



Public

Memo

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Treatment of silver in solubility calculations

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1 Introduction

This Memo describes the treatment of silver in the SR-Site solubility calculations and addresses an issue relating to the handling of Ag-108m in these calculations, raised by SSM in a request for additional information (SSM2011-2426-4.7.2.i).

The treatment of the solubility of Ag in SR-Site is described in SKB (2010, p 294); for a summary see section 2 below. The vast majority of the silver is found in the PWR control rods. These consist of an alloy of Ag-In-Cd, and a proportion of the silver is in the form of the activation product Ag-108m. The uneven distribution of Ag-108m in the control rods, which was not discussed in SR-Site, is addressed in section 3 below.

2 Treatment of silver in SR-Site: pessimistic immediate dissolution of all Ag

As is specified in SKB (2010, p 294), the control rod (Ag-Cd-In) alloy is pessimistically assumed to be immediately released to solution at the time of canister failure. In the scenarios involving early canister failure, such as shear due to an early earthquake, solubility limits are used in the calculation of solution composition inside the canister. The assumption made is that all silver (mostly non-radioactive) is, at once, released to solution; thus, the solubility limit is reached with regards to the solubility limiting phase. There is no need, in this scenario, to consider where in the control rod the activation products are located. It should be noted that the control rod alloy is covered by a ca 0.5 mm thick steel cladding. The protective effect of the cladding is pessimistically disregarded in the calculation, assuming the alloy is immediately available for dissolution.

The logic following this assumed scenario is that the amount of Ag-108m in solution corresponds to the ratio between Ag-108m and total amount of silver in an average canister. This ratio is about 1/250. This is the ratio used in the calculations.

The reason behind this pessimistic treatment is the lack of leaching data of control rods in relevant repository conditions.

Pessimistic, unrealistic assumptions vs. probable behaviour of the alloy in repository conditions: a discussion

The assumption of an immediate and complete dissolution of the control rods results in an unrealistic isotopic dilution of the radioactive silver, since its concentration is higher at the tip of the rod longitudinally and at the outer periphery of the rod where the neutron flux is higher. On the other hand, metallic silver is a noble metal and is not expected to be corroded in anoxic waters. The most reasonable corrosion mechanism for metallic silver would be through sulphide attack (McNeil and Little 1992). This would result in extremely low concentrations of dissolved silver. In order to estimate a corrosion rate for metallic silver, one would have to assume a certain flux of an oxidant. The most probable oxidants would be radiolytic ones, produced by the radiation field of the irradiated control rods. In this case only low LET gamma and beta radiations have to be considered. The oxidant production from low LET radiations in waters saturated with even low concentrations of hydrogen (as e.g. used in nuclear reactor water) is extremely limited (Pastina et al. 1999, Pastina and LaVerne 2001). A few percent H₂ is supposed to protect even steel details in the primary circuit of a nuclear reactor from corrosion.

However, no experimental data exist for silver corrosion under relevant conditions, and without data it is difficult to assume an oxidant production rate resulting in silver corrosion. For this reason SKB is preparing tests of control rod corrosion with regard to estimate silver releases. The tests concern un-

irradiated control rods in simulated groundwaters (to estimate de-alloying and its effects on potential silver releases) and irradiated control rods under inert atmosphere or low (10 % H₂ in Ar) concentrations of hydrogen.

The treatment of Ag corrosion in SR-Site is pessimistic in the extreme. The elements In and Cd may be released rapidly from the alloy, but far from instantaneously, through dealloying. The silver, however, is a noble metal in metallic form, and will corrode through sulphidation, if dissolved sulphide is available, to insoluble silver sulphide. A more realistic instant release fraction for silver from the control rods would be zero. It is the lack of relevant, quality assured experimental data which lies behind the unrealistic, pessimistic treatment of Ag corrosion in SR-Site.

3 Account of uneven distribution of Ag-108m in the control rods

It is highly pessimistic to assume immediate release of all Ag to the solution. On the other hand, the resulting concentration of Ag is limited by its solubility, meaning that the amount of Ag available for release from the canister interior is still limited. In the safety assessment SR-Site, it was assumed that the saturated solution would have the same ratio of radioactive Ag-108m to stable Ag as the total Ag inventory, thereby yielding a certain concentration of Ag-108m in the solution. However, if Ag-108m is concentrated to the outer part of the control rod, a higher concentration of Ag-108m in solution would result if only this outer part is dissolved.

Studies of used control rods show that the majority of the activation products are located in the lower part of the ca 3.8 m long rods. More specifically, for rodlets studied in Studsvik the activity decreases to almost zero at around 35 cm from the bottom of the rodlet (SKBdoc 1396058). PWR control rods were also studied by Lepel et al. (1995) showing a very large change in concentration of Ag-108m along the rod, from about 200 Bq/g to ca 1×10^9 Bq/g at the very bottom of the rod. This axial distribution originates from how the rods are used in the reactor, and the immediate effect of the rods on the neutron fluence when lowered into the core. This means that almost all activity is found at the bottom end of the control rod, and that this axial distribution may influence the release of radionuclides more than the radial distribution.

Since the radial concentration profile of activation products is expected to vary according to neutron fluence, this profile will vary along a single rod and between different rods in the control rod cluster. In any case, the profile concentration of activation products can be expected to be highest near the surface and decrease inwards. This is exemplified by the studies presented in Zakharov et al. (2000) and Kryger et al. (1995), who presents radial profiles of neutron capture in B₄C absorber rodlets. For this material, burnup in the centre of the rodlet is only ca 5% while in the outer rim (about 1mm) it is about 50%. The burn-up profile for this absorber material is not very sharp; the same is expected for the Ag-Cd-In alloy.

Assuming all Ag-108m is formed in the outer layer

It is here assumed that neutron absorption only takes place uniformly in the first mm into the alloy. From this information it is possible to calculate the volume of material which contains Ag-108m. By only considering the affected surface layer, the ratio between radioactive and stable Ag becomes higher. This ratio can then be used in a calculation similar to the one already performed for SR-Site, but adjusted to only involving immediate release of the affected surface layer of the control rod. With a control rod radius of r_0 mm and an unaffected inner part of r_1 mm, the concentration of Ag-108m in the affected layer increases a factor F relative to a situation where the isotope is uniformly distributed over the control rod cross section, where F is

$$F = \frac{r_0^2}{r_0^2 - r_1^2}$$

With $r_0 = 5$ mm and $r_1 = 4$ mm, one obtains $F = 25/9$.

The effect on dose and risk would be an increase with a factor of 25/9, i.e. by about a factor of 3. Considering that the peak dose from Ag-108m is about three orders of magnitude below the dose corresponding to SSM:s risk limit, see Figure 5-6 in TR-10-50, this treatment of the release of Ag-108m, that takes into account the uneven distribution of Ag-108m in the control rods, still yields a dose that is more than two orders of magnitude below the risk limit.

In conclusion, also when the uneven distribution of Ag-108m in the control rods is taken reasonably into account, the calculated risk from Ag-108m is negligible.

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