



Public

Memo

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# Corrosion of the copper canister inside due to radiolysis of remaining water in the insert

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## 1 Background

The Swedish Radiation Safety Authority (SSM) has requested additional information on various aspects of SKB's licence application for the disposal of spent nuclear fuel. As part of this request, SSM has asked about the possibility of corrosion in the gap between the cast iron insert and the copper shell (SSM 2012, question 6).

Specifically, SSM requests a description of how condensation of water remaining in the copper canister can cause corrosion in local areas, due to radiolysis and on how the corrosion at the canister inside is influenced by volatile fission products as e.g. iodine.

This memo is a supporting document for SKB:s answer to SSM:s request.

## 2 Prerequisites for the analysis

The most important design premise for the copper canister relevant for the discussion that follows is that at most 600 g of water is allowed to be left inside the insert (SKB 2009). Any water not removed in the drying process is most probably located inside fuel rods with a damaged zirkaloy cladding. These 600 g of water would correspond to the presence of 12 damaged fuel rods per canister, each rod containing 50 g of water (SKB 2010a).

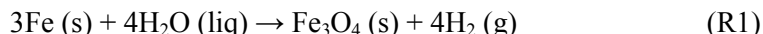
The number of known damaged fuel rods is small. In addition, there may be damaged rods that are not known to be damaged. The rods will be distributed randomly among the canisters and the probability that there will be several damaged rods in one canister is low. The assumption that 600 g water is present inside the iron insert is thus very pessimistic.

In the analysis it is further assumed that the seal between the cast iron insert and the steel lid is not tight, and thus that water and volatile substances including radionuclides can migrate to the gap between the iron insert and the copper shell. The detailed dimensions of the canister are taken from SKB (2010b).

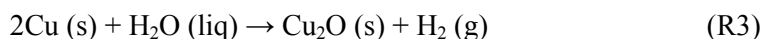
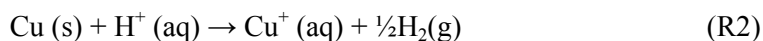
### 2.1 Thermodynamics for copper and iron corrosion

The thermodynamic corrosion product of iron in water is magnetite, which forms according to reaction R1.

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For the temperatures of interest here, the equilibrium hydrogen pressure for R1 is on the order of  $10^3$  bar. The corrosion of copper in pure water is determined by either thermodynamic or kinetic limitations, by the reactions R2 and R3.



As discussed by Macdonald and Sharifi-Asl, the equilibrium hydrogen pressure is less than  $10^{-13}$  bar for copper in pure water of pH 7, even at the elevated temperature inside the canister (Macdonald and Sharifi-Asl 2011). This implies that if copper and iron are present in the same closed volume, copper should not corrode by pure water because corrosion of iron will generate a hydrogen pressure that exceeds the equilibrium pressure for copper corrosion by several orders of magnitude. Even if one would assume that the experimentally measured hydrogen pressure by Szakálos and co-workers is established, copper corrosion would cease when the hydrogen pressure exceeds  $10^{-3}$  bar as found in those experiments (Szakálos et al. 2007).

Nevertheless, corrosion in the system at hand is not determined by equilibrium thermodynamics, since corroding species are generated by gamma radiolysis of water. It is pessimistically assumed that the radiolysis products react with copper and/or iron surfaces before they have had time to recombine to water.

## 2.2 Temperature and temperature differences in the canister

During the first few hundreds of years considerable heat is generated in the canister by the energy released from the decaying radionuclides. The maximum energy production rate is 1.7 kW. A large part comes from the decay of Sr-90 and Cs-137, both with half-lives of about 30 years. After 200 years the energy production rate has decreased with about 99%. The temperature in various parts in the KBS-3 system is shown in Figure 1. The figure shows the results of the thermal calculation for Forsmark as reported in SR-Can (SKB 2006). The description of the thermal evolution in the near field was slightly modified for the updated safety assessment SR-Site (SKB 2011, Chapter 10.3.4). On average, less than one of 6,000 canister positions would have a peak buffer temperature larger than  $95^\circ\text{C}$ . The large majority of the canisters, about 98%, will have a peak buffer temperature lower than  $90^\circ\text{C}$ . Since the canister temperature has been shown to be around  $10^\circ\text{C}$  higher than the buffer temperature during the initial warm phase (Figure 1), this means that the majority of the canisters will have a peak temperature lower than  $100^\circ\text{C}$ .

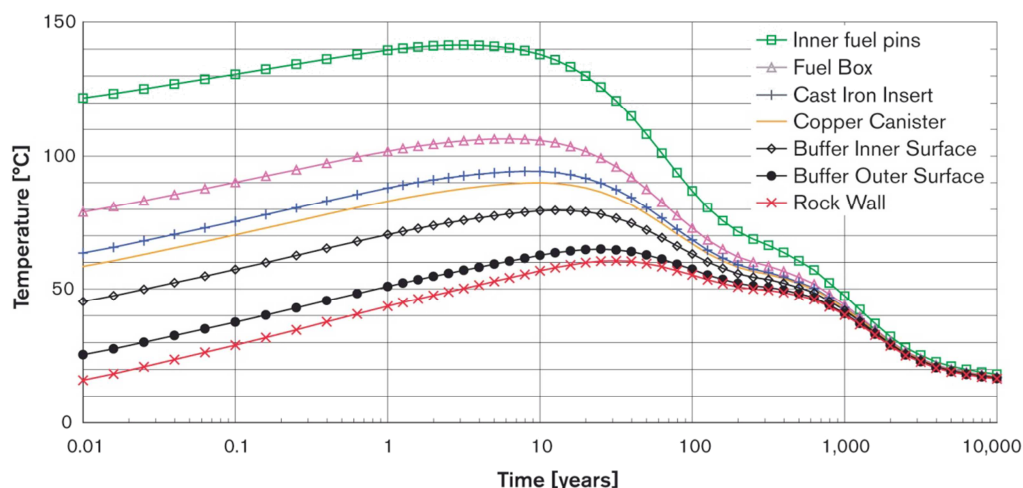


Figure 1. Temperature evolution in a canister (SKB 2006, Figure 9-17).

### 2.3 Basic constants and data used in the calculations

Avogadro number	$6.02 \times 10^{23}$ per mol
Density of copper	$8.96 \times 10^6$ g/m <sup>3</sup>
Molar mass of copper	63.546 g/mol
Molar mass of water	18.0 g/mol
Gamma Q-value for Cs-37 decay <sup>1</sup>	$0.662 \times 10^6$ eV/decay
G-value for oxidising species <sup>2</sup>	0.035 species/eV

#### Gamma attenuation coefficients for 662 keV gamma radiation from Cs-137<sup>3</sup>

Water	0.1 cm <sup>-1</sup>
Iron	0.6 cm <sup>-1</sup>

#### Canister data (BWR except void volume for PWR)

Void volume in canister, estimated <sup>4</sup>	0.7 m <sup>3</sup>
Gap width between iron and copper	1.5 mm
Inner canister length	4.62 m
Dimension square form hole in iron insert	0.16 m
Inner surface area of copper canister	15 m <sup>2</sup>

#### Fuel data (BWR)

Number of fuel rods in a canister	720
Void volume fraction in iron insert <sup>5</sup>	0.5
Nuclide inventory	see Table 1 and 2

1 Qvalue calculator, National Nuclear Data Center Brookhaven, [www.nndc.bnl.gov/qcalc](http://www.nndc.bnl.gov/qcalc). (Figure 4.5 in Chopin and Rydberg, 1980, gives the same value).

2 Approximate G values for the most common primary oxidising species [http://www.kth.se/polopoly\\_fs/1.226777!/Menu/general/column-content/attachment/rad\\_chem2.pdf](http://www.kth.se/polopoly_fs/1.226777!/Menu/general/column-content/attachment/rad_chem2.pdf), (Lide, 2008) give similar values.

3 Values taken from (Lide, 2008).

4 Value taken for PWR (SKB 2010e, Table 4.3). This has the smallest volume, which is conservative as water will condense earlier.

5 Estimated from geometry of insert. Results are not sensitive to this value

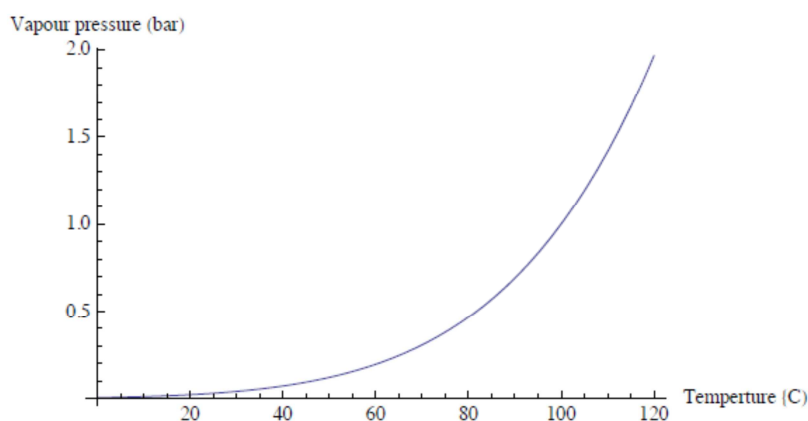
### 3 Analysis and discussion

#### 3.1 Structure

In this section we will first assess the evaporation and condensation of water inside the canister in order to determine when and where water will exist as vapour and as liquid. Then a simple mass balance calculation is performed to determine the maximum amount of copper that can corrode if all water is used for this. After that, we discuss the possibility that the copper shell is cathodically protected by its contact with the iron insert, a mechanism that may limit the corrosion of copper. Thereafter the rates of formation of corrosive agents by radiolysis are estimated for different conditions and in different locations in the interior of the canister. Furthermore the release of volatile corrosive radionuclides and their contribution to corrosion is estimated. It is emphasized that except for corrosive radionuclides coming from the fuel, the mass balance based estimate of corrosion by water is the largest possible amount of corrosion whatever the detailed reaction mechanisms, since the corrosive species come from the radiolysis of water itself.

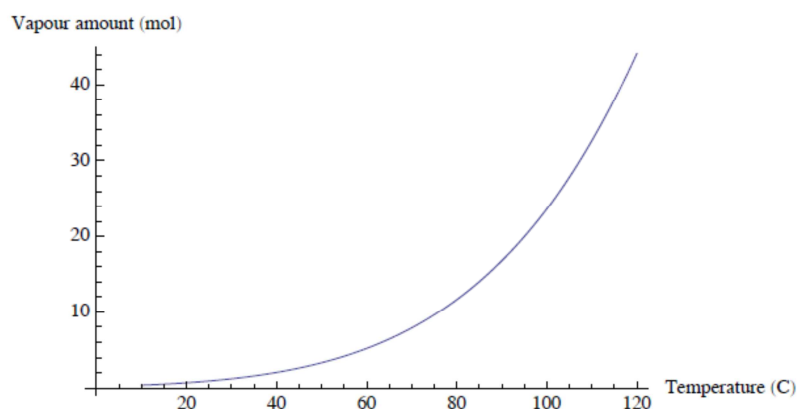
#### 3.2 Vaporisation and condensation of water

The temperature in the fuel rods is much higher than in the iron insert, which in turn is a few degrees (°C) higher than the copper shell. The water in the fuel rods can evaporate and condense on the cooler inner walls of the copper canister. There is enough water for part of it to be able to liquefy in the temperature range of interest. Figure 2 shows the vapour pressure of water at different temperatures.



**Figure 2.** Vapour pressure at different temperatures.

Figure 3 shows the amount of water that exists as vapour at different temperatures. The total void volume available is about 0.7 m<sup>3</sup>.



**Figure 3.** Amount of water as vapour at different temperatures (for a void volume of  $0.7 \text{ m}^3$ ).

As shown in Figure 3, the maximum allowed amount of water in the canister (600 g or 33.3 mol) will all be vaporised at  $110^\circ\text{C}$ . Below this temperature a fraction of the water always will exist as liquid in the cooler parts of the canister. Most of this liquid water will be found in the lower part of the 1.5 mm wide gap between the iron and copper. If all the water is liquid it would have a height of 134 mm in the gap. This is 3% of the entire height.

### 3.3 Mass balance considerations

A worst case conservative scenario can be analysed by assuming hypothetically that:

- i) the maximum allowed amount of water (600 g) is collected as liquid at the bottom of the canister-insert gap (which gives a 134 mm high liquid column if the gap is assumed to have a uniform thickness)
- ii) the total amount of water is split by radiolysis
- iii) the radiolysis products only reacts with and corrode the copper surface (and not the iron surface)

Under these assumptions the corrosion depth at the lower 134 mm part of the copper shells inside would be 1.1 mm.

If on the other hand the corrosion would occur evenly over the entire inner copper surface the corrosion depth would be 0.03 mm. Since condensation will occur over the whole inner copper surface, it seems unlikely that the corrosion would be localised to the lower part of the gap.

The mass balance approach sets an upper bound on the amount of copper that can corrode by reaction with water. One mole of water will at most corrode two moles of copper, whatever the reaction mechanism is, and whether the reaction occurs via radiolysis of water or not.

### 3.4 Iron as a sacrificial anode

The iron insert rests on the copper and they are in electrical contact, which would promote a preferred corrosion of the more reactive iron. Even if there is corrosion at the bottom of the iron insert forming magnetite according to reaction formula (R1) the electrical contact remains because magnetite conducts electricity. Thus, the iron insert could still act as a “sacrificial” anode and protect copper from corroding.

### 3.5 Radiolysis

Gamma radiation from the spent fuel pellets penetrate the zircaloy tube walls and will to some extent cause radiolysis of water in the interior of the canister. The water molecules dissociate into oxidising and reducing species, such as hydrated electrons ( $e_{aq}^-$ ), OH radicals,  $H_2$ ,  $H_2O_2$  and other species with different re-combination kinetics. Oxidising species can react with copper- and iron surfaces unless they recombine with reducing species in either solution or gas-phase. This mechanism can conceivably corrode copper, especially if the copper and iron are not in electrical contact with each other. This would, however, lead to a build-up of the concentration of reducing species, mostly hydrogen, which would tend to reduce the copper oxide.

Furthermore, due to the corrosion of iron (R1), a partial pressure of hydrogen will be built up in the gap. This hydrogen gas will reduce the amount of corrosive species resulting from gamma radiolysis of water, i.e. molecular hydrogen acts as a scavenger of oxidizing species (Pastina et al. 1999, Pastina and LaVerne 2001), which will make the environment less corrosive.

#### 3.5.1 Radiolysis in the insert and in the gap between insert and copper shell

In this section the rate of production of oxidizing species is estimated assuming that these can react with the copper surfaces before recombining to water. It may be noted that in operating nuclear reactors most species recombine before they have had time to react with the construction steel in the reactor. In a reactor the hydrogen pressure is much smaller than what is expected inside the canister where iron corrodes and where the hydrogen cannot escape. Nevertheless, due to differences in geometry, surface to volume ratio and construction materials, the kinetic situation in the insert-canister gap might differ from the situation in a reactor.

During the first hundred years Cs-137 dominates the production of gamma radiation. In the example below the following data for a BWR canister is used.

Activity Cs-137	$4.21 \times 10^{15}$ Bq (SKB 2010c, Table 6-4)
Q-value	$0.662 \times 10^6$ eV/Cs-atom (BIPM 2008)
Fraction of decay to produce gamma	0.946 (BIPM 2008)
G-value (primary production of ox. species) <sup>6</sup>	0.035 species/eV

For comparison with the G-value given above, it may be noted that if all energy was efficiently absorbed by water, and if water was split into  $H_2$  and  $O_2$ , then 0.42  $H_2O$  molecules would be consumed per eV energy unit (estimated by Gibbs free energy of formation of water from  $H_2$  and  $O_2$ ).

If all the 422 W gamma radiation (at early times) from Cs-137 in a BWR canister were to produce oxidising species (primarily OH radicals and  $H_2O_2$ ) with the G-value 0.035, then

4 688 mol oxidising species would be produced annually, corresponding to 84 400 g water. This is much more than the 600 g of water assumed to be present. However, only a fraction of the radiation is absorbed by water, the rest is absorbed by fuel, iron, copper and material outside the canister. The absorbed fraction can be determined using Lambert-Beer's law.

$$I = I_0 e^{-\mu x}$$

where,  $I_0$  is the intensity of the incoming radiation,  $I$  is the intensity that passes a thickness  $x$  of the medium and  $\mu$  the linear attenuation coefficient. The attenuation is proportional to the number of atoms of the medium through which the radiation passes and is independent of the density of the medium. This implies that the attenuation of water vapour and liquid water is the same per mass unit

<sup>6</sup> G-value for production of the most important primary products, electrons ( $e_{aq}^-$ ), OH radicals,  $H_2$ ,  $H_2O_2$  and other species vary between 0.005 and about 0.03 species/eV.

[http://www.kth.se/polopoly\\_fs/1.226777!/Menu/general/column-content/attachment/rad\\_chem2.pdf](http://www.kth.se/polopoly_fs/1.226777!/Menu/general/column-content/attachment/rad_chem2.pdf)

of water through which the radiation passes. Calculated results of absorbed energy and production of oxidising species are presented below for two cases to illustrate what happens in vapour and liquid in the canister.

In the first case all water is vaporised. Most of the vapour is present in the large voids in the iron insert, about  $0.7 \text{ m}^3$ . The void volume of the gap between the iron and copper is  $0.022 \text{ m}^3$ . As the mass based attenuation coefficient is constant the mass of water vapour in the voids will attenuate the amount of energy in the same way as if the water was spread out as a thin film on the inner surfaces of the iron insert. This considerably simplifies the calculations as there is no need to account for the complex geometry of the void volume. The estimate made is conservative as no energy loss in the fuel pellets and the zirkaloy cladding surrounding these is accounted for. The small amount of vapour in the gap between iron and copper can be disregarded in comparison because the radiation intensity is considerably attenuated during passage through the iron.

The area of the surfaces in the iron insert is  $35 \text{ m}^2$  and the thickness of the hypothetical water film evenly spread over this area is  $17.4 \times 10^{-6} \text{ m}$ . The linear attenuation coefficient in water for gamma radiation from Cs-137 (662 keV) is about  $0.1 \text{ cm}^{-1}$ . Using Lambert-Beer's law it is found that a fraction 0.999826 of the radiation passes the water film and thus a fraction 0.000174 of it is absorbed in the water. This means that 14.7 g/year water is split into oxidising and reducing species at early times. The radiation intensity in the gap between iron and copper is only 5% of that in the voids in the iron insert and the mass of water there is much smaller.

Accounting for the decay of Cs-137 with a half-life of 30.15 years, it is found that after 100 years 328 g water has undergone radiolysis. When all Cs-137 has decayed the corresponding amount of water consumed would be 364 g. Since practically all vapour radiolysis occurs in the iron insert it is reasonable to assume that practically all of the oxidising species produced in the insert react with iron and not with copper in a scenario where all water exists as vapour.

In the insert-canister gap, having only 3.1% of the total void volume and being exposed to only 5% of the radiation intensity, a minor fraction of the total amount of water undergoes radiolysis. This fraction can be estimated as  $0.05 \times 0.031 = 0.00155$  of the amount of water split in the insert void, i.e. 0.5 g water by the time all Cs-137 has decayed. If half of this corrodes copper spread out over the entire copper surface the corrosion depth would be  $14 \times 10^{-6} \text{ mm}$ .

The situation is different if we instead assume that all the water has condensed to liquid in the gap between iron and copper because it is cooler there than in the insert as discussed in section 3.2. The water column in the gap is 1.5 mm thick and stands to a height of 134 mm. The gamma radiation reaching the gap is only 5% of that in the insert and the water is exposed to a fraction of this intensity that is equal to the height of water column (134 mm) to the entire height of the gap (4630 mm). This fraction is  $0.05 \times 134/4630 = 0.00145$ . Lambert-Beer's law gives that 1.5% of this energy is absorbed in the water. The energy from Cs-137 gamma decay absorbed in the water in this case is 0.0090 watt. Initially 1.04 g/year water is split and after 1000 years 45 g has undergone radiolysis. If the entire mass of oxidising species would react with copper, the corrosion depth would be 0.07 mm. If one were to account for the effect of gradually disappearing water (due to consumption via corrosion), the lowest part of the 134 mm height might corrode to about twice the depth.

It may be noted that the assumption that only the gamma energy from Cs-137 need to be accounted for is reasonable because alpha and beta particles do not penetrate the metal tube (zirkaloy cladding) of the fuel rods, but deposit their energy in the fuel pellet and possibly in the minute amount of vapour inside the pellets and cladding. Any produced oxidising species are not likely to escape from the rod and to migrate to the gap between the insert and the copper shell.

### **3.6 Oxidising radiolysis products generated by instant release fraction IRF in damaged fuel rods**

Some nuclides in the fuel can migrate to the surface of the pellets and accumulate there during reactor operation. Some of these nuclides are or can form volatile species and escape from the damaged rods.

These are primarily tritium (as THO), C-14 (as CH<sub>4</sub> or CO<sub>2</sub>), I-129 (as I<sub>2</sub>) and Cl-36 (as Cl<sub>2</sub>). Cs-137 and Sr-90 could conceivably escape dissolved in water if liquid water enters the damaged rods. The latter can be excluded during the first hundred years after deposition when the temperature is above 100°C, and they have decayed considerably by the time liquid water can exist in the fuel rods. In Table 1 the activity of nuclides in the instant release fraction are shown. The nuclides that may be considered to be able to escape are marked as bold and highlighted in yellow. The table also shows how much water that would undergo radiolysis if all the energy of the beta radiation, practically only from Cs-137, would be absorbed in the water. This is a conservative assumption because only a fraction of the energy is absorbed by the water, the remaining energy is deposited in the metals in the canister. In total 0.002 mol/year oxidising species could be produced via beta radiolysis of water under these assumptions.

Even if other nuclides in Table 1 could migrate to the canister-insert gap, these could produce at most 2.3 mol/year oxidising species, generated mostly by Cs-137 beta decay. This would during early times consume about 40 g water per year. This example is meant to illustrate an entirely hypothetical case because it is not conceivable that these nuclides, except those marked yellow can migrate to the gap between the iron and copper.



**Table 1. Activity of nuclides from the instant release fraction in a BWR canister with 12 damaged fuel rods, Q-values, rate of production of oxidants. Data from SKB (2010d): Inventory from Table E-1 in year 2045, Correction factor from Table E-2, IRF (Instant Release Fraction) from Table 3-4; nuclides that may be considered to be able to escape are marked as bold and highlighted in yellow.**

Nuclide	Activity in a BWR canister w. 12 damaged rods, Bq	Q-value, keV, per beta and gamma decay	Energy/s deposited in water, eV/s	Split <sup>7</sup> molecules H <sub>2</sub> O/s	Split mol H <sub>2</sub> O/year
Ag-108m	1.88E+11	1646	3.10E+17	6.20E+15	3.25E-01
<b>C-14</b>	1.28E+08	156	2.01E+13	4.01E+11	<b>2.11E-05</b>
Cd-113m	1.25E+10	323	4.02E+15	8.04E+13	4.22E-03
<b>Cl-36</b>	5.54E+05	710	3.93E+11	7.86E+09	<b>4.13E-07</b>
Cs-135	1.87E+07	269	5.03E+12	1.01E+11	5.28E-06
Cs-137	1.85E+12	1176	2.18E+18	4.36E+16	2.29E+00
<b>H-3</b>	1.05E+11	19	1.95E+15	3.91E+13	<b>2.05E-03</b>
<b>I-129</b>	1.10E+06	189	2.08E+11	4.16E+09	<b>2.18E-07</b>
Mo-93	1.07E+05	407	4.34E+10	8.68E+08	4.56E-08
Nb-93m	1.16E+10	31	3.57E+14	7.14E+12	3.75E-04
Nb-94	4.57E+07	1200	5.49E+13	1.10E+12	5.76E-05
Ni-59	5.70E+07	1073	6.12E+13	1.22E+12	6.43E-05
Ni-63	6.04E+09	67	4.05E+14	8.10E+12	4.25E-04
Pd-107	3.25E+05	34	1.11E+10	2.22E+08	1.16E-08
Se-79	4.27E+05	151	6.44E+10	1.29E+09	6.76E-08
Sn-121m	5.40E+06	401	2.16E+12	4.33E+10	2.27E-06
Sn-126	2.24E+05	378	8.46E+10	1.69E+09	8.88E-08
Sr-90	1.07E+11	546	5.82E+16	1.16E+15	6.11E-02
Tc-99	3.74E+07	295	1.10E+13	2.21E+11	1.16E-05

### 3.7 Oxidation by iodide and chloride

Table 2 shows the amount of different nuclides from the IRF (Instant release fraction) that could escape and dissolve in liquid water. Only the yellow marked Cl-36 and I-129 are volatile and oxidising and can chemically react with copper and iron. Jointly they could, in all, oxidise 0.08 mol copper to Cu(I) if one atom of Cl or I oxidizes one atom of Cu. It is, however, likely that they react with iron before they reach the copper. Of the other nuclides only H-3 and C-14 can form volatile compounds but are not likely to oxidise copper.

<sup>7</sup> Same number, expressed as equivalents, of oxidising and reducing species are formed. Given as split moles of water to refer to reaction (R1)

**Table 2. Inventory in a BWR canister of nuclides that have an IRF fraction and the amount that could dissolve in water (SKB 2010d).**

Nuclide	Nuclide in an BWR canister, year 2045, mol	IRF fraction, (SKB 2010d, Table 3-4)	mol dissolved in water
Ag-108m	0.109	1	0.1086
C-14	0.036	0.092	0.0033
Cd-113m	0.001	1	0.0008
<b>Cl-36</b>	0.009	0.086	<b>0.0008</b>
Cs-135	6.730	0.029	0.1952
Cs-137	8.690	0.029	0.2520
H-3	0.006	1	0.0058
<b>I-129</b>	2.700	0.029	<b>0.0783</b>
Mo-93	0.000	0,012	0.0000
Nb-93m	0.050	0.017	0.0008
Nb-94	0.234	0.018	0.0042
Ni-59	1.621	0.012	0.0194
Ni-63	0.229	0.012	0.0027
Pd-107	4.780	2.00E-03	0.0096
Se-79	0.136	4.20E-03	0.0006
Sn-121m	0.007	1.90E-04	0,0000
Sn-126	0.339	3.00E-04	0.0001
Sr-90	5.430	2.50E-03	0,0136
Tc-99	17.900	2.00E-03	0.0358

### **3.8 Comparison with other studies**

Sjöblom and co-workers studied corrosion in copper canisters that had been electrically heated during a long time to simulate real conditions (Sjöblom et al. 2013). They found that copper as well as iron had corroded in some locations. It was found that the corrosion was caused by decay products of polymeric materials on electrical insulators and cable glands. Such heater installations and materials will not exist in the copper canisters that will be deposited with spent nuclear fuel. These observations are thus not relevant for the present report.

## **4 Conclusions**

A mass-balance with 600 g water would corrode the copper shell to a depth of 1.1 mm locally if all water were to react with copper and the corrosion was to take place in the bottom of the gap between the iron insert and the copper shell where liquid water could form. Such a corrosion scenario is not likely for at least three reasons

- 1) it is highly unlikely that one canister would contain as many as the 12 damaged rods needed to result in 600 g water left in the canister
- 2) the iron insert would not be inert, but would also consume oxidizing species by corrosion
- 3) corrosion of copper would not occur only in contact with liquid water but on all surfaces wetted by condensation

Nevertheless, gamma radiolysis of water will occur, and the radiolysis products will to some extent corrode the copper canister. More realistic estimates of the extent of radiolysis mediated corrosion shows that this mechanism could locally corrode the copper canister to a depth of 0.07 mm. Volatile corrosive components (chlorine and iodine) from damaged fuel rods have a negligible effect on the total amount of corrosion.

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