Tritium in Decommissioned Reactors

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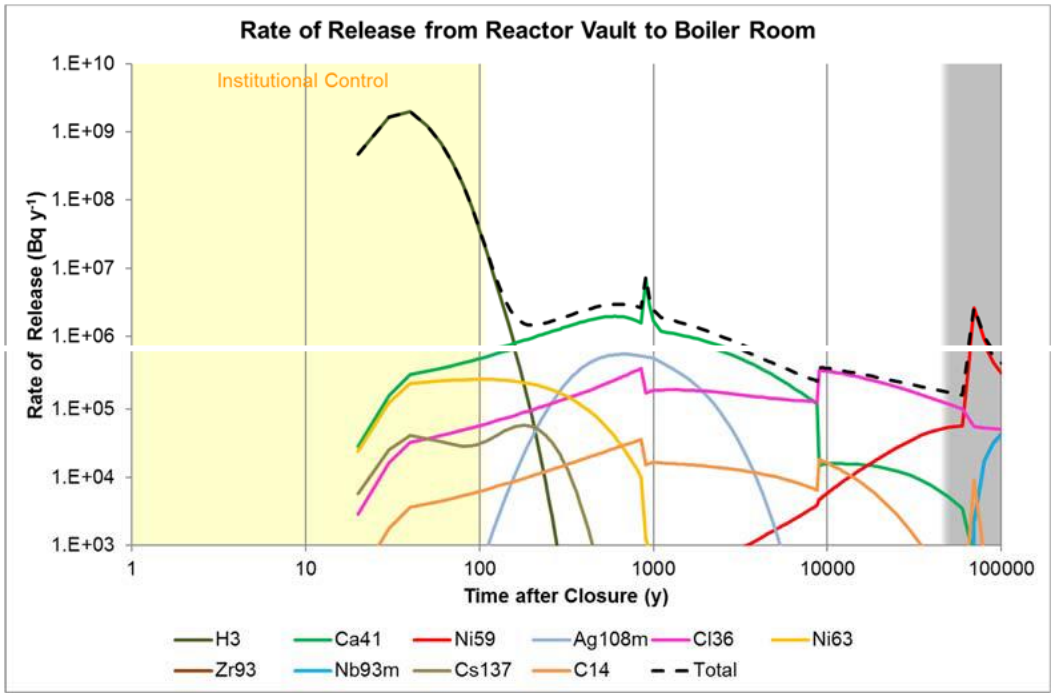
Tritium is the most significant nuclide present in decommissioned reactors as evidenced by their very high annual releases even after many years following their decommissioning. See https://www.ianfairlie.org/news/continued-radioactive-emissions-from-old-closed-nuclear-reactors/

The figure below indicates release rates for various nuclides from the reactor vault into the plant's boiler room as estimated by Canadian Nuclear Laboratories from the Rolphton reactor in Canada. It can be seen that tritium release rates for the first 100 years are about 3 orders of magnitude greater than most other nuclides and 5 orders of magnitude greater than carbon-14.

Source: CNL (2016a) Technical Document WR-1 Reactor Radiological Characterization Summary and Radionuclide Inventory Estimates. WLDP-26100-041-000-0001. August 2016.



According to Krasznai (1993), in the early 1990s, high tritium concentrations up to 82,000 Bq/g were found in the concrete structures at the NPD reactor at Rolphton near Chalk River in Canada. This had been closed with their fuels removed since ~ 1985. These tritium concentrations were very high compared with ~300 Bq/g for C-14 the next highest concentration of a nuclide (Krasznai, 1993).

The high tritium concentrations in decommissioned reactors are due to

- neutron activation of hydrogen, deuterium and Li-6 impurities,
- tertiary fission (fission yield <0.01%) and
- diffusion from high levels of tritium in cooling water and moderator (Kim et al, 2008).

As stated by Kim (2009) "During the lifetime of nuclear sites tritium becomes incorporated into the fabric of the buildings. **When nuclear decommissioning works and environmental assessments are undertaken it is necessary to accurately evaluate tritium activities in a wide range of materials prior to any waste sentencing**."

Conventional computer models can give unreliable predictions of tritium concentrations in decommissioned reactors. For example, according to Corcoran et al (2017), steel containment vessels used for >20 years "**exhibit tritium burdens greatly exceeding those predicted by simple gas solution in the parent metal."**

Investigations into the location of, and activity release from, vessel materials indicate the existence of two major tritium sources:
(1) the bulk metal where in-depth contamination arises from diffusion/solution; and
(2) a highly active surface layer, responsible for holding the main tritium inventory (Corcoran et al, 2017).

Models based on neutron activation codes alone may have incorrectly predicted tritium levels. As stated by Kim et al (2008) "Without an appreciation that two forms of tritium exist in concrete reactor bioshields, the H-3 content of samples may be severely underestimated using conventional analytical approaches. The two forms are strongly-bound tritium and loosely-bound tritium. The former originates from neutron capture mainly on trace (1 part per 20,000) lithium (Li-6) within mineral phases, and requires temperatures in excess of 800 °C to achieve quantitative recovery. The weakly-bound form of tritium can be liberated at lower temperatures (100 °C) as HTO and is associated with dehydration of hydrous mineral components."

In metals, tritium is retained by absorption of free water in the hydrated surface oxidation layer, by H ingress into bulk metal and also as lattice-bound tritium produced by neutron activation. (Nishikawa M et al, 2006).

In addition, Croudace et al (2014) found that significant tritium was incorporated in non-irradiated metals (e.g., stainless steel and copper), following prolonged exposure to tritiated water vapour (HTO) and/or tritium/hydrogen gas (HT) in nuclear facilities. In irradiated metals, an additional type of tritium was formed internally through neutron capture reactions. The amount formed depended on the concentration and distribution of trace

lithium and boron in the metal.

Computer models often use unconservative assumptions. For example, if they use measured nuclide levels of the concrete bioshields on their external surfaces, that is where the lowest levels exist while the highest levels are on the other end, nearest the reactor core.

As concluded by Kim et al (2008) "**These findings exemplify the need to develop robust radioactive waste characterization procedures in support of nuclear decommissioning programs**".

References

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