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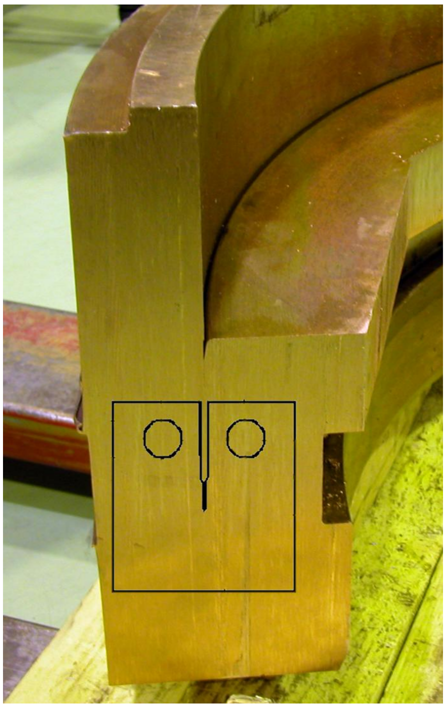

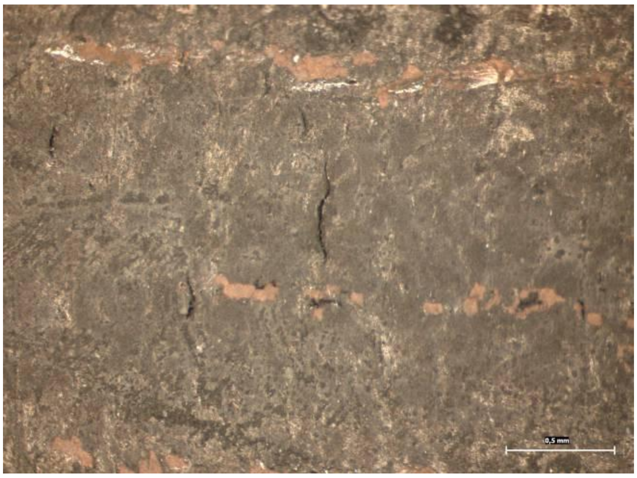

KYT 2010 / PIKE

Long-term integrity of copper overpack
– Final report 2009

Authors: Juhani Rantala, Jorma Salonen, Pertti Auerkari, Stefan Holmström / VTT
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Summary

The microstructural changes during low temperature (175°C), low stress creep experiments on base material and FSW welds of OFP copper appear to be largely restricted to the widening recovery zones at stressed grain boundaries, and to the increasing rate of grain boundary creep cavitation at the tip of multiaxial (CT) creep testing specimens at 175°C after 25000 h. This cavitation appears to be related to local strain, as demonstrated by the concurrently increasing rate of notch/crack tip opening.

Clear extension of creep cracking with low creep ductility has been confirmed in OFHC copper (up to 8000 h), while much higher creep ductility has been retained in OFP copper so far. Longest CT tests for OFP copper have reached the 25000 h mark, and the longest duration of the uniaxial tests has exceeded 70000 h (8 years, continuing) but with some surface cracking. The test results together with an updated material model have been applied in FE to characterise the effects of multiaxial stress at the crack tip. The optimised assessment with a somewhat reduced apparent activation energy predicted a safe creep rupture life of 43000 ± 9000 years at 100°C/50 MPa, which is significantly less than in earlier assessments. In the combined corrosion and creep testing with welded CT specimens immersed in aerated simulated Olkiluoto groundwater at 90°C, only limited grain boundary corrosion has been found at the notch edges after 1900 h of testing. So far, no indications have been observed of significant localised environmental damage, such as stress corrosion cracking.

Confidentiality	Public

Espoo 29.1.2010

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Preface

This report provides the final report of the project “Long term integrity of copper overpack” (PIKE), including and summarising the experimental, modelling and life assessment activities as well as the results and status of the project up to December 2009/January 2010. 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.

Espoo, January 29, 2010

Authors

Contents

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 principal objectives of the project are

- to determine experimentally and model the long term mechanical (creep) behaviour of the copper overpack, including effects of low stresses, multiaxiality, defects and reduced ductility; and
- to determine experimentally and model the combined creep/corrosion impact of the expected oxygen potential transition on the predicted life of the overpack.

The particular technical objectives for the year 2009 have been:

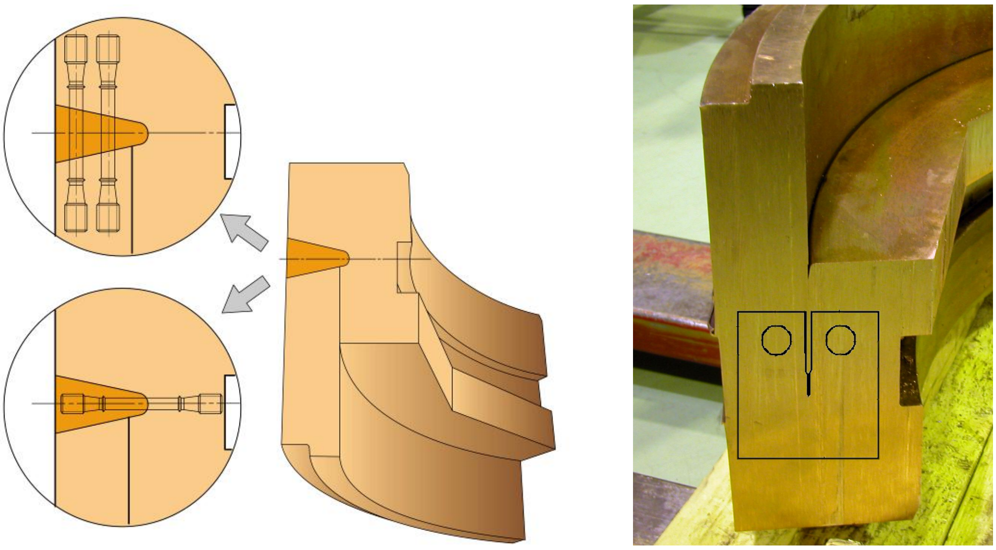
- to clarify the impact of low stresses for uniaxial and multiaxial creep, and to extend the multiaxiality effect of accelerated damage at low stress regime for Cu-OFP and the model material Cu-OFHC

- to investigate the detailed mechanisms of the observed microstructural changes in low-temperature and low stress creep of OFP copper
- to conduct extended CT specimen testing with real weld joints of OFP copper (above 100°C for creep, about 90°C for combined creep/corrosion) for damage assessment and to support mechanical modelling; and
- to conduct extended FEA for transferring the improved materials models to the structural analysis and to improve the corresponding models for the impact of welds and defects.

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 (HUT) 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].

For creep modelling, the combined Wilshire and LCSP models have been applied and further developed [5-7,15] to support robust FE analyses under non-homogenous stress and strain fields.



a) b)

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

3 Results

3.1 Uniaxial testing - general

The planned uniaxial creep testing program of OFP copper has been completed, except for the specimen V1 (150°C/120 MPa) that has reached 70000 h (8 years) of testing time and a true strain well beyond 10%. At 63760 h the test was interrupted for visual inspection and physical measurements. At that point small surface cracks were observed (Fig. 2), without appreciable necking of the specimen. After inspection, the test was continued at the same loading conditions as before. The creep test 5K at 250°C/80 MPa has been completed after 7837 h of testing. The test K3 at 200°C/70 MPa is running at 14650 h. One uniaxial test has been completed for the Cu-OF material and another one is on-going. All testing results have been used to support creep modelling and to set the initial loading levels in multiaxial (CT) testing.

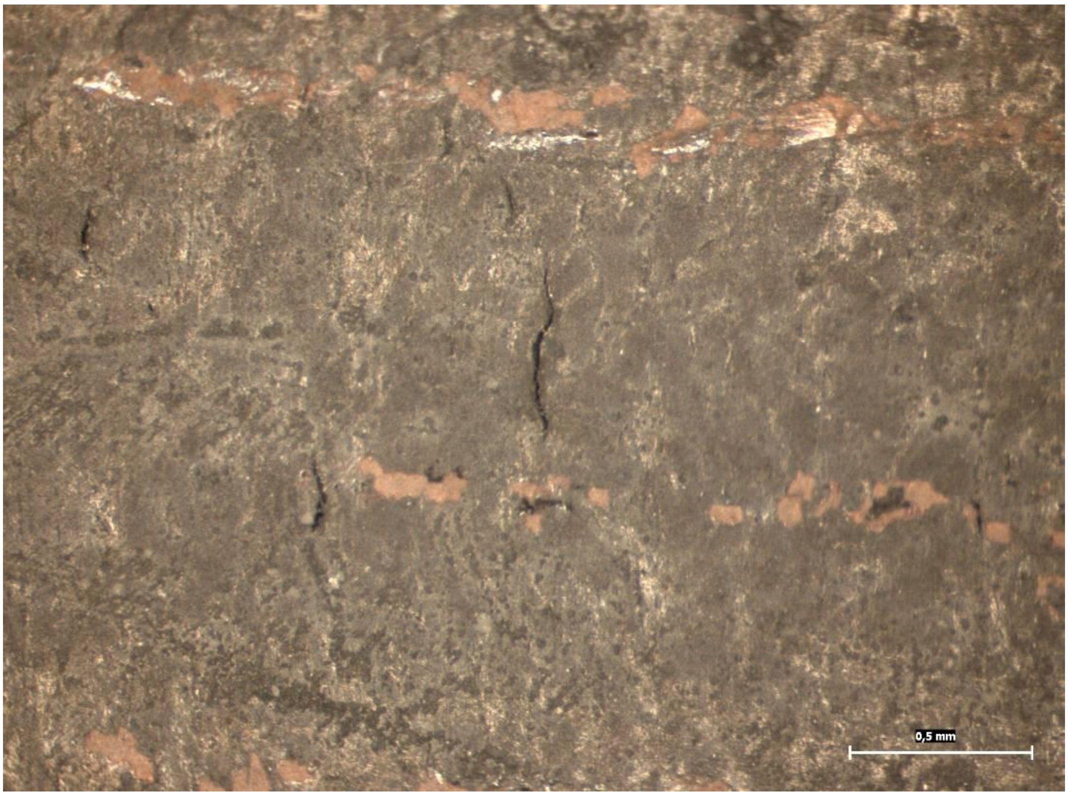


Fig. 2. Surface cracks in the uniaxial specimen V1 after 63760 h at 150°C.

3.2 Multiaxial testing - general

CT creep testing

The specimen CS3 (OFP copper) with a FSW joint was initially 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 five testing cycles (5000 h each, up to 25000 h) the test was interrupted for microstructural and damage 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 25000 h has been completed at HUT. The next test interruption will be done after an additional 5000 h of testing when the specimen has reached a total of 30000 h in August 2010.

The CT specimen is shown in Fig 3. After 25000 h of exposure the joint tip has opened about 0.13 mm, indicating local deformation at the notch tip, see Fig. 4. After 15000 h of exposure, the joint faces (“crack mouth”) had opened by about 0.03 mm for near zero crack growth. Although the opening was measured at a different location at 15000 h and at 25000h it is obvious that the opening process is now accelerating.

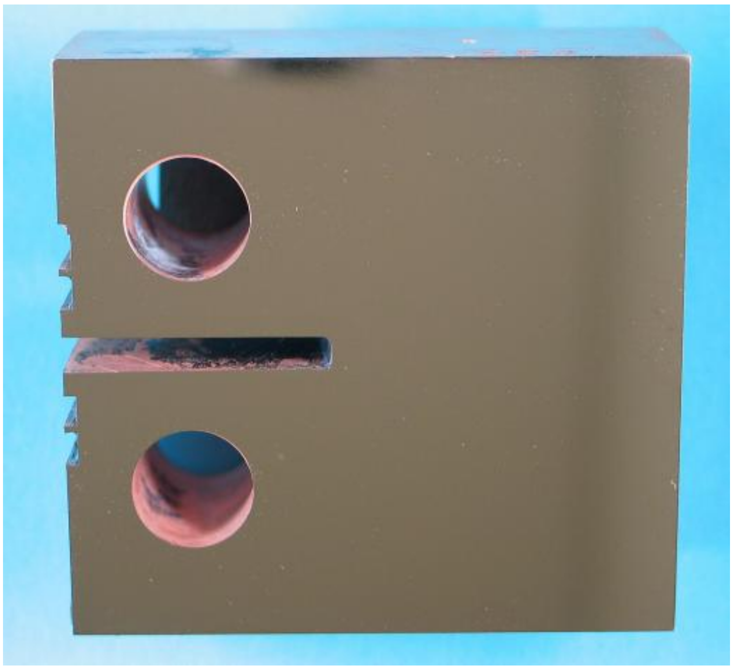


Fig 3. CT-specimen (CS3, OFP) after the first test cycle (175°C / 35.1 MPa)

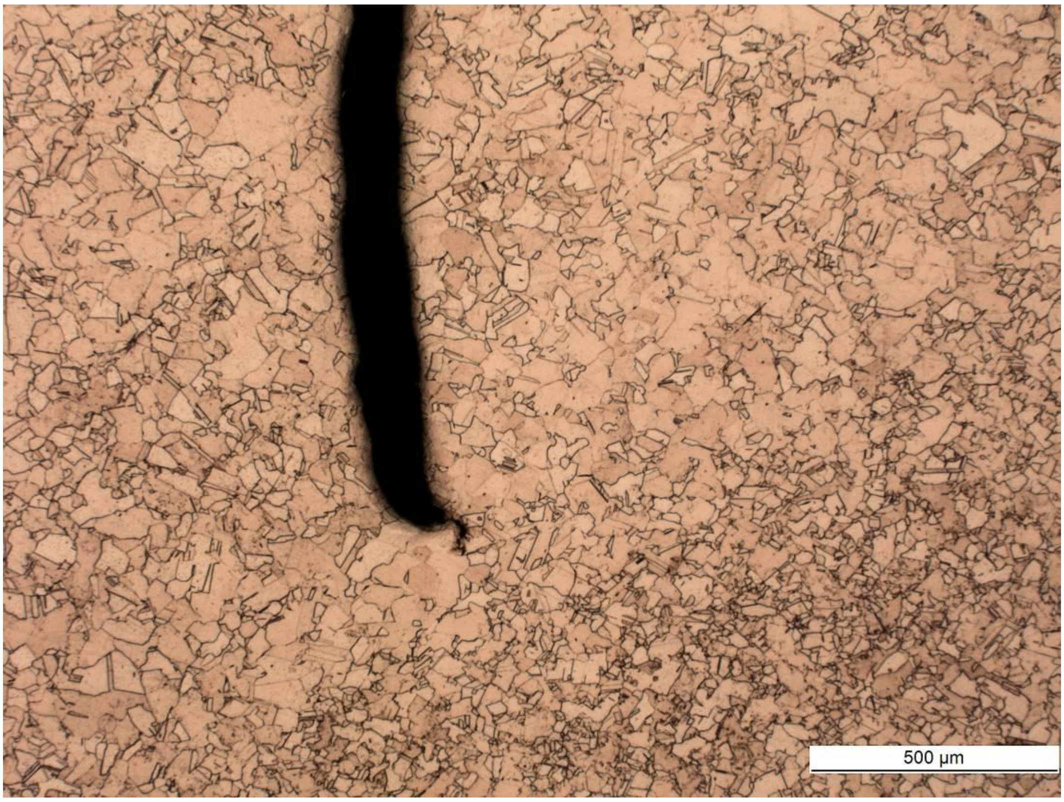


Fig 4. FSW joint tip and the FSW/BM border line in specimen CS3 after 25000 h (175°C) of testing.

Parallel CT testing with a similarly sized Cu-OFHC 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 at 9 MPa√m. Intergranular cracking to about 8 mm required a notch mouth opening of only about 0.1 mm. In the second test reduced load of 8.25 MPa√m, cracking with brittle appearance and crack branching was observed after about 6200 h of testing, see Figs. 14 and 15. The third test at 175°C and 7.75 MPa√m has been completed after 8334 h. A fourth test at 6 MPa√m is running at 710 h and is expected to last for more than 15000 h.

Combined creep and corrosion (CT) testing

For combined creep and corrosion testing, the new testing facility has been applied. 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 circulated at a temperature of 90°C.

The test facility has been designed to minimise evaporation, during which only water is evaporated and can be compensated for by adding distilled water. The conductivity of the salt water is monitored manually in order to control and maintain the salt concentration.

The second combined corrosion and creep testing with welded CT specimens in the aerated transient conditions using simulated Olkiluoto groundwater has been completed and the specimen has been inspected. In the first test of 227 h only local pitting corrosion was observed. After the second test of 1907 h grain boundary corrosion (intergranular attack) was observed at the both side faces of the specimen mainly in the middle of the notch, see Figs 5-6. As there was less corrosion towards the joint tip, Fig. 7, and no evidence of stress corrosion at the joint tip, it has been decided to continue the test for an additional 3000 h. After this additional exposure the test specimen will be sectioned for metallography in order to inspect possible corrosion along the crack front instead of inspections on the surface as has been done so far.

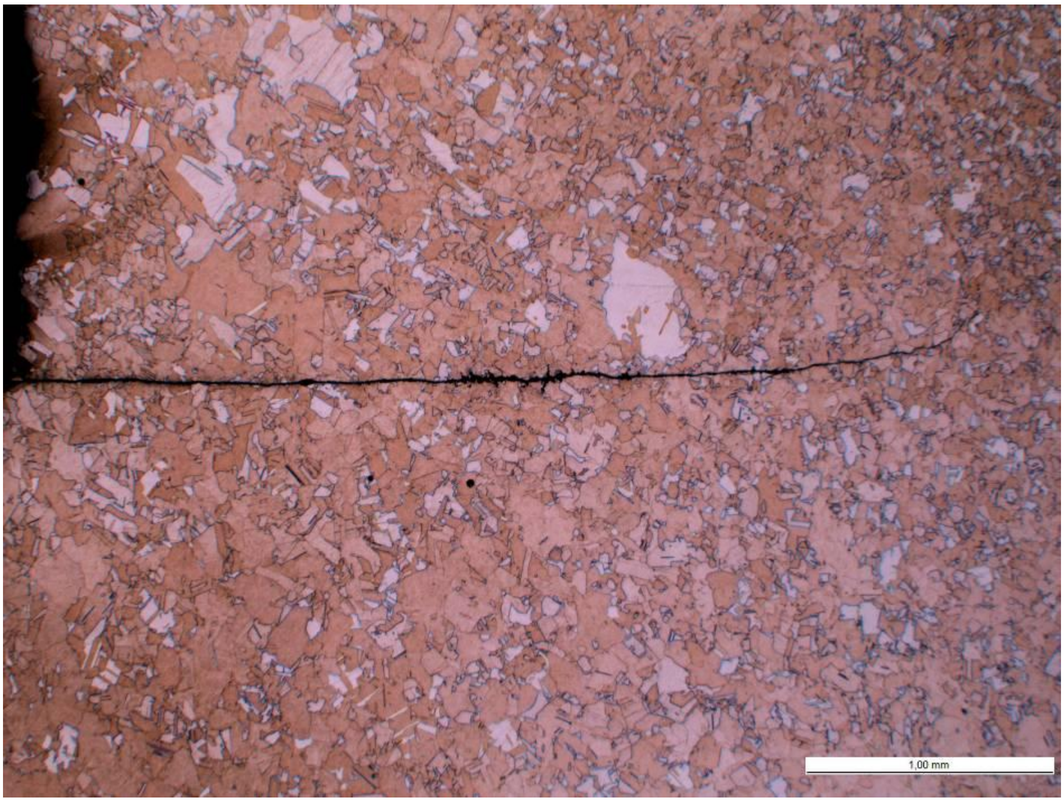


Fig. 5. The natural defect in specimen CS1 after 1907 h of testing in salt water at 90°C.

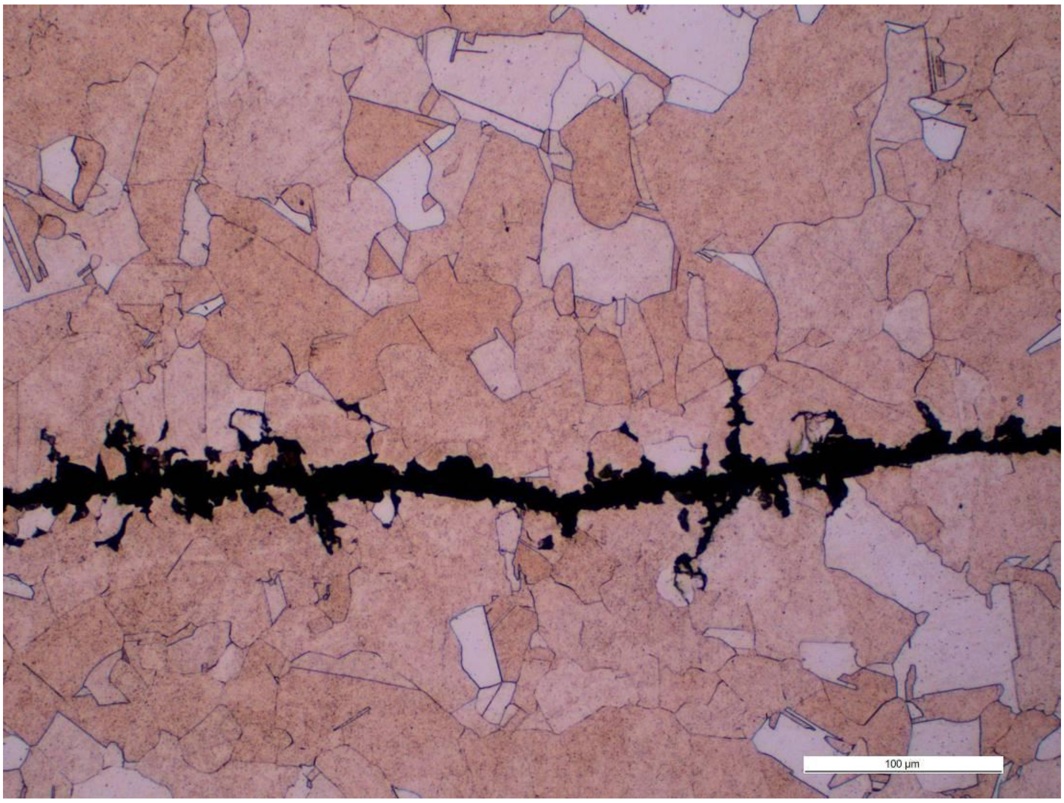


Fig. 6. A detail of Fig. 5 from the middle of the notch. Intergranular attack at the grain boundary.

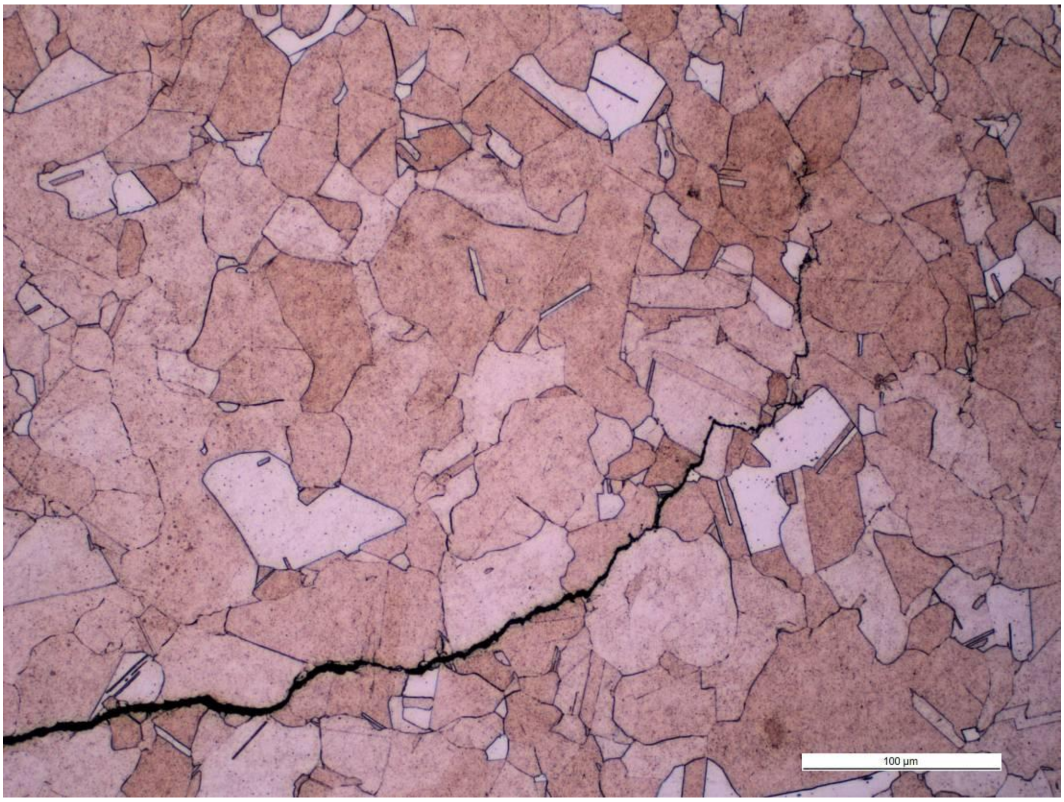


Fig. 7. A detail of Fig. 5 from the region at the joint tip.

3.3 Metallography

CT: Cu-OFP

The OFP copper CT-specimen with friction stir weld after interruptions at 10, 15, 20 and 25kh of testing was subjected to metallography with optical (Fig. 8) and scanning electron (EBSD) microscopes (Figs 9-13). Regarding the microstructural evolution in general, the earlier conclusion [3] was that the loaded grain boundaries show diffusion controlled recovery zones as shown in Fig. 8 that grow with increasing time, temperature and stress (strain). During the latest inspection the amount of these zones was found to have increased. The nature of the grain boundary zones which appear like ridges in Fig 9 needs further investigations during the next interruption after reaching 30000 h of testing.

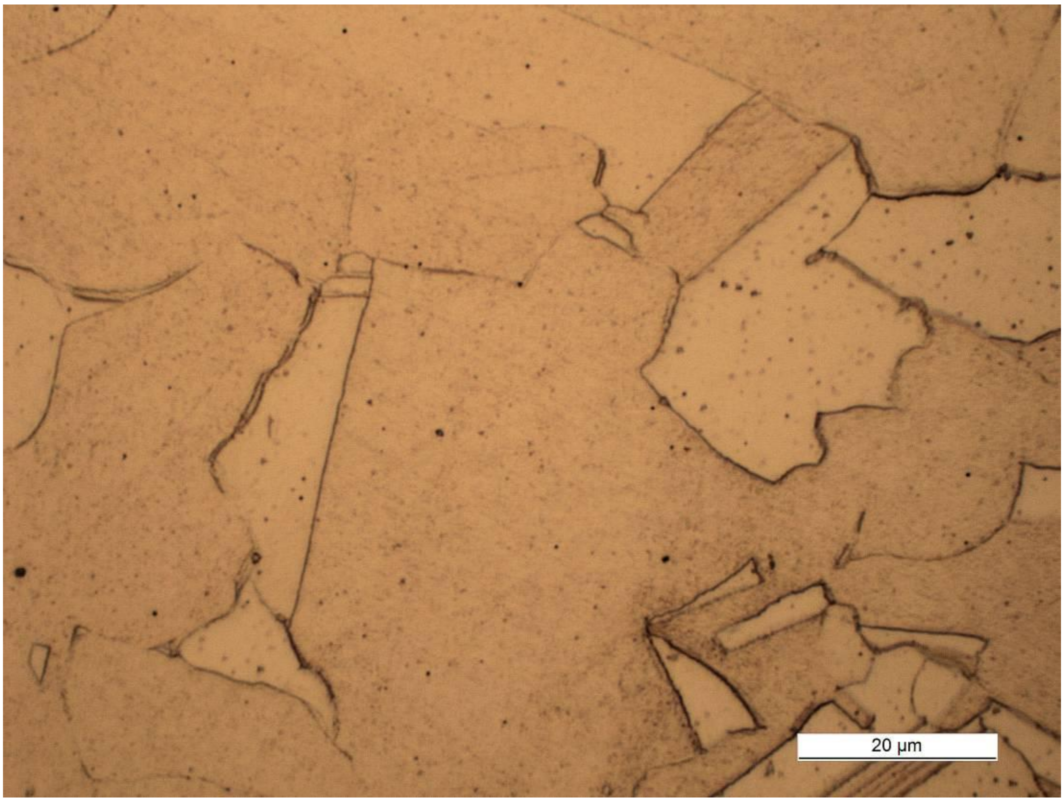


Fig. 8. The effect of grain boundary widening near the joint tip in the specimen CS3 in optical microscopy.

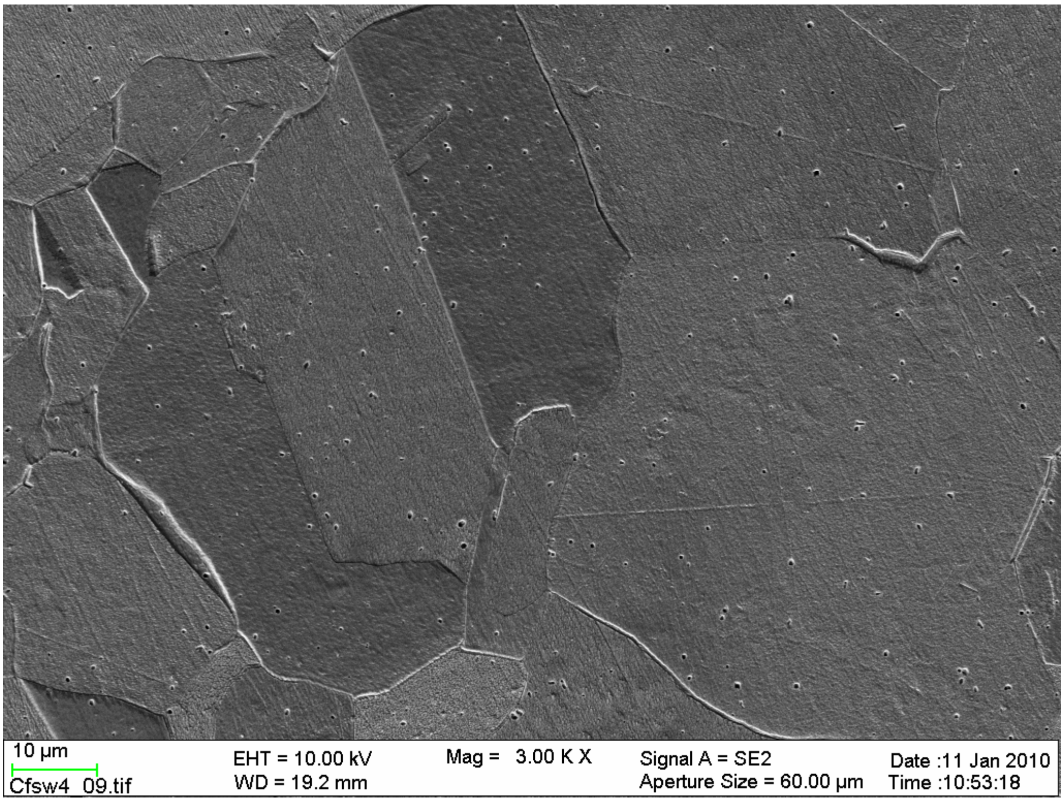


Fig. 9. An example of grain boundary widening near the joint tip in the specimen CS3 in scanning electron microscopy.

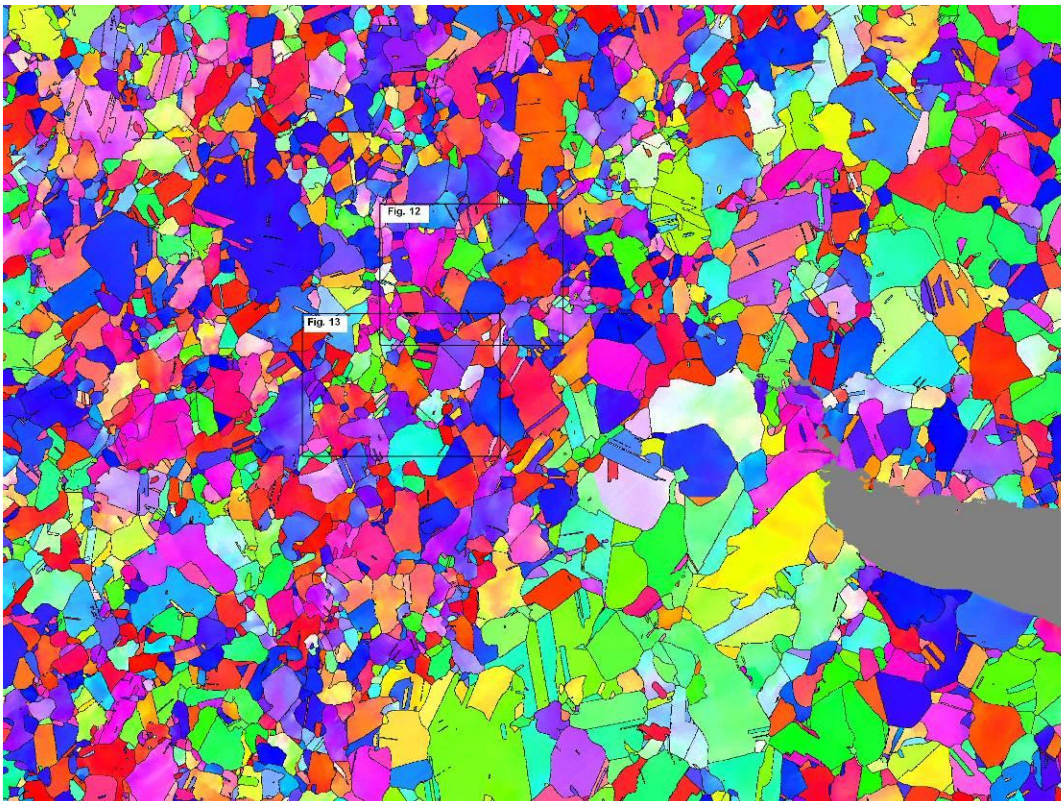


Fig. 10. EBSD grain orientation map IPF-Z, with locations of Figs 12-13 shown. The total width of the image is 1.07 mm; crack tip region shown in light grey.

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 at the joint tip can be seen in Fig. 10 in the form of an EBSD grain orientation map after 25000 h. The location of Figs. 12-13 is shown in Fig 10. Strain localization in the weld zone is shown in Fig. 11. These mechanisms have been elucidated further in a previous paper [13].

Grain boundary cavitation has been observed in front of the notch tip and the cavity density is clearly increasing when approaching the joint tip region. In the inspection at 20000 h only a single cavity was detected, which means that the damage mechanism has now reached a phase where cavity density starts to increase rapidly. Cavities can be seen on the grain boundaries in Fig. 12. A nearby location is shown in an EBSD misorientation map in Fig. 13, which shows that the deformation is concentrated around the cavities. The cavity density drops markedly after a distance of 0.75mm ahead of the crack tip.

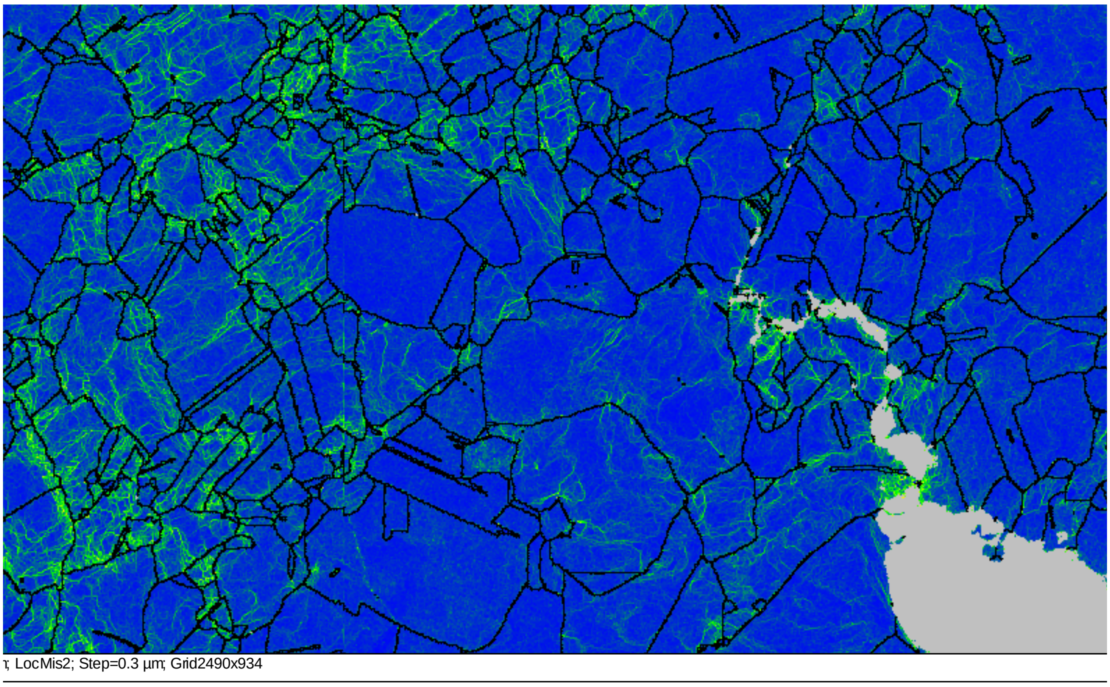


Fig 11. Local misorientation map at the FSW/BM interface (after 25000 h, main joint plane horizontal); misorientation shown in green. The width of the picture is 470 µm.

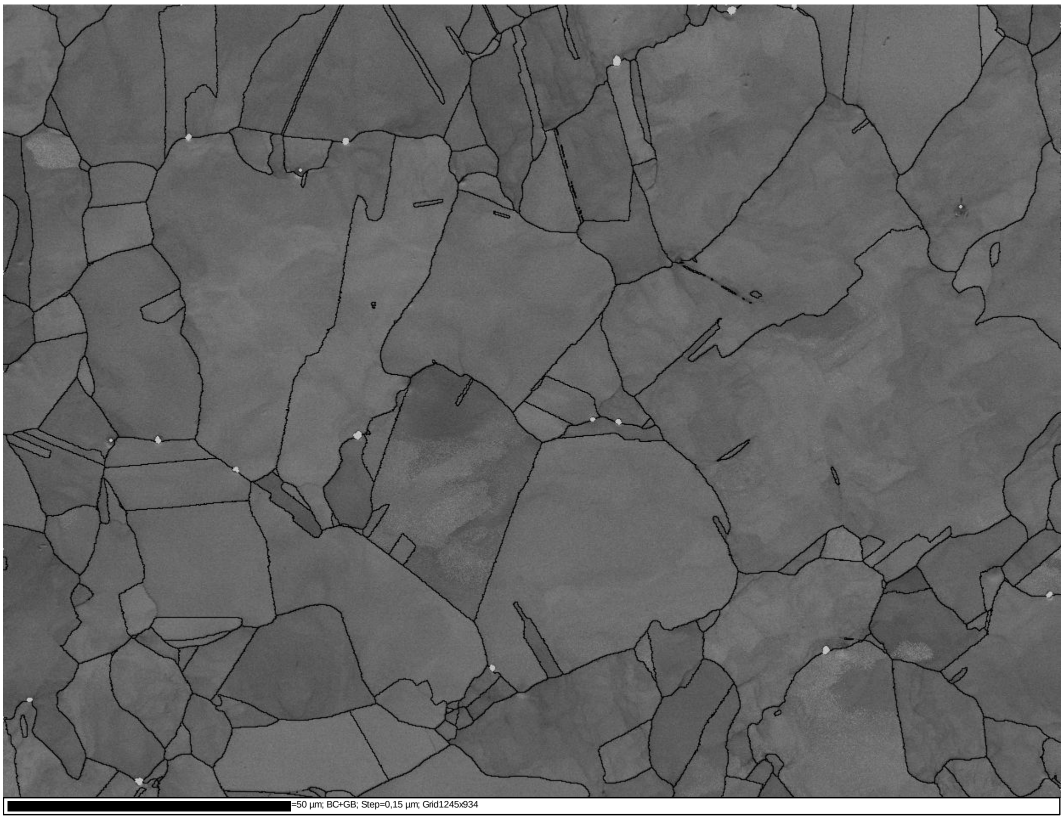


Fig 12. Grain boundary cavities near the crack tip of the CT specimen after 25000 h of testing (stir region); scale bar 50 µm; location indicated in Fig. 10.

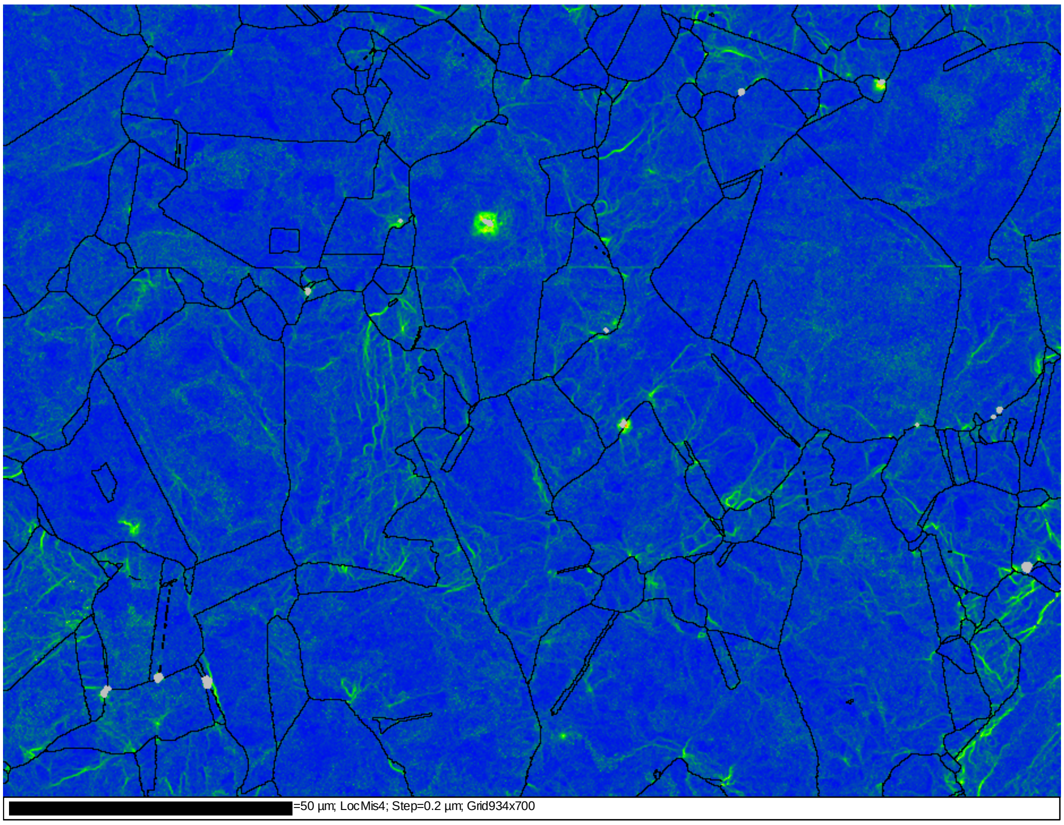


Fig 13. EBSD local misorientation map after 25000 h. Scale 50 µm. The location is indicated in Fig. 10.

CT: Cu-OFHC

The first CT specimen test at a load of 9 MPa√m at the testing temperature of 175°C with base metal Cu-OFHC showed in only 1000 h creep cavitation, crack initiation and growth on the grain boundaries with brittle appearance (Fig 14), i.e. grain boundary damage with low strains of the grain interiors. The second CT-specimen at a reduced loading of 8.25 MPa√m has also shown brittle type of creep crack initiation and early growth. The third test at 175°C and 7.75 MPa√m has been completed after 8334 h and has shown low ductility and remarkable crack branching as in Fig. 14, almost identical in appearance as in the previous test. The metallographic examination (Fig. 15) confirmed that the cracking mechanism was grain boundary cavity initiation and growth. Also twinning caused by deformation is seen in Fig. 15. A fourth test at 6 MPa√m is running at 710 h and is expected to last for more than 15000 h.

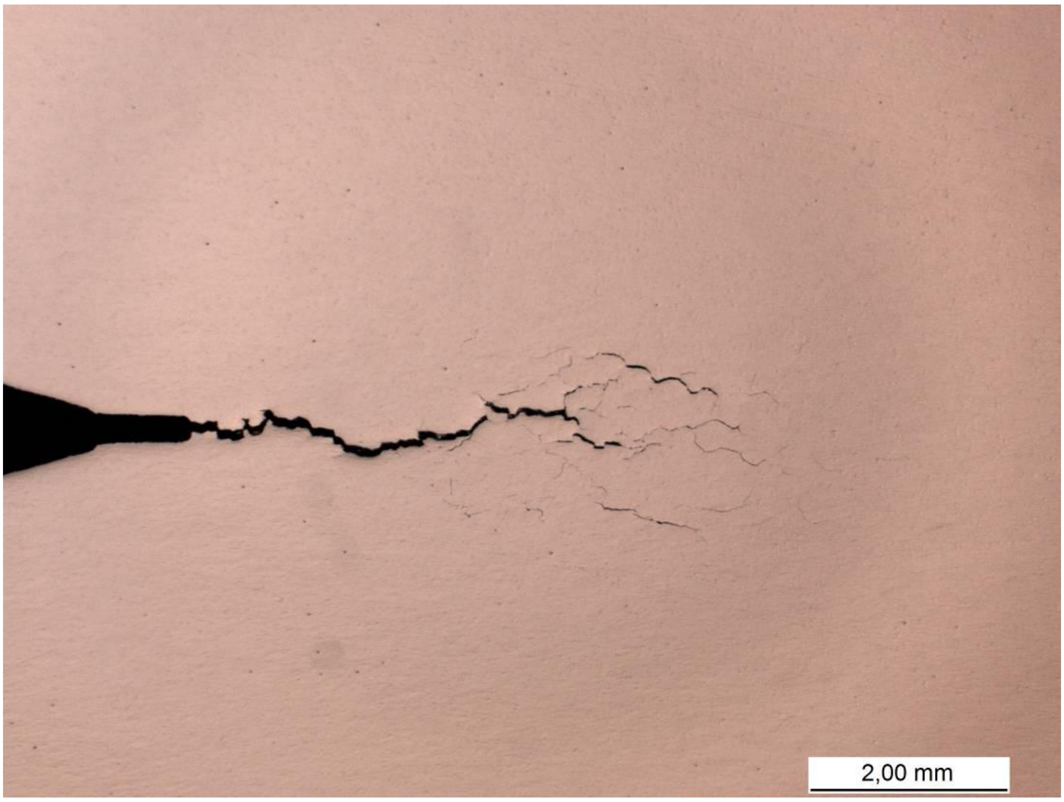


Fig 14. Crack branching in the third Cu-OF CT specimen after a test of 8334 hours.

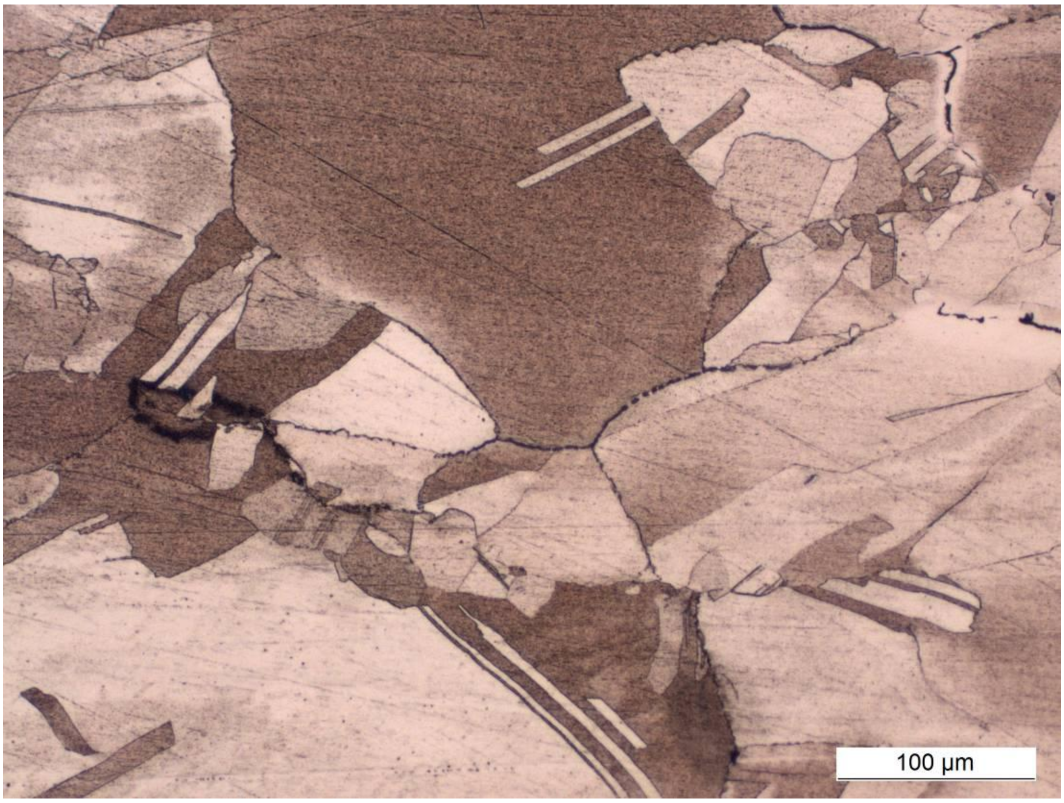


Fig 15. Grain boundary cracking in the third Cu-OF CT specimen after 8334 h of testing.

3.4 Creep modelling

The LCSP creep model developed at VTT has now been applied in FE analysis. Primary, secondary and tertiary creep terms are included in the analysis. In Fig 16 the strain distribution of a CT specimen is shown. The analysis was then extended to characterise the state of multiaxiality at the crack tip in order to apply the LICON methodology to the life prediction. The distribution of the multiaxiality parameter H (= max. principle stress / von Mises effective stress) at the crack tip of a CT specimen is shown in Fig. 17. The corresponding distribution based on the Norton creep analysis has been calculated for comparison (not shown in the figure).

The FE LCSP implementation is based on a formulation utilizing the principles J_2 incremental plasticity in describing the viscoplastic flow. In the numerical implementation the strain rate provided by the LCSP model is subjected to a random walk like routine to evaluate the consistency of the FE strain increment particularly in the presence of high strain rates, such as those found near crack tips. The analyses can be carried out either using a local or non-local form of the LCSP model, where in the latter a spatial size dependency is introduced to the constitutive law field variables. Labour related to modification of the implementation or introduction of a completely new material are lessened by the introduction of a Python interface between the FE routine and software used for its derivation, eliminating the need for user interaction or re-coding the constitutive law itself.

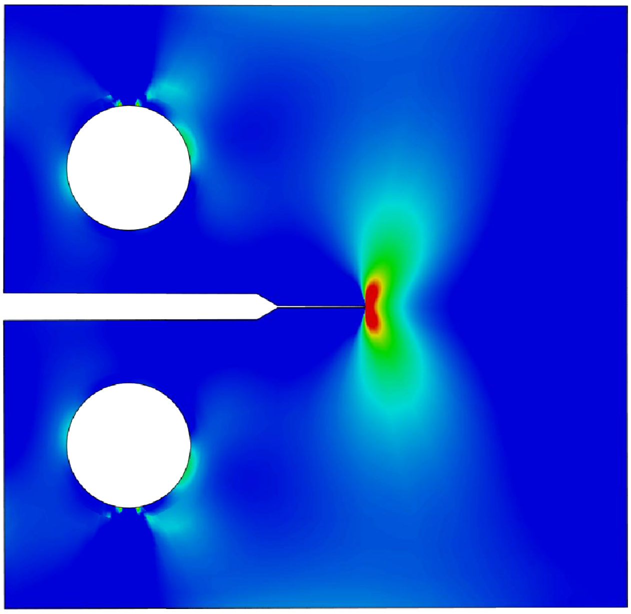


Fig. 16. Strain distribution of a CT specimen calculated using the LCSP creep model.

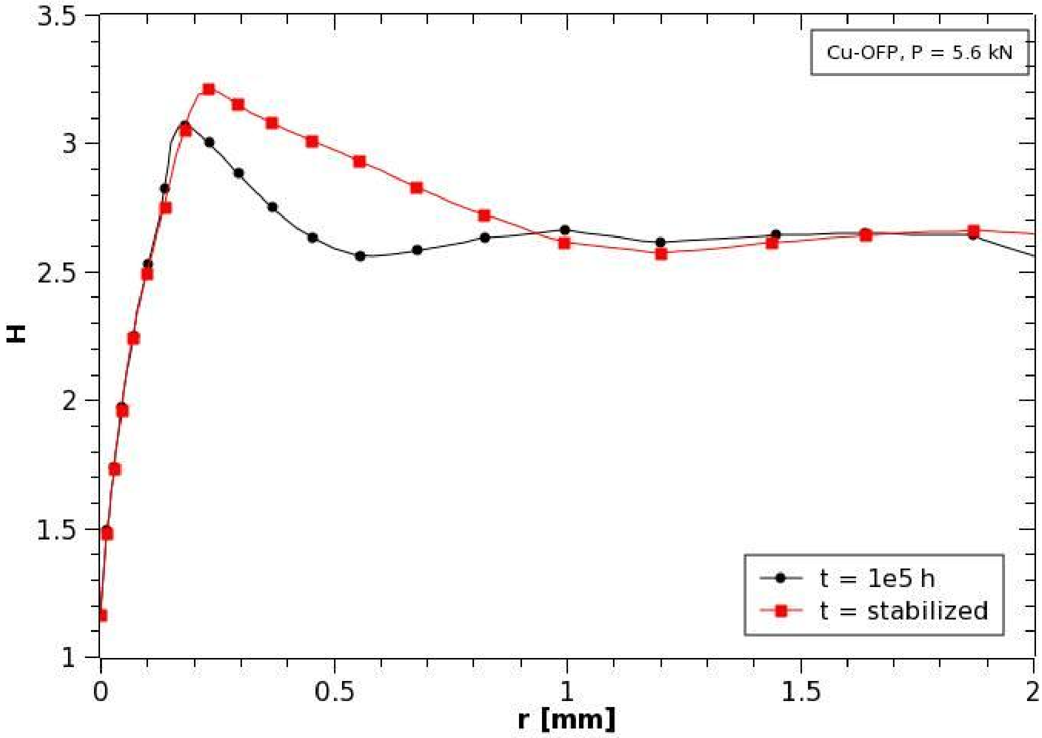


Fig. 17. Distribution of the multiaxiality parameter H at the crack tip of a CT specimen calculated by the LCSP model for the OFP copper.

3.5 Strain and rupture time prediction

The capability of the LCSP 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. 18.

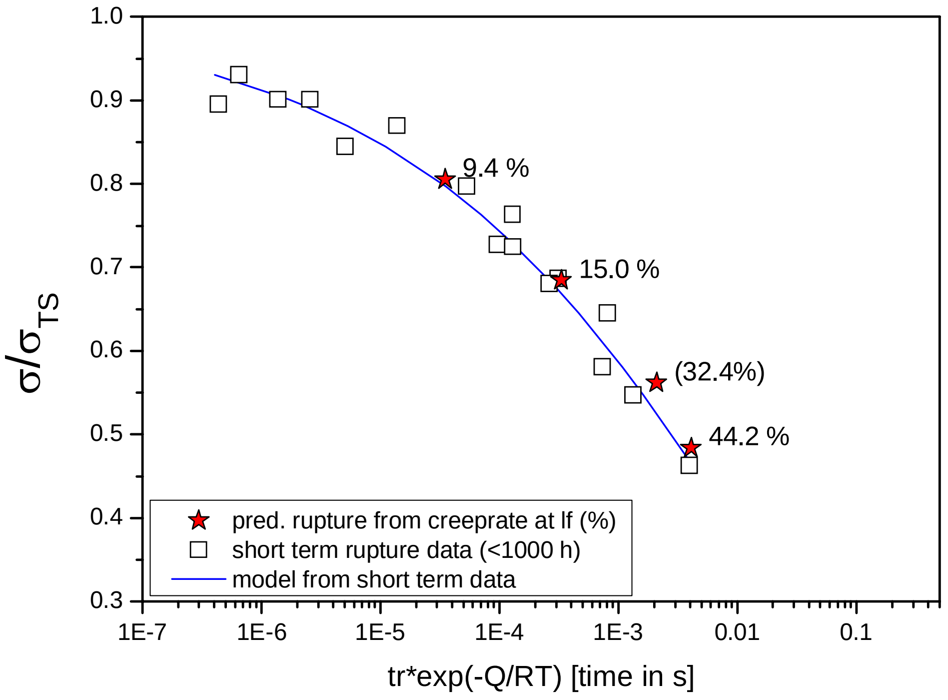


Fig 18. Predicted time to rupture from the observed strain rate (stars, at life fractions shown) in comparison with short-term rupture data (squares) and corresponding model (line); the life fraction value in brackets refers to a test interrupted at over 25600 h.

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 θ-model [12].

4 Life prediction at 50 MPa / 100°C

Extrapolating the above models for the OFP canister copper (corresponding to all available creep data combined), the expected life at 50 MPa and 100°C requires extrapolation of the model (see Fig. 19) to a normalised stress (σ/σ_{TS}) of just below 0.3. The corresponding predicted rupture time at 100°C is 43000 ± 9000 years, which is again less than the specified safe life even if a life of 100000 years is considered as a requirement (note however that the foreseen temperature is much less already after some 1000 years if not earlier). This new prediction is based on the Wilshire rupture model [15], an apparent activation energy of 90 ± 3 kJ/mol, and allows an uncertainty of ±2.5% in the ultimate tensile stress at temperature, and ±2°C in temperature. The predicted life fraction consumed during the first 10 000 years with the temperature profile presented by Raiko [14] is at 50 MPa less than 1% [16]. The predicted life is lower than those obtained earlier, e.g. that reported in the previous yearly report of this project. It should be noted that much of the predicted shortening is due to the optimised apparent activation energy being reduced from about 104 kJ/mol (i.e. that for expected self-diffusion of copper) to 90 kJ/mol.

SSM has shown special interest in the effect of defects in friction stir welds based on a conference presentation made by VTT in 2009 [2]. This issue was discussed in detail during a VTT visit to SSM in Stockholm in November 2009.

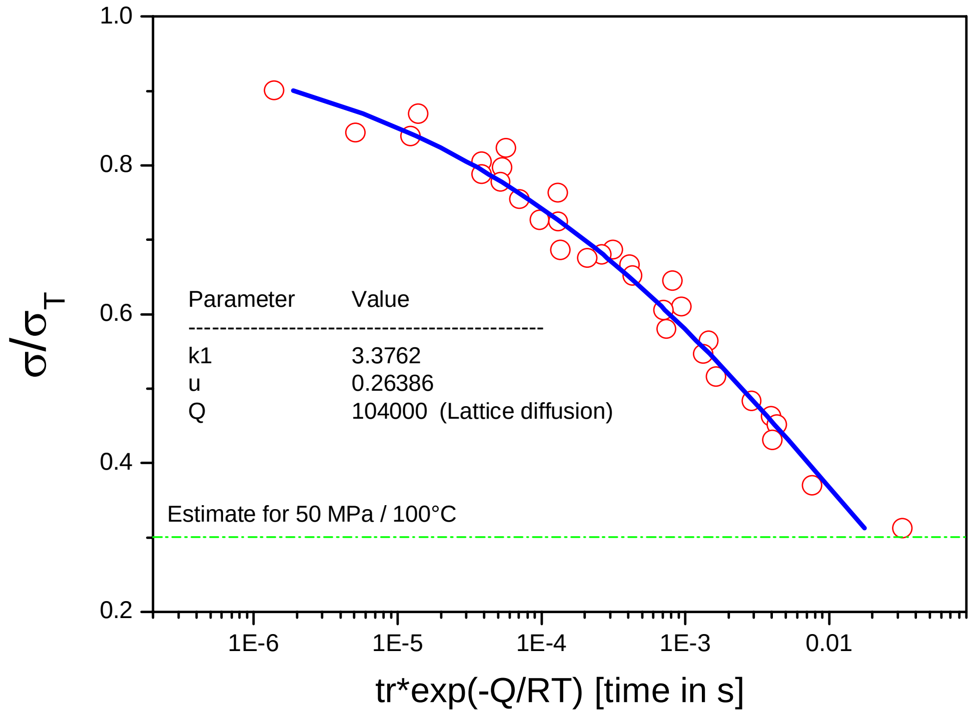


Fig 19. The master curve with the OFP copper rupture data; the horizontal line corresponds the formerly common approximate max loading of the overpack.

5 Conclusions and summary

The uniaxial creep testing program with the long term testing (now exceeded 70000 h) continues to support creep modelling. The results from CT tests and earlier tests with nominally uniaxial but defective specimens [3] suggest that multiaxility is important in limiting life. This has also stimulated the cooperative effort between the Finnish and Swedish programs.

In the friction stir welded (FSW) CT specimen creep testing at 175°C, after 25000 h the joint tip has shown signs of grain boundary cavitation. The next inspection will take place after an additional 5 000 h. However, for damage modelling it is of interest that interrupted testing of the longest uniaxial specimen has also shown distributed microcracking of the type not reported previously. The new test results together with updated material and FE models have been used to update the creep life assessments for the assumed future service history of the copper overpack. In parallel CT testing using Cu-OF base material, much faster intergranular creep damage and crack branching has been demonstrated.

For combined creep and corrosion testing, a most recent 1900 h test under aerated simulated Olkiluoto groundwater at 90°C has been completed and grain boundary corrosion (intergranular attack) was observed at the both side faces of the specimen mainly in the middle of the notch. No evidence of stress corrosion cracking has been observed. The test will be continued.

The microstructural changes during low temperature (175°C), low stress creep experiments on base material and FSW welds of OFP copper appear to be largely restricted to the widening recovery zones at stressed grain boundaries, and to the increasing rate of grain boundary creep cavitation at the tip of multiaxial (CT) creep testing specimens at 175°C after 25000 h. This cavitation appears to be related to local strain, as demonstrated by the concurrently increasing rate of notch/crack tip opening.

Clear extension of creep cracking with low creep ductility has been confirmed in OFHC copper (up to 8000 h), while much higher creep ductility has been retained in OFP copper so far. Longest CT tests for OFP copper have reached the 25000 h mark, and the longest duration of the uniaxial tests has exceeded 70000 h (8 years, continuing but with some surface cracking. The test results together with an updated material model have been successfully applied in FE to characterise the effects of multiaxial stress at the crack tip. The optimised assessment with a somewhat reduced apparent activation energy predicted a safe creep rupture life of 43000 ± 9000 years at 100°C/50 MPa, which is significantly less than in earlier assessments.

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