Long-term integrity of copper overpack – Final report 2008

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Summary

The microstructural changes during low temperature, low stress creep experiments outside the welds appear to be largely restricted to the widening recovery zones of grain boundaries, without change in crystallographic orientation between the zone and the adjacent parent grain. Also a sign of grain boundary cavitation has been observed. In spite of the local stress concentration at the notch tip no creep acceleration has been observed.

Much higher creep cracking rate and lower creep ductility have been confirmed in OFHC copper than in OFP copper, but some grain boundary damage indications have been found also in the latter in multiaxial (CT) testing at 175°C. Longest CT tests for OFP copper have reached the 20000 h mark, and longest uniaxial tests have exceeded 63100 h (7 years, continuing). The new test results together with updated material and FE models will be used to update the creep life assessments for the assumed future service history of the copper overpack. In the combined corrosion and creep testing with welded CT specimens in the aerated transient conditions using simulated Olkiluoto groundwater only local pitting corrosion has been observed. The test series will be continued.
Preface

This report provides a preliminary final report of the project “Long term integrity of copper overpack” (LEIA), including and summarising the experimental, modelling and life assessment activities as well as the results and status of the project up to January 2009. The project is a part of the Finnish national research program on nuclear waste management, 2006-2010 (KYT2010). The financial support by this program is gratefully acknowledged.

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Authors
## Contents

Preface ...........................................................................................................................................2

1 Introduction .................................................................................................................................4
   1.1 Background .........................................................................................................................4
   1.2 Objectives .........................................................................................................................4

2 Materials and methods ..............................................................................................................5

3 Results .......................................................................................................................................6
   3.1 Uniaxial testing - general .................................................................................................6
   3.2 Multiaxial testing - general ...............................................................................................6
   3.3 Metallography ......................................................................................................................9
   3.4 Creep modelling and life assessment .................................................................................13

4 Discussion ...............................................................................................................................19

5 Conclusions and summary ......................................................................................................20

References ....................................................................................................................................20
1 Introduction

1.1 Background

The current concept of managing spent nuclear fuel in Finland and Sweden involves encapsulation of the fuel in metallic canisters that are placed into a deep underground repository [1]. The required design life is of the order of glaciation cycles to reduce the radioactivity of the contents close to the background level. The temperature of the canister is expected to peak at about 100°C during the first 1000 years, with gradual cooling to the level of the bedrock environment. For the protective copper (Cu-OFP) overpack of the canister, creep and corrosion are included as potential damage mechanisms under the repository conditions [2, 3]. Although relatively mild in usual engineering terms, the repository conditions imply a significant challenge to the life estimates for such damage mechanisms, as the expected design life is one to four orders of magnitude longer than for ordinary engineering structures designed against these mechanisms.

This work is dealing with both damage mechanisms in an effort to provide a realistic model for life prediction and long term behaviour of the copper overpack. This research particularly includes

- assessment of damage mechanisms and their interaction: primary creep, damage interaction in groundwater, impact of oxidation and thermal degradation
- material and life modelling of creep, damage and corrosion, and their combined action; and
- evaluation of long term materials properties of the welded copper overpack, and the expected impact on the overpack life in the repository.

The project is a part of the Finnish national research program on nuclear waste management 2006-2010 (KYT2010). The project also includes specific issues requested and defined by SSM (formerly SKI, Sweden).

1.2 Objectives

The technical objectives for the current period of the project (2007-2008) are:

- to extend the experimental database well beyond 10000 h of testing, to support modelling and life assessment
- to clarify the character and significance of the observed material changes (thermal and other) in the experiments
- to develop a realistic description of the expected creep damage as well as creep strain and life modelling under relatively low temperatures (about 100°C) and very long hold times; and
- to provide a well justified life prediction for the welded copper overpack under the final repository conditions.
2 Materials and methods

The OFP copper material for the experiments on the friction stir welded (FSW) test material was obtained from a full scale section provided by SSM/SKI (Sweden) and the Swedish program for canister studies. This section included a cylindrical part (material code T31) and a cover lid (lock TX 82) welded together and marked KL 059 on the outer surface of the cover lid. Chemical analysis, short term mechanical properties and initial microstructures of the materials have been reported previously in [2]. In addition, a batch of low-phosphorus copper (Cu-OF) was added to the testing program to explore the effect of composition (phosphorus) on ductility and creep cracking. The OFP test materials (Fig. 1) were subjected to uniaxial and multiaxial (compact tension, CT) creep testing with and without a simulated Olkiluoto groundwater environment. The CT specimen notch for welded OFP copper applied the natural gap tip of the joint, while the notch for (parent) Cu-OF material used an EDM notch with a tip width of 0.3 mm. For testing CT specimens of OFP copper in aerated groundwater, a new testing facility was used with circulating medium at 90°C. Metallography using optical, scanning electron and FESEM/EBSD (TKK) microscopy has been applied for as-new materials and test specimens after testing periods. Interrupted testing has been applied for multiaxial testing to inspect for damage evolution. For load setting and interpretation of the results, life modelling with extended parametric and other techniques has been applied, including finite element (FE) analysis for the CT specimens [2-4].

![Fig 1. Extraction (Cu-OFP) of a) uniaxial and b) CT specimens for testing](image)

For creep modelling, the combined Wilshire and LCSP models have been applied and further developed [5-7] to support a robust FE analyses under non-homogenous stress and strain fields.
3 Results

3.1 Uniaxial testing - general

The planned uniaxial creep testing program has been completed, except for the specimen V1 (150°C/120 MPa) that has exceeded 63 100 h (1/2009, more than 7 years) of testing time with an elongation well beyond 10% true creep strain. However, the specimen has not yet reached the minimum strain rate position, and will be continued under the next KYT programme. The other creep test results on OFP copper have been reported previously [3]. The on-going uniaxial tests with the Cu-OF material have not yet show failures. All testing results have been used to support creep modelling and to set the initial loading levels in multiaxial testing.

3.2 Multiaxial testing - general

CT creep testing

The specimen CS3 with FSW was loaded at \( F = 6400 \) N, resulting in front face displacement (FFD) of 0.15 mm after the first testing cycle at 175°C. For this specimen initially the ratio \( a/w = 0.376 \), giving \( K_I = 40.3 \) MPam\(^{0.5}\). The corresponding (plane stress) Mises equivalent stress is 35.1 MPa. After each of the four testing cycles (at 5040, 10000, 15000 and 20000 h) the test was interrupted for microstructural examination of the tip region. Early initiation of grain boundary separation (grain boundary cracking) was observed at the tip region. The metallography inspection after the latest test interruption of the current test program after 20000 h has been carried out at TKK. This CT test continues to provide valuable information and therefore the test will be continued under the next KYT programme.

The CT specimen is shown in Fig 2. After 15000 h of exposure, the joint faces (“crack mouth”) had opened by about 0.03 mm for near zero crack growth.
Parallel CT testing with a similarly sized Cu-OF base material specimens at 175°C was initiated so that the initial mouth opening, corresponding to the first fast straining, was comparable to that in the tested Cu-OFP specimens. This was done to compensate for the lower creep strength level of Cu-OF. Nevertheless, in the first test a crack with brittle appearance started to grow relatively fast in less than 1000 h. Intergranular cracking to about 8 mm required a notch mouth opening of about 0.1 mm. In the second test reduced load, cracking with brittle
appearance and crack branching was observed after about 6200 h of testing, see Figs. 9 and 10.

**Combined creep and corrosion (CT) testing**

For combined creep and corrosion testing, the new testing facility has been applied (Figs 4 and 5). This facility consists of a loading frame built into a constant load creep testing machine, with the specimen grips and the CT specimen designed to allow for continuous monitoring of the load line opening. The specimen is inserted to a glass jar where the simulated Olkiluoto groundwater (aerated) is made to circulate at a temperature of 90°C.

The testing program aims for at least 5000 h of testing with similar interruptions as for testing in air. The test facility has been moved to another laboratory room and has been updated to minimise vapourisation. During the test the temperature and load line displacement are monitored continuously and the conductivity is monitored manually in order to control the concentration which will change as a result of vapourisation.

The first combined corrosion and creep testing with welded CT specimens in the aerated transient conditions using simulated Olkiluoto groundwater has been completed and only local pitting corrosion has been observed at the moment. The test series must be continued before any conclusions can be drawn regarding the possible corrosion effects.

![Image of testing facility](image-url)
3.3 Metallography

CT: Cu-OFP

The OFP copper CT-specimen with friction stir weld after interruptions at 10, 15 ans 20kh of testing was subjected to metallography in optical and scanning electron (EBSD) microscopes (Figs 6-8). Regarding the microstructural evolution in general, the earlier conclusion [3] was that the loaded grain boundaries show diffusion controlled recovery zones that grow with increasing time, temperature and stress (strain). This conclusion, however, has been questioned after the inspection after 20kh of testing. What appears to be a widening grain boundary zone could be due to the preparation of the sample. This would confirm the earlier notion from TEM results of much narrower grain boundary zones that the zone shares the crystallographic orientation with the originating grain.

With sufficient strain like in some of the longer term uniaxial tests, the grain boundaries will become decorated by a large number of small grains, i.e. the process results in recrystallisation. No formation of the recovery zones has been observed within regions of lower stress/strain, such as outside the tip region of the CT specimens. The boundary between the friction stirred region and base metal
can be seen in Fig. 6 in the form of an EBSD local misorientation map. Strain localization is seen in the weld zone. The mechanism is studied further at TKK [13].

![Local misorientation map at the FSW/BM interface (after 20000 h, main joint plane horizontal); local misorientation shown in green.](image)

Distinguishing between genuine grain boundary cavities and artefacts produced by the polishing process is difficult in copper, which is a soft material. In one case a genuine creep cavity has been detected about 1mm ahead of the crack tip, see Fig. 7 after 15000h of testing. The same location after 20000h is being inspected. The genuine nature of the cavity is confirmed by Fig. 8, where the local misorientation created by creep deformation can be seen.

Microcracking has appeared at the grain boundaries of the tip region as extensions on the main crack (joint). These microcracks did not exceed about 25 µm in length after 20000 h of testing time.
Fig 7. A grain boundary cavity after 15000h of testing in a grain boundary triple point.

Fig 8. The same location as in Fig. 7, EBSD local misorientation map. The color key is shown in the inset.
CT: Cu-OF

The first CT specimen test at the testing temperature of 175°C with base metal Cu-OF showed in only 1000 h creep cavitation, crack initiation and growth on the grain boundaries with brittle appearance, i.e. grain boundary damage with low strains of the grain interiors (Fig 9). The second CT-specimen at a reduced loading has also shown brittle type of creep crack initiation and early growth. The second specimen showed low ductility and, rather unexpectedly, remarkable crack branching as shown in Fig. 9. The metallographic examination (Fig. 10) confirmed that the cracking mechanism was grain boundary cavity initiation and growth.

Fig 9. Crack branching in the second Cu-OF CT specimen after a test of 6200 hours.
3.4 Creep modelling and life assessment

The classical rupture modelling produce unacceptably big errors when post assessed heat to heat. The new approach using the activation energy based Wilshire approach has here been implemented and utilised for modelling OFP canister copper. The main advantage of the model is its robustness and it is evidently superior in minimising data scatter. The model is also shown to be a good base for the new logistic creep strain prediction model. Both rupture and strain master curves have successfully been produced for the OFP copper and calculated minimum strain rates on public domain data verify the model.

The strain model has also been used for comparative life prediction at 50 MPa and 100°C that in terms of stress and temperature are is beyond recommended extrapolation range. The predicted life of 283 000 years is shorter than expected. The result is dependent on the activation energy driving creep and the used 104 kJmol⁻¹, value for boundary diffusion is very close to the optimal for the data fit. That the activation energy at intermediate temperatures (75-195°C) will be less than for self-diffusion was already stated in 1956 [9] for OFHC coppers, otherwise obeying the same general laws as for high temperatures creep.

It is however to remember that this thermal loading case refers to a upper limit for the canister outer wall and a more thorough assessment taking the whole predicted temperature distribution into account might produce life times meeting the required 1000000 years of life. It is also to remember that the case assumes a stress level that does not take into account local material properties (as in welds).
and multiaxial constraints lowering the creep ductility. These factors will again shorten the expected life.

Using the above model for creep rupture the creep strain model “Logistic creep strain prediction” (LCSP) method can readily be used for creep strain modelling. In its uniaxial form the LCSP predicts the full creep curve from the creep rupture time together with only three additional “shape” parameters.

The logistic creep strain prediction presentation of time to strain is defined as

\[
\log(t_e) = \frac{\log(t_r \cdot \alpha + C)}{1 + \left(\frac{\log(\epsilon)}{x_0}\right)^p} - C
\]

(1)

where \(t_e\) is time to strain, \(t_r\) is the true time to rupture or a predicted one acquired from a creep rupture model (as a function of stress and temperature, \(\epsilon\) the strain and \(x_0\), \(p\) and \(C\) are fitting constants (for each creep curve) defining the curve shape. The constant \(\alpha\) is for correcting strain at rupture towards the creep ductility \(\epsilon_R\).

Eq 2. can we rewritten in a form giving strain at specified time:

\[
\log(\epsilon_e) = \left(\frac{\log(t_r) + C}{\log(t_e) + C} - 1\right)^\frac{1}{p} \cdot x_0
\]

(2)

By differentiating Eq. (2) with respect to time, the resulting strain rate as a function of stress and temperature can be written as

\[
\dot{\epsilon} = -\epsilon \cdot k_1 \cdot k_2 \cdot x_0
\]

(3)

where \(\epsilon\) is given by Eq. (2), and

\[
k_1 = \frac{(LTF - 1)^\frac{1}{p}}{p}
\]

(4)

and

\[
k_2 = \frac{\log(\alpha \cdot t_r + \beta)}{(\log(t_r + \beta)^t \cdot t_r \cdot (LTF - 1))}
\]

(5)

An example for the modelled OFP-copper versus measured creep response is shown in Fig. 11.
<table>
<thead>
<tr>
<th>Time (h)</th>
<th>Strain (mm/mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>0.01</td>
</tr>
<tr>
<td>100</td>
<td>0.1</td>
</tr>
<tr>
<td>10^2</td>
<td>1</td>
</tr>
<tr>
<td>10^3</td>
<td>10</td>
</tr>
<tr>
<td>10^4</td>
<td>100</td>
</tr>
<tr>
<td>10^5</td>
<td>1000</td>
</tr>
<tr>
<td>10^6</td>
<td>10000</td>
</tr>
</tbody>
</table>

Fig. 11. Actual and predicted creep strain prediction for batch 500 [10] specimen at 200°C / 120MPa (upper figure), T17 specimen at 150°C / 120MPa (lower figure), the predicted line overlaps the measured data if the predicted rupture time is increased by a factor of 3.
Fig. 12. Measured vs. predicted time to engineering strain. Z=6

The OFP data was fitted to Eq. (1) as time to strain utilising the measured time to rupture. The resulting $p$ and $x_0$ parameters showed to be both stress and temperature dependent. A preliminary attempt has been done for parameter dependence as shown in Fig. 13 and the parameter values and functions listed in Table 1.

Table 1. Shape factors (Eq.2) of the creep strain model for OFP copper ($\sigma$ in MPa, $T$ in °C)

<table>
<thead>
<tr>
<th>Factor</th>
<th>$x_0(\sigma/\sigma_{TS},T)$</th>
<th>$p(\sigma/\sigma_{TS},T)$</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Value</td>
<td>-2.179+4.397\ln(\sigma/\sigma_{TS})-0.008\cdot T</td>
<td>7.235+0.460(\sigma/\sigma_{TS})\ln(\sigma/\sigma_{TS})-0.012\cdot T</td>
<td>3.5</td>
</tr>
</tbody>
</table>

To validate the acquired creep strain model minimum creep strain predictions were made for public domain data [11]. The calculated predictions correspond extremely well with the two OFP copper batches where the strain rates had been reported as seen in Fig. 14. Note that the shape functions should not be used for extrapolation beyond the range of data before confirmation of the creep shapes has been properly validated.
Fig. 13. Creep curve shapes as a function of normalised stress and temperature.

Fig. 14. Creep strain model predicted minimum creep rates compared to reported ones [batch 400,500]
The capability of the model to predict well the strain rates can be taken to suggest inversely a fair ability to predict time to rupture from relatively early strain data of unfailed specimens. To test this, four points were selected from different creep curves (VTT tests) so that the points were at different locations in terms of life fraction. The creep strain rates at these locations were determined, and time to rupture was solved for the same strain rate at the specified time. In these tests the minimum strain rate occurred approximately at 30% life fraction. The results of the exercise are shown in Fig. 15.

![Fig 15. Predicted time to rupture from the observed strain rate (stars, at life fractions shown) in comparison with short-term rupture data (squares) and corresponding model (line); the life fraction value in brackets refers to a test interrupted at over 25 600 h.](image)

The strain rate related prediction holds great promise in predicting the creep response of the OFP copper towards the real canister temperature and stress states by testing at lower stresses aiming at long durations of testing with small accumulated strains. The LCSP model can be considered as an equally suitable but simpler than the classical 0-model [12].

4 Life prediction at 50 MPa / 100°C

Extrapolating the above models for the OFP canister copper (corresponding to tensile and creep properties of T31 and T17 batches) the expected life at 50 MPa and 100°C requires extrapolation of model (see Fig. 16) to a normalised stress ($\sigma/\sigma_{TS}$) of just below 0.3. The corresponding predicted rupture time for this stress state at 100°C is 283 000 years, which is less than the specified safe life if 1000000 years is considered as a requirement. It is also considerably lower than earlier predicted for 10% strain using the minimum commitment method [4].
Discussion

The uniaxial creep testing program has been nearly finished except for one long term test (at 150°C/120 MPa) that has exceeded 63 100 h and continues to support creep modelling. Additional long term testing would still be useful, but the results from CT tests and earlier tests with nominally uniaxial but defective specimens [3] would suggest that multiaxiality could be more seriously life-limiting.

The FSW CT specimen creep test of Cu-OFP was initially loaded to front face displacement of 0.15 mm in the first testing cycle at 175°C. After three test cycles to 15 000 h the joint faces showed an (internal) opening of only 0.03 mm in cold relaxed state. Nevertheless, after 20 000 h the crack tip shows microcracking up to about 25 um in length on some grain boundaries that are roughly parallel to the main crack/joint plane. This suggests that further test cycles will be able to propagate cracks under the selected fairly low levels of loading. To compensate for differences in strength, parallel CT testing using Cu-OF base material specimen at 175°C was initiated so that the initial mouth opening was similar to that in Cu-OFP testing. However, the first test showed relatively fast creep cracking in less than 1000 h. Intergranular cracking to about 8 mm required a notch mouth opening of about 0.1 mm. The second test of 6200 h demonstrated transgranular creep damage and crack branching. The microstructural changes that have been observed seem to warrant some future TEM studies to clarify the details of the observed microstructures.

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**Fig 16.** The master curve with the OFP copper rupture data; the horizontal line corresponds the formerly common approximate max loading of the overpack.
For the first time, some microcracking appears at the grain boundaries of the tip region, mainly as extensions on the main crack (joint) plane. These microcracks do not seem to exceed about 25 µm in length after 20 000 h of testing time.

For combined creep and corrosion testing, a new facility was designed and constructed for loading CT specimens under groundwater environment. The main difference to the previous approach with internally pressurised vessel specimen is that the loading configuration will better allow for monitoring of the emerging damage. Although the test temperature is necessarily lower, it does approach the actual foreseen repository conditions, and the test facility has so far shown no sensitivity to disturbance by deposits.

6 Conclusions and summary

The microstructural changes during low temperature, low stress creep experiments outside the welds appear to be largely restricted to the widening recovery zones of grain boundaries, without change in crystallographic orientation between the zone and the adjacent parent grain. Also a sign of grain boundary cavitation has been observed. In spite of the local stress concentration at the notch tip no creep acceleration has been observed. Much higher creep cracking rate and lower creep ductility have been confirmed in OFHC copper than in OFP copper, but some grain boundary damage indications have been found also in the latter in multiaxial (CT) testing at 175°C. Longest CT tests have reached the 20000 h level, and the longest uniaxial test has exceeded 63 100 h (7 years, continuing). The new test results together with updated material and FE models will be used to update the creep life assessments for the assumed future service history of the copper overpack. Combined corrosion and creep testing with welded CT specimens continue in the aerated transient conditions using simulated Olkiluoto groundwater. So far only local pitting corrosion has been observed.

References


12. Dutton, R., A methodology to analyze the creep behaviour of nuclear fuel waste containers. AECL-11249, COG-95-06