Behaviour of Carbon-14 Released from Activated Steel in Repository Conditions – a Key Issue in the Long-term Safety of Decommissioning Waste

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Fortum

Contents of the presentation

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- final disposal of activated components in Loviisa NPP
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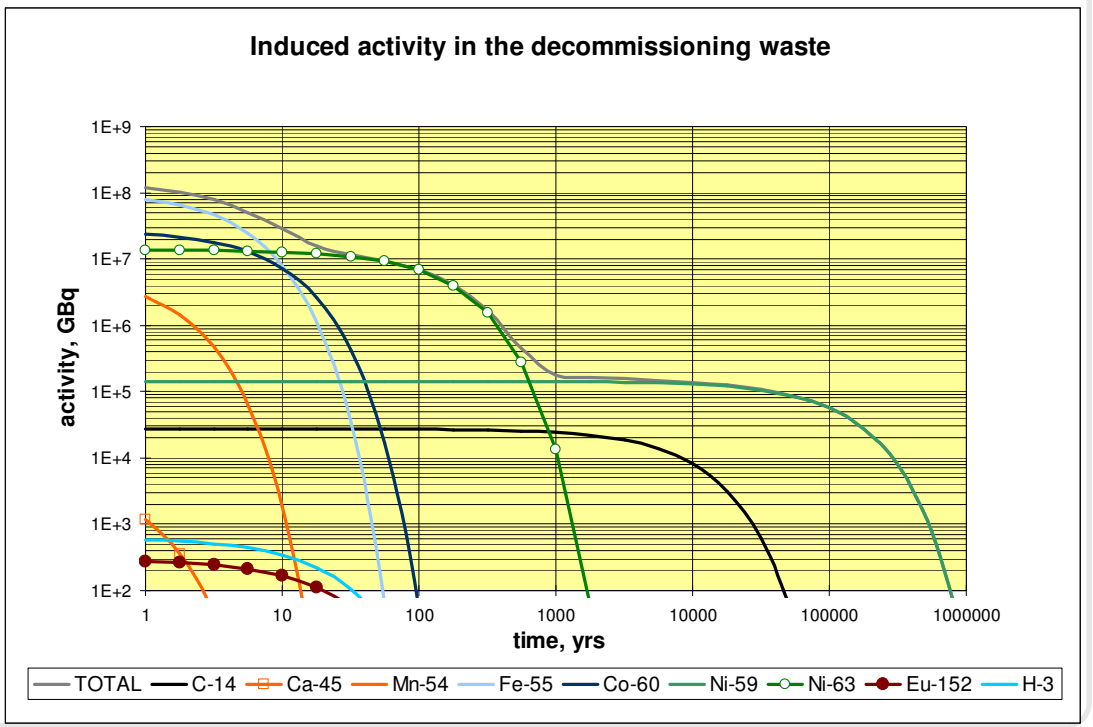
- possible mechanisms to affect the speciation
- ideas for further experimental work - and related problems

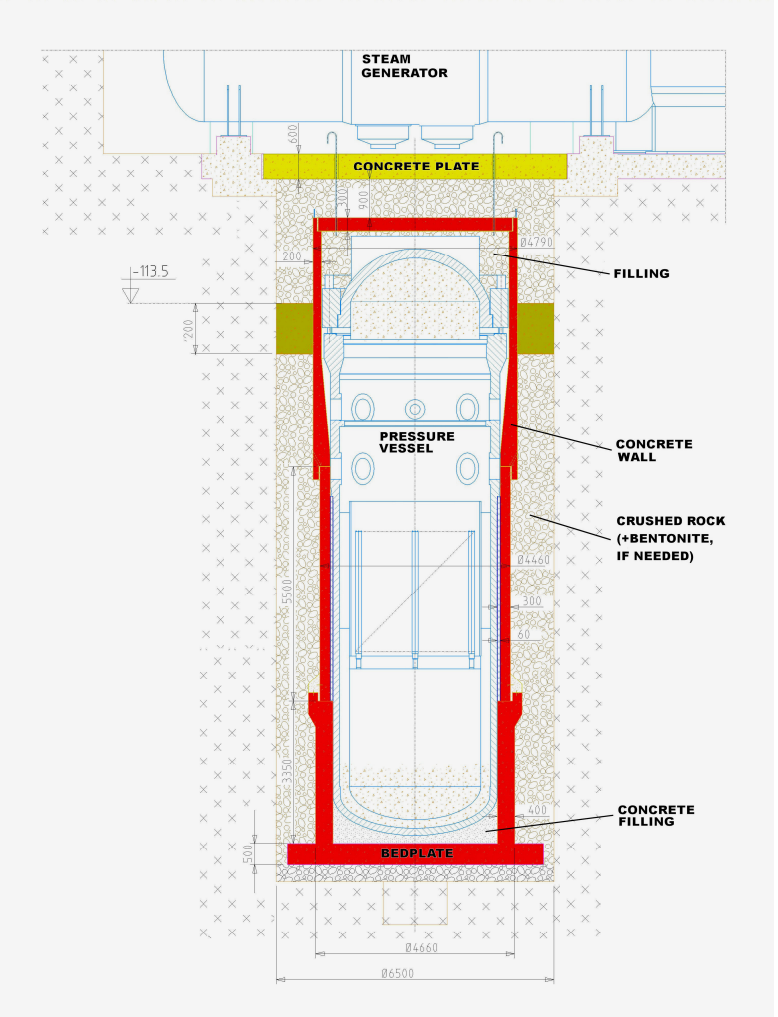
Origin and activity of C-14 in activated metal components

- Neutron activation, three reactions:
  - N-14 (n,p) C-14
  - C-13 (n,g) C-14
  - O-17 (n,a) C-14
- In an LWR, in stainless steel the reaction from N-14 dominates

Induced activity in the decommissioning waste of the Loviisa NPP



Final disposal of activated components; case: Loviisa NPP, reactor pressure vessel with internals

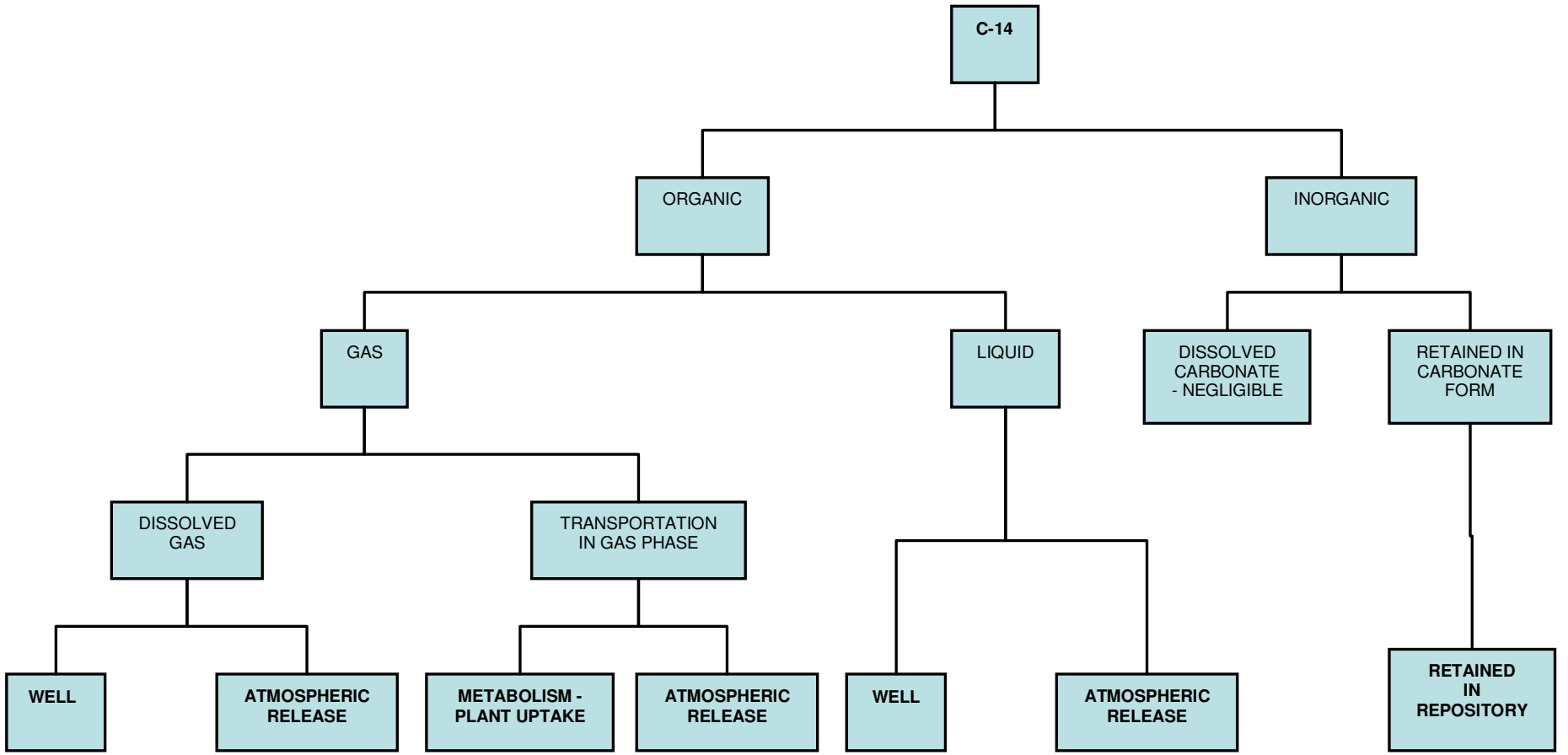


- One-piece removal of large components
- Reactor internals are packed in the RPV
- The nozzles are sealed.
- Surrounding concrete silo structure
- Low corrosion rate

Release from steel and chemical speciation of carbon

- Carbon is assumed to be released from steel, as the steel corrodes.
- In anaerobic conditions in cementitious environment the corrosion rate is low (~0.1 µm/a).
- Diffusion from steel is believed to be low compared to corrosion.
- Chemical speciation *in* the activated steel (carbide?; what happens if C-14 is formed from N-14 as nitride? High neutron energy involved.)
- Chemical speciation after being released from the activated steel?
  - carbonate? => low solubility and large amount stable carbonate in the cementitious repository conditions => OK
  - soluble organic form? => possibly very mobile
  - gaseous organic form? => solubility, mobility
- According to Pourbaix diagram the organic forms should not be stable in the alkaline and reducing repository conditions; still there are indications of their existence.

Possible migration routes for C-14 from the repository to the biosphere



Some comments on the available literature and the conducted experimental work

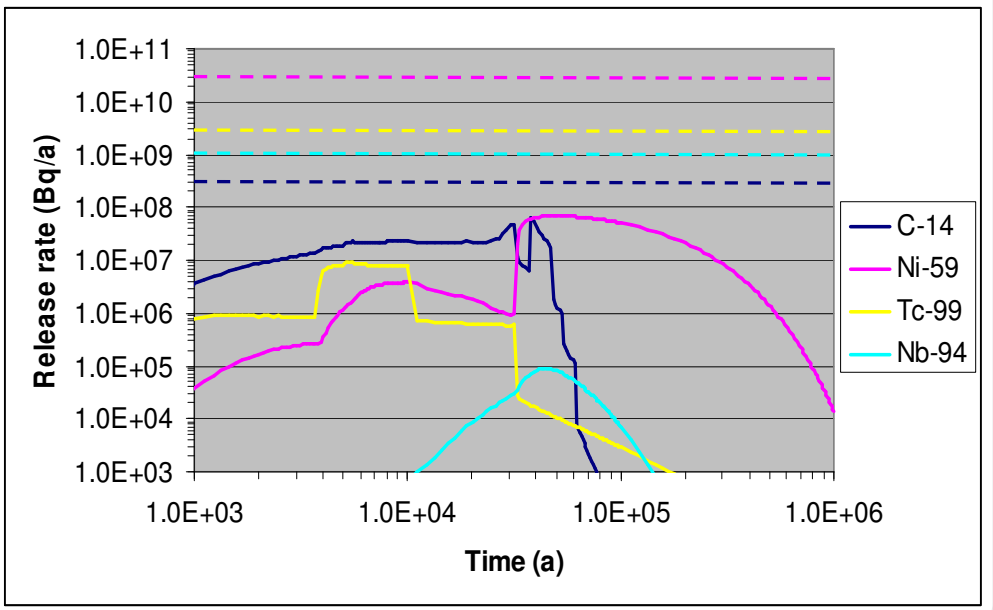
- Only few relevant articles on experimental results are available.
- Experimental works by Deng et al. (1997) and Kaneko et al. (2003) are often cited
  - open questions regarding coverage, representativeness and relevance for the repository issue
    - phenomenology to some extent unclear
    - redox conditions may not be representative
    - Both gaseous and soluble organic compounds were detected
    - No straightforward quantitative conclusions can be drawn.
- A master's thesis (Kuitunen 2007) was financed by Fortum for the safety assessment of Loviisa NPP
  - Literature survey
  - ***…even though the formation of organic species has not been confirmed, their existence cannot be denied and this should be taken into account in the future safety assessments.***
- Research is going on…

Approach chosen for the safety assessment of Loviisa NPP decommissioning plan

- Release from the metal according to the corrosion rate
- Soluble and mobile chemical form assumed
- Sensitivity analysis: gaseous chemical form

- => C-14 is the dominant nuclide

Release rates as 1000 years' averages compared to regulatory constraints (dashed lines)



Possible phenomena/mechanisms to tackle the problem

- chemical reactions between organic forms and carbonate; kinetics?
- some catalysts to enhance the reactions
- isotopic exchange
- radiolysis to decompose the organic molecules
- microorganisms
- …

- behavior of the organic species: sorption, diffusion?

- => Need for further experimental work

Questions and ideas for further experimental work - and related problems

- A long phenomenological chain from the steel to the biosphere; which phenomena are to be investigated?
- Is C-14 chemically similar to C-12 in steel => research work with radioactive substances.
- Accelerated corrosion is probably needed in the experiments, but how not to disturb the speciation of carbon?
- The reported experiments should be repeated with additional measurements.
- All the chemical species should be measured.
- If assumed to be organic => possible reactions and their kinetics (possible to experiment with inactive substances).
- Long timescales characterize the repository conditions => how to accelerate the experiments.
- Research within NKS?

Conclusions

- C-14 in activated steel components may be one of the major dose contributors in the decommissioning waste.
- Chemical speciation of C-14 in repository conditions is not completely clear.
- Organic species may be significantly more mobile than inorganic ones.
- The problem is characterized by a long phenomenological chain from the steel to the biosphere.
- Further research is necessary to reveal the main chemical reactions and their kinetics.
- A more detailed – and possible less conservative – modelling can be applied only if the phenomenology is sufficiently well known.

Thank you for your attention!