed:



The solubility is at a minimum for near-neutral pH conditions. In a Boom clay environment, with pH values around 8.2, beryllium is passivated throughout the stability region of water. The corresponding general corrosion rate will therefore be very low.

Although the general corrosion rate of pure beryllium in pure water is negligibly low, *localised corrosion* can present a problem for commercial alloys containing impurities that are exposed to non-pure water. Small quantities of chloride and sulphate in the environment can have a strong impact on the corrosion rate of beryllium, by promoting pitting corrosion on surface impurity sites in the beryllium. Several factors influence the initiation and propagation rate of localised corrosion of beryllium alloys, including the presence of aggressive ions in the environment, metallurgical impurities, metallurgical history, and irradiation history. The presence of aggressive ions such as chloride in particular can initiate pitting corrosion. Static corrosion immersion tests of pure beryllium in demineralised water, and in demineralised water to which chloride, fluoride, or sulphate were added in small amounts, showed that corrosion products were present on the beryllium surface after an immersion time of only a few hours in a 5 ppm chloride solution [18]. In the context of a geological disposal in Boom Clay, we encounter chloride concentrations of approximately 27 ppm [19]. It is clear that the 'direct disposal' of metallic beryllium, which in the longer term would come into contact with the backfill of the disposal galleries, may cause corrosion problems. In the case of conditioning, the corrosion mechanism and rate will depend on the specific chemistry of the immobilising matrix. At present, there is a clear lack of quantitative data on the corrosion of beryllium, especially in slightly alkaline environments representative for geological disposal in clay layers. One goal of the current research programme is to provide such data.

## 5.2. Long-term corrosion tests

To provide quantitative data for the performance assessment of disposal of fusion beryllium waste, we initiated long-term corrosion tests in simulated disposal environments. These tests consist in immersing beryllium electrodes in Boom clay slurries and monitoring the corrosion potential over a period of at least two years. In additional tests, the potentiodynamic polarisation curves of beryllium in Boom clay slurries will be recorded, allowing to determine the critical potentials for localised corrosion:  $E_{np}$ , the critical pitting potential, above which new pits nucleate and grow, and  $E_{pp}$ , the repassivation potential, below which pits repassivate and above which pits continue to grow (but no new pits necessarily nucleate). Comparing the values of the corrosion potential with the values of the critical pitting potential and the repassivation potential, in combination with a knowledge of the corrosion kinetics, allows predicting the extent of localised corrosion. Furthermore, in the case localised corrosion can be excluded in the course of the disposal, the electrochemical tests allow us to predict the extent of general corrosion.

## 5.3. Glass corrosion tests

In order to provide quantitative data for the safety assessment of the disposal of vitrified beryllium waste, we will perform leaching tests of beryllium glass powder in clay water.

## 6. FIRST APPROACH TO PERFORMANCE ASSESSMENT IN GEOLOGICAL DISPOSAL

In section 2.2, the methodology for performance assessment in the case of surface disposal was presented as a step towards the development of fusion-specific waste acceptance criteria. In the current section, the case of deep geological disposal is discussed.

The chemo-toxic impact of beryllium waste on the biosphere was estimated using the Belgian reference deep geological repository concept. This concept is based on isolation of waste in a deep clay layer (Figure 3) and relies

on the multibarrier concept, in which both natural and engineered barriers (e.g. conditioning matrix, container...) contribute to the containment of the waste. In the Belgian disposal concept for high-level radioactive waste from fission reactors for example, waste containers containing the conditioned waste are placed in long tubular metallic overpacks in the centre of concrete-lined galleries which are backfilled with bentonite. For our calculations, we considered the clay layer as the only barrier against release of radionuclides, i.e. we did not take into account the retarding effect of the engineered barriers. The small fraction of long-lived radionuclides that escapes is diluted in the surrounding aquifers. The radiological impact was also roughly assessed by analogy with existing waste from fission.

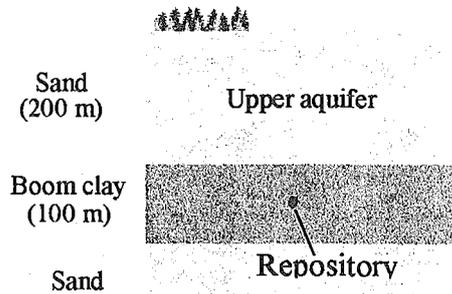


Figure 3. Schematic representation of geological disposal in a deep clay layer.

6.1. Chemical impact

The quantity of beryllium foreseen for future power production Tokamaks is about 64 ton per reactor in Plant Models 1 and 2 of the SEAFP-2 study [3]. Plant Model 3 considers a much higher beryllium quantity, 4930 ton per reactor, due to the use of beryllium as a neutron multiplier in the breeder blanket, but this is probably not realistic from the point of view of the production of such a large quantity of beryllium, and its economic aspects. To model the chemical toxic impact, we considered the disposal of 100 T Be, which roughly corresponds to the beryllium waste produced by one fusion reactor. To make a first rough estimation of beryllium release in the upper aquifer, we took several conservative assumptions on

the migration parameters: pore diffusion coefficient equal to  $2 \cdot 10^{-10} \text{ m}^2 \cdot \text{s}^{-1}$  (typical value for performance calculations in Boom clay), a retardation factor equal to 300 (typical for moderately sorbed elements), and a solubility limit equal to  $10^{-9} \text{ M}$  ( $\text{mol} \cdot \text{dm}^{-3}$ ). Those values are quite conservative, as beryllium is identified in the literature to strongly sorb on clay minerals, and to be very insoluble (geochemical models predict  $10^{-12} \text{ M}$  for beryllium solubility at pH around neutral).

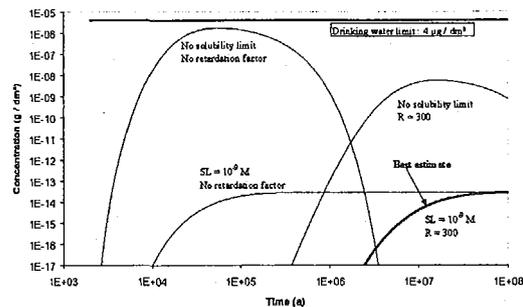


Figure 4. Concentration of beryllium in the upper aquifer remains under the limit for drinking water, even for the most conservative assumption.

The results of the migration model presented in Figure 4 describe the influence of the assumptions taken for the migration parameters on the beryllium concentration in the upper aquifer. It shows that the conservative value of  $10^{-9} \text{ M}$  for the solubility limit guarantees by far that beryllium won't be a problem in the aquifers. The only effect of no retardation (no sorption) is that the beryllium concentration reaches its plateau sooner. To see the effect of the solubility on the beryllium output, we performed the simulation without solubility limit. Even in this case, and with no retardation, beryllium concentrations remained under the health standard for drinking water set by the EPA [20]. We can conclude from those preliminary calculations that, even if the migration parameters for beryllium still need to be better studied, the disposal of several 100 tons of beryllium in the Boom clay should raise no concern about the possible chemical toxic impact on the biosphere.

## 6.2. Radiochemical impact

Beryllium waste from fusion is not only toxic, it is also radioactive, mainly due to neutron activation of impurities during its lifetime in a Tokamak reactor. Most of its activity comes from  $^3\text{H}$ , produced in beryllium neutron reactions. Given its short half-life, it will have totally decayed before coming out of the clay layer. However, the activation of impurities present in the beryllium might lead to the production of long-lived radionuclides (see section 3.2.2). Indeed, such radionuclides were found to be responsible for the long-term dose in the case of an intrusion scenario [21].

Most of activation products of impurities in the beryllium will rapidly decay to very low activities. By comparing the radiological inventory with some typical fission wastes, it appears that radioelements in beryllium waste have too little activity or too short half-lives to significantly influence the radiological impact of the repository on the biosphere (provided that the concentration of U in beryllium is minimised).

## 7. CONCLUSIONS

This paper describes the Belgian research programme on beryllium waste from fusion reactors. Although recycling is the preferred waste management option, we have hitherto focussed on the aspects of disposal, because for a limited amount of neutron-irradiated beryllium, this is the only viable option. In the fusion reactor plant models considered in the SEAFP-2 study, the first wall beryllium would not be acceptable for surface disposal, mainly due to the high tritium content. However, fusion-specific waste acceptance criteria need to be developed to derive more appropriate maximum activities for fusion-specific radionuclides. These waste acceptance criteria will have to be derived with performance assessment, for which quantitative data on the beryllium source term will be needed. To provide such data, we are currently running long-term corrosion tests.

## ACKNOWLEDGMENT

The authors wish to thank Ing. Steven Smets and Mr. Kris Penasse for their technical assistance. This work has been partly financed by the European Commission in the framework of the European Fusion Development Agreement. However, the content of the publication is the sole responsibility of its publishers and it does not necessarily represent the views of the Commission or its services.

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