Used Fuel Container Designs and Lifetime Prediction

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USED FUEL CONTAINER DESIGN AND LIFETIME PREDICTION

- Adaptive Phased Management (APM), involves encapsulating the Canadian used nuclear fuel in durable containers and sealing the containers in a deep geological repository built in either crystalline or sedimentary rock.
- NWMO's R&D work on used fuel container development includes studies on the performance of corrosion barrier materials in repository environments.
- This presentation reviews and summarizes the results of research studies on the corrosion behaviour of copper and carbon steel as used fuel container materials and discusses the implications for container service life.

example deep geological repository concept

in-floor configuration

- Capacity: 3.6 million bundles
- Repository Area: ~1.5 x 1.5 km
- Depth: 500 m
- Container Capacity: 360 fuel bundles
- Container Placement Configuration: in-floor borehole

The containers are placed vertically inside boreholes drilled in the room floor, surrounded by highly compacted bentonite blocks.



copper used fuel container design



container design concept

- Capacity: 360 CANDU fuel bundles in three baskets containing 120 bundles each
- Corrosion barrier: 25 mm thick copper outer vessel.
- Structural support: 102 mm thick carbon steel inner vessel.
- Maximum external load: 45 MPa
- Design life: >100,000 years

An alternative UFC design of similar geometry, consisting of a single carbon steel vessel is also being evaluated.



Used Fuel Container

for the Used Fuel Container



uniform corrosion

At RH values higher than ~ 60% a thin water film will form on the copper surface, and metallic copper can be oxidized to ionic forms, Cu⁺, Cu²⁺. This process can occur if copper donates electrons to a receptor in the surrounding environment, such as oxygen molecules (O_2).

$$2Cu^{0} + \frac{1}{2}O_{2} \rightarrow Cu_{2}O$$
⁽¹⁾

Under anoxic conditions, corrosion could proceed as per the equations below, however, these reactions are thermodynamically not sustainable.

$$2Cu^{0} + H_{2}O \rightarrow Cu_{2}O + H_{2}$$

$$(2)$$

$$Cu^{0} + H_{2}O \rightarrow CuOH + \frac{1}{2}H_{2}$$
(3)



uniform corrosion (continued)

Under anaerobic conditions, copper corrosion would need impurities such as sulfide ions (HS⁻) to sustain the process:

$$2Cu^{0} + H^{+} + HS^{-} \rightarrow Cu_{2}S + H_{2}$$
(4)

The unlikelihood of reactions (2) and (3) has been challenged by two Swedish scientists. Gunnar Hultquist and Peter Szakálos (from the RIT). Their findings suggest that, contrary to thermodynamic predictions, water can act as an oxidant for copper [5]. However, these findings have been contested by other scientists and are the subject of ongoing study.



effect of salinity

Corrosion of copper under the effect of salinity is often assessed based on thermodynamic equilibrium, but thermodynamic data for high chloride concentrations are quite limited. The high salinity of groundwaters in Canadian sedimentary rock makes it necessary to conduct further research. A program with that objective is under development.

Based on existing data, the maximum wall loss on a copper container caused by uniform corrosion in sedimentary rock can be estimated at of 0.17 mm over a period of 10⁶ years. This is a conservative estimate, assuming a high initial oxygen inventory and not taking credit for oxygen consumption by competing processes.



localized corrosion

- Several studies have concluded that under repository conditions copper would not undergo pitting corrosion but rather a surface roughening process caused by the temporary separation of anodic and cathodic sites.
- The hypothetical effect of pitting of the copper container surface was assessed using a statistical extreme-value analysis that yielded an estimated maximum pit depth of 6 mm after 10⁶ years.
- Using the surface roughening model, considered more realistic for a deep geological repository environment, a 0.1 mm wall thickness loss is predicted over the same period.

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corrosion in compacted bentonite



Initially aerated Na-Ca-CI synthetic groundwater (~1 mol/L Cl⁻) 50:50 sand:bentonite, $\rho_d = 1.67$ Mg m⁻³, 50°C, 733 days (SKB TR-01-23, AECL-10397, Corros. Sci. <u>33</u> (1992) 1979)



stress-corrosion cracking

- Research results suggest that in a deep geological repository SCC is unlikely under aerobic conditions since the pre-requisite conditions of corrosion potential, interfacial pH, and SCC agent concentration do not exist simultaneously at the container surface.
- Based on the nature of the repository environment, SCC does not appear to be a threat to the integrity of copper used fuel containers and is excluded as a process that may cause container failure of a copper UFC in a deep geological repository.
- In addition to this low risk, suitable engineering provisions can be applied to further minimize the probability of SCC attack on copper, making it unlikely to be a significant corrosion mechanism.



microbiologically influenced corrosion

- Canadian experimental studies of microbial activity show that water activity below 0.96 effectively limits microbial culturability, that swelling pressures above 2 MPa reduce bacteria survival and that, overall, the repository environment can suppress microbial activity in the bentonite.
- Microbial activity may induce SCC of copper since bacteria can produce sulphides, ammonia, nitrite and acetate ions. Therefore to assess the impact of MIC it is important to predict where and when microbial activity will occur.
- A large body of evidence indicates that in highly compacted bentonite microbial activity either does not occur or proceeds at a very slow rate.
 MIC rates based on sulfide transport rates yield an estimated wall thickness loss of 1 mm over a period of 10⁶ years.



effects of radiation fields on corrosion

- Corrosion of the copper UFC may be influenced by gamma fields at the container surface via radiolysis of the moist air, producing oxidizing and reducing radicals and molecular species, like O₂, H₂O₂ and O₂-. The reducing species include H, e⁻ and H₂.
- Studies on effects of γ-radiation on the corrosion of copper conducted in Canada, Switzerland and USA suggest no detrimental effects. On the contrary, results indicate that for dose rates in the range of 10-100 Gy.h⁻¹ a more compact protective surface film is formed, which results in a lower corrosion rate.

carbon steel uniform corrosion

In aerobic environments, the anodic dissolution of Fe is supported by the cathodic reduction of either O_2 or H_2O .

$$4Fe + 3O_2 + 2H_2O \rightarrow 4FeO(OH)$$
(5)

As the environment becomes anoxic, the Fe(III) corrosion products are expected to convert to Fe(II) via Reaction (8) (Schikkor reaction). The overall reaction is expression (9).

$$2FeOOH + Fe + 2H_2O \rightarrow 3Fe(OH)_2 \tag{6}$$

carbon steel uniform corrosion - 2

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Fe + 2H_2O \rightarrow Fe(OH)_2 + 2H_2  (7)
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 $Fe(OH)_2 \rightarrow Fe_3O_2 + 2H_2O + H_2$ (8)

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3Fe + 4H_2O \rightarrow Fe_3O_4 + 4H_2
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The corrosion rate at this stage is controlled by the rate of film growth, with magnetite (Fe_3O_4) films forming in aqueous solution and carbonate-containing Fe(II) films forming in compacted bentonite.

(9)

carbon steel localized corrosion



- Carbon steel may be susceptible to some degree of localized attack, especially in high-carbonate groundwaters. This is more likely to occur under aerated conditions. Similarly to copper containers, the carbon steel containers are expected to experience surface roughening rather than distinct pitting.
- Using conservative assumptions it is estimated the carbon steel used fuel container may have a wall thickness loss of 8-24 mm due to localized corrosion through the repository aerobic phase.

- SCC
- MIC
- hydrogen generation and transport
- hydrogen degradation of carbon steel
- corrosion induced by radiation

The above mechanisms are treated in a similar manner as for the copper containers, having similar consequences as those observed for the copper UFC environment. Only the major differences are addressed here.

copper container lifetime prediction



- Through its lifetime, the copper UFC is assumed to be subject primarily to uniform corrosion and pitting. The extent of uniform corrosion is conservatively assessed using mass-balance calculations. Other processes, such as SCC and MIC, are considered to be unlikely in the repository environment
- Corrosion caused by the initially trapped O₂ is equivalent to ~80 μm of wall penetration. Corrosion due to sulphide, either from the groundwater, from pyrite impurities in the clay, or from microbial activity accounts for an additional 0.27 mm of corrosion over a period of 10⁶ years.
- The extent of pitting is assessed using a pitting factor, with a value of between 2 (considered realistic) and 5 (considered to be conservative). Therefore, in total, the maximum wall penetration is predicted to be from 1.5-2.0 mm after 10⁶ years.

c-steel container lifetime prediction

• Uniform corrosion:

Long-term anaerobic corrosion rate as measured in on-going experiments is ~1 μm /year

- Localized corrosion during aerobic phase:
 8-24 mm depending upon assumed aerobic phase duration
- Conservative estimate of the total corrosion depth in 10,000 years: ~35 mm of the proposed 100 mm wall thickness
- Confidence in prediction results derives from:
 - an understanding the repository environment evolution
 - predicted benign long-term conditions with slow uniform corrosion
 - corrosion data supported by archaeological evidence
 - an extensive database for assessing localized corrosion



wall thickness loss due to corrosion

Repository Phase	Corrosion rate	Thickness loss in	Thickness loss in
		10,000 yrs	100,000 yrs
Aerobic	20-200 µm/a (uni.)	9-34 mm	9-34 mm
(~ 50a)	160-480 µm/a (loc.)		
Unsaturated, anaerobic	0.01-0.1 µm/a	0.1-1 mm	0.1-1 mm
(10,000 a) Saturated, anaerobic	0.1- 1.0 µm/a	0 mm	9-90 mm
(> 10,000 a) Total Thickness Loss		~ 9-35 mm	~18-125 mm