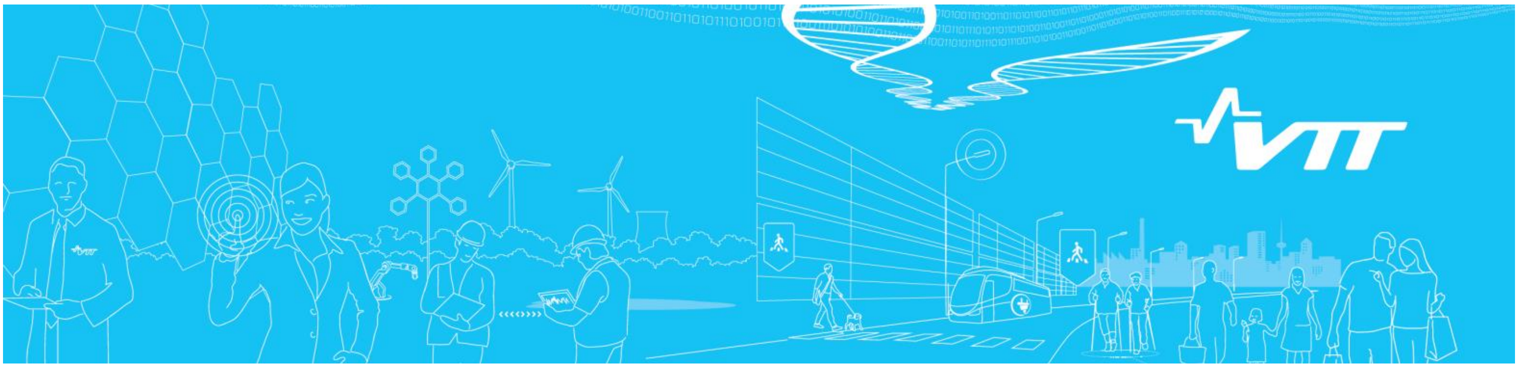

RESEARCH REPORT

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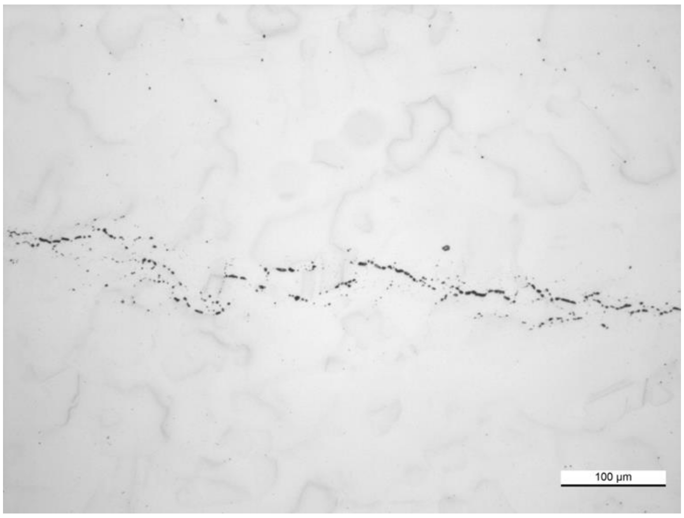

KYT 2014 / MICO

Material integrity of welded copper overpack – Annual report 2014

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Tapio Saukkonen (Aalto)

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Summary

The research programme on the creep issues of the nuclear waste disposal canister copper material has continued with long-running uniaxial and multiaxial experiments as well as relaxation testing to support the material modelling activities, which produce appropriate material models to be used in the FE analysis, which will be used to assess the stresses and strains in the critical locations of the canister.
The relaxation testing is ongoing and it has proven necessary to use a servo-mechanical testing machine in order to generate good quality test data. The initial FE prediction of strain rates calculated by the LCSP creep model and a relaxation model suggests a clear difference, but this needs to be verified after a better relaxation model is defined based on much more data.
The most important result of this year has been the observation of cracking in the oxide particle zone at the friction stir weld root in the longest running CT test, which was terminated after 50 000 hours for sectioning and metallographic investigations. The oxide particles have nucleated creep cavities which have eventually lead to cracking of the oxide particle zone in spite of the low stress state 8 mm ahead of the notch tip. This observation is in line with the earlier results from uniaxial creep tests with a radial sample from the weld root. The weakness only manifests itself in long-term testing because the cavity nucleation and growth process is a slow process. Because of the low strength of the oxide particle zone welding in air should be avoided and vacuum or inert gas should be used instead.

Confidentiality	Public

Espoo 27.2.2015

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Preface

This report is the annual report of the project “Material integrity of welded copper overpack” (MICO2014), including and summarising the experimental, modelling and life assessment activities as well as the results and status of the project up to the end of 2014. This project is part of the Finnish national research program [1] on nuclear waste management, 2011-2014 (KYT2014). The research has been carried out by J. Rantala (project manager and experimental work), P. Auerkari (Principal scientist), A. Laukkanen and T. Andersson (FEA) / VTT and T. Saukkonen / Aalto University (scanning electron microscopy, SEM).

For reasons of clarity the structure of this report follows the new task structure of tasks proposed for the KYT2018 research project PRECO as far as applicable.

The financial support and other guidance by this program, STUK (Finland) and SSM (Sweden) are gratefully acknowledged.

Espoo, February 27, 2015

Authors

Contents

1. Introduction

1.1 Background

In the repository for spent fuel, the temperature of the canister surface is expected to peak at about 75-90°C before the first hundred years [2], with gradual cooling to the level of the bedrock environment (Fig 1). The top temperature will depend on the rate of wetting in individual disposal holes, which might vary a lot, depending on the flow of water in the bedrock. The development of the bentonite swelling pressure will also depend on the rate of wetting.

For the protective copper (Cu-OFP) overpack of the canister, creep and corrosion are included as potential damage mechanisms under the repository conditions [2]. Although relatively mild in usual engineering terms, the repository conditions imply a technical challenge to life estimation for ensuring the integrity of the overpack. This is because of the discrepancy between the longest achievable laboratory test (less than decades) compared to the design life that is of the order of glaciation cycles (about 10^5 years) to reduce the radioactivity of the contents close to the background level. The time difference by a factor of about 10^4 also exceeds the usual range of extrapolation from laboratory experiments to real service conditions in most (or any) comparable engineering applications.

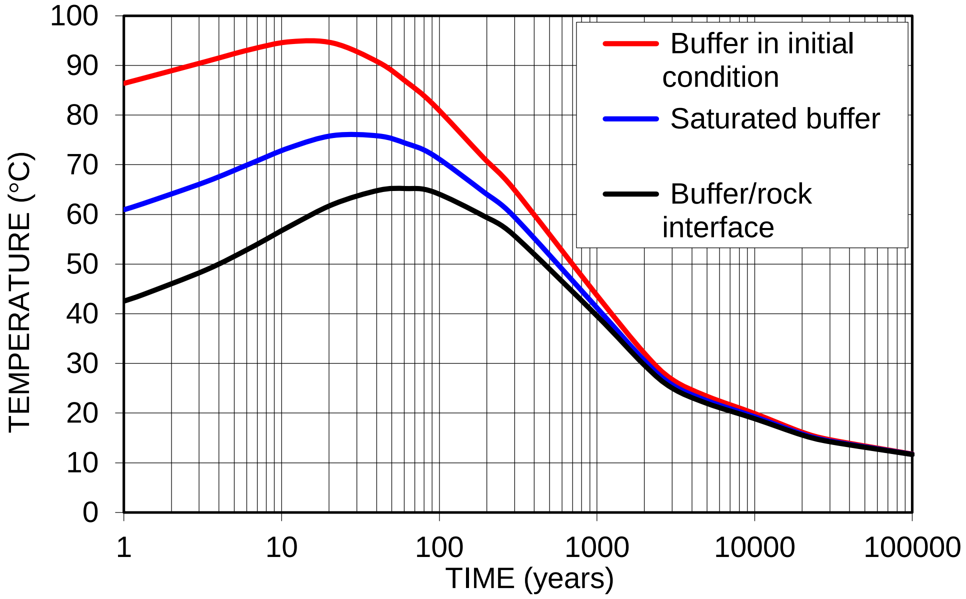

Fig 1. Predicted temperature evolution at the canister surface for EPR fuel [2]; the red curve assumes dry environment with a 10 mm gap around the canister.

This work will deal with both damage mechanisms to provide a realistic model for life prediction and long term behaviour of the copper overpack, including in particular:

- assessment of damage mechanisms and their possible 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 2011-2014 (KYT2014). The project also includes specific issues defined by SSM (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 2014 have been:

- to produce an independent FE verification of the stresses and strains in the canister
- to continue the longest creep tests to beyond 100 000 h to support modelling and the longest CT-specimen to 50 000 h.
- study of creep deformation and electron microscopy (Aalto University)

2. Materials and methods

The OFP copper material for the experiments including friction stir welded (FSW) material from a full scale section provided by SSM/SKI (Sweden) and the Swedish program for canister studies. The CT-specimen CS3 was removed from a cylindrical part (material code T31) and a cover lid (lock TX 82) welded together. Chemical analysis, short term mechanical properties and initial microstructures of the materials have been reported elsewhere [2].

For relaxation experiments a 50 mm thick block of OFP copper plate with a reference code X579 was supplied by Posiva Oy.

Metallography using optical, scanning electron and FESEM/EBSD (Aalto University) microscopy has been applied for as-new materials and test specimens after each given testing period. 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.

For creep modelling, the combined Wilshire and LCSP models have been applied and further developed [3, 4] to support robust FE analyses under non-homogenous stress and strain fields.

3. Relaxation behaviour

In the previous annual report of 2013 [5] it was reported that the FE creep analysis of the copper canister showed rapid relaxation of stresses in the highly stressed areas. In this analysis the LCSP creep model was used. However, it is known that for many other metals a uniaxial creep model does not normally predict relaxation very well at all, and therefore it was decided that dedicated relaxation testing and modelling was necessary. Therefore some initial tests on OFP copper were carried out in the bellows testing rig at 80°C with initial strains of 0.28% and 0.44%, and three classical relaxation models were applied. An example of the data fitting is shown in Fig. 2.The initial results were reported in a separate report [6]. The Kohlrausch relaxation model was applied in FE in order to compare the prediction of relaxation by the LCSP creep model and the initial relaxation model. Fig. 3 shows that the predicted strain rates are very different by the two models. This example only considers the first few hours after load application. When more relaxation data becomes available the models will be refitted and then the behaviour of the whole canister will be predicted by the LCSP creep model and the relaxation model.

The specimen size in the pneumatic bellows testing rig is rather small: diameter 4 mm and 10 mm gauge length. The relaxation testing standards (EN 10319-1 and 2, ASTM E1012 – 12e1) require a specimen which is typically twice as long as a standard uniaxial test specimen. This was not feasible in the bellows testing rig. Therefore it was decided to machine long uniaxial test specimens. However, for relaxation testing it is important to avoid cold deformation during the specimen manufacturing and this is very difficult for copper. Therefore it was decided to machine long specimens with square cross-section using wire erosion. The cross-section was 8.9*8.9 mm which gives an equal area as in a standard 10 mm specimen. By wire erosion it was possible to machine long specimens with a 80 mm long parallel section without cold deformation. However, the residual stresses in the plate material caused some slight bending of the specimen, but did not result in an error greater than 0.01 mm in the final thickness of the relaxation specimens.

The long relaxation specimens were tested at 80°C in a servo-hydraulic MTS materials testing machine. Eight tests were carried out with a planned duration of one week. Axiality of the loading arrangement was checked by strain gauges on the specimen. The specimen was first heated to the test temperature in a resistance furnace and temperature was let to stabilize. The initial strains varied between 0.44% and 20%. The loading rate was fixed such that in all cases the initial strain was reached in 10 minutes. After this the (constant) strain reading and the load drop were recorded. Unfortunately only one test produced a reliable result while in all the other tests either pulses in the hydraulic fluid circuit or some electrical disturbances caused some load peaks, which invalidated the test results. Therefore in 2015 a slower screw-driven servo-mechanical test machine will be used for the relaxation tests. Because of the insufficient number of valid test results the relaxation models [6] have not yet been re-fitted. Therefore the FE analysis with a relaxation creep model has to wait until a good relaxation model has been defined, see chapter 5.

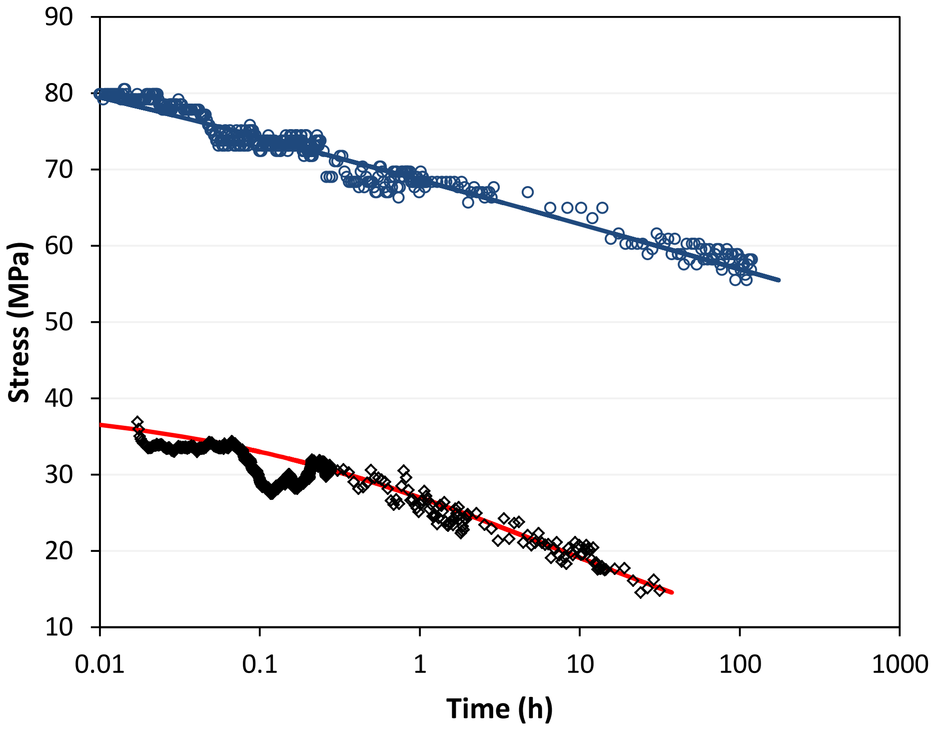

Fig. 2. Relaxation test data at 0.28% and 0.44% initial strain fitted with Söderberg relaxation model.

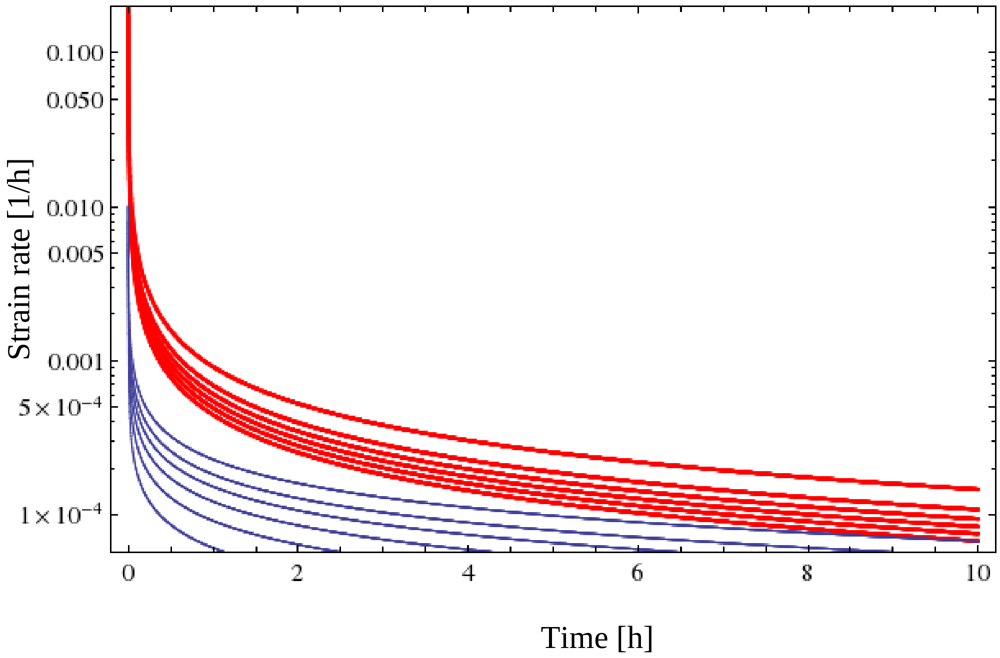

Fig. 3. Comparison of strain rates predicted by FE by using the LCSP creep model (red curves) and the Kohlrausch relaxation model (blue curves).

4. Effect of multiaxiality

This activity consists of two different types of experiments:

- Notched bar tests in uniaxial loading
- CT-specimens for OFP copper and pure OFHC copper

The notched bar test series was initiated already in 2013 in a M.Sc thesis [7]. The test series has been continued and two tests are currently running. Each test specimens has two identical notches. Two specimen geometries had been used: a round specimen with a net diameter of 7.1 mm in a 10 mm cylindrical specimen and a square specimen with an identical cross-sectional area and notch depth. The notch root radius is 0.17 mm, machined with wire erosion. Two tests are currently running. The test results plotted with effective stress are shown in Fig. 4 with the reference data. The data shows that the round and square specimens produce comparable results and that the notched bar data fall on a line with a strength reduction factor (SRF) of 0.78.

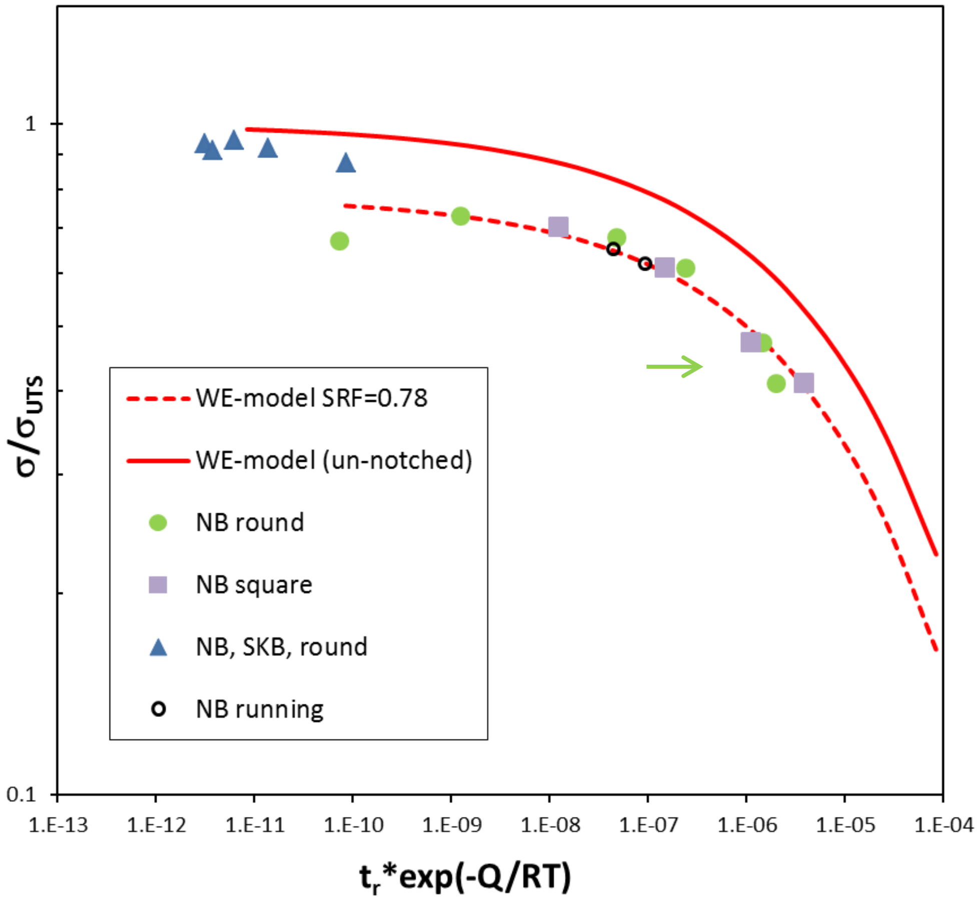

Fig. 4. Notched bar rupture data with round and square specimens compared against reference data.

The long-running CT-specimen specimen (CS3) with a friction stir weld, tested at 175°C at 35 MPa had reached a total testing time of 50 000 hours after which it was decided that the test is stopped and the specimens will be sectioned to see whether the crack has started to grow in the middle of the specimen and how the grain boundary cavity density has developed and how the oxide particle zone has behaved.

It is seen that practically no crack growth had happened at the notch tip. The length of the vertical crack in Fig. 5 is only about 30 μm. The cavity density in the middle of the specimen near the notch tip in Fig. 6 is about 600 $1/mm^2$, which is of the same order of magnitude of what was reported earlier from the side face of the same specimen during the test interruptions. It was expected that the cavity density would be bigger in the middle of the specimen. However, the specimen preparation makes a big difference in how many cavities are seen in the micrographs, so this result has to be regarded as temporary. The optical and SEM investigation of the specimen is ongoing.

The most important result of the CT-specimen CS3 is that the oxide particle region about 8 mm ahead of the notch tip has practically fractured. The location of the oxide particle region is seen in Fig. 7 with a close-up picture in Fig. 8. In Fig. 7 it is seen that the oxide particle zone starts about 8 mm ahead of the notch and bend towards the outer surface of the canister following the process zone of the friction stir weld. The vertical part of the oxide particle zone has fractured as is shown in Fig. 8. No cracking was observed in the upper parts of the particle line in Fig 7, but the specimens will still be studied in more detail with SEM. In the FE analysis of a copper CT-specimen it was calculated that the vertical stress 8 mm ahead of the crack tip is only about 13% of the maximum vertical stress ahead of the notch tip. This means that the strength of the oxide particle zone is strongly reduced, because the oxide particle zone has cracked (Fig. 7 and 8), while the material in front of the notch tip (Fig. 5) has not even cavitated remarkably although the vertical stress in front of the crack tip is about seven times higher than in the oxide particle zone.

The friction stir weld block, from which the CT-specimen was machined, was welded in air while the current practise is to do the welding in protective (inert gas) atmosphere. Therefore it is likely that in the new welds there will be much less oxide particles. Some oxide can appear because there is an oxide scale on the initial surfaces to be joined, but the amount will be much smaller than in the pictures shown below. At the root of the weld the proportion of original (oxidised) surface and weld material is much bigger than near the outer surface of the canister, so it is likely that the problem will mainly concentrate on the root area only. However, the behaviour of the same location in the new welds has to be verified experimentally.

As long as it is not proven that welding in inert gas or vacuum will help avoid the oxide particles it must be assumed in the FE analysis that the joint line hooking and the oxide particle region will have a strongly reduced strength. In the FE analysis of a copper CT-specimen it was calculated that the vertical stress 8 mm ahead of the crack tip is only about 13% of the maximum vertical stress ahead of the notch tip. The fact that in spite of the low stress state the oxide particle region had practically cracked is an indication that the oxide particles will act as nucleation locations for grain boundary cavities. This is in line with the earlier results [8] of the radial cross-weld creep tests, see Fig. 10. The radial specimen orientation is shown in the lower left-hand corner in Fig. 9. The rupture times of the radial specimens in Fig. 10 are close to the reference data at high stresses, which indicates that the strength loss of the oxide particle zone does not show at high stresses, but becomes more pronounced at low stresses. The weakness only manifests itself in long-term testing because the cavity nucleation and growth process is a slow process. It is therefore recommended that a suitable damage mechanics model should be applied in the future FE analysis where the oxide particle zone is modelled.

FSW cross-weld uniaxial creep tests have been performed by SKB in the axial direction of the cylinder [9], see Fig. 11. These tests have not shown strength reduction as a result of possible oxide particles at or near the process zone, but unfortunately no FSW cross-weld testing has been carried out at stresses below 130 MPa, see Fig. 10. This is an obvious shortcoming which SKB and Posiva have to rectify. This would be recommendable because any weakness produced by small particles will not show in rupture data at high stresses but rather at low stresses and long testing times, as shown in Fig. 10. At the conditions where FSW cross-welds have been tested the rupture strains are better than 30% while most of the EB cross-weld data fall below this value as shown in Fig. 12.

The long-running uniaxial creep test at 120 MPa and 152°C has been running for 104 000 hours (11.9 years). It has been decided that this test will not be interrupted anymore for inspections before rupture, because other creep tests have shown that every interruption adds a new primary creep component to the creep curve which consumes the deformation capacity of the material and will shorten the creep life.

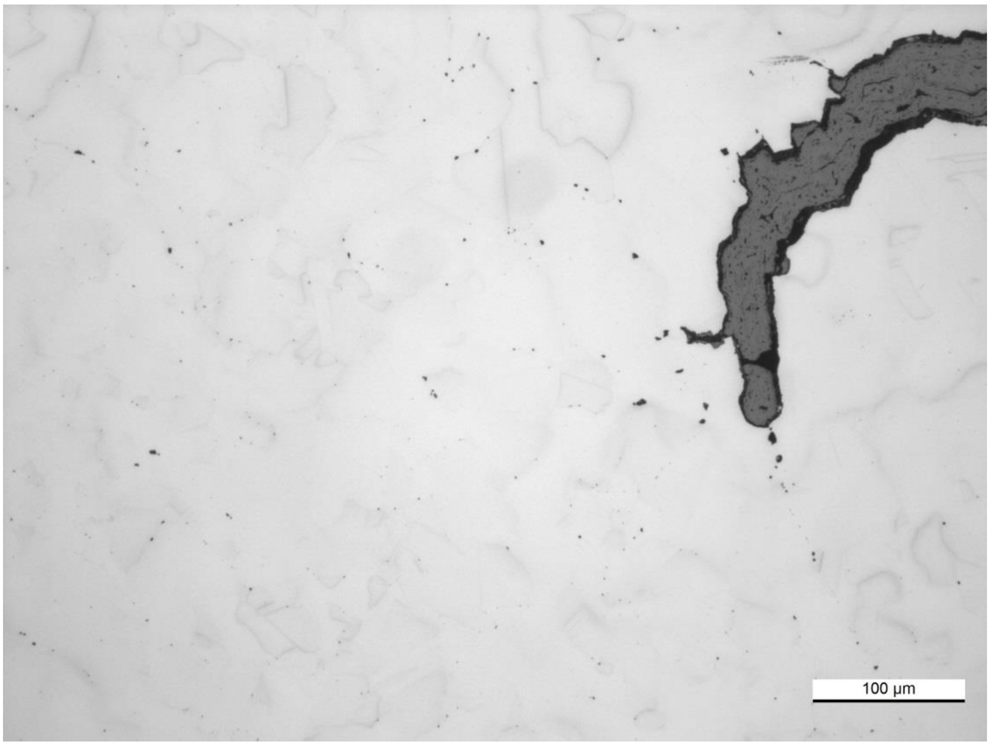

Fig. 5. Tip of the natural notch and the joint line hooking in the middle of the CT-specimen CS3 after 50 000 h of testing. There is hardly any crack growth and not much cavitation.

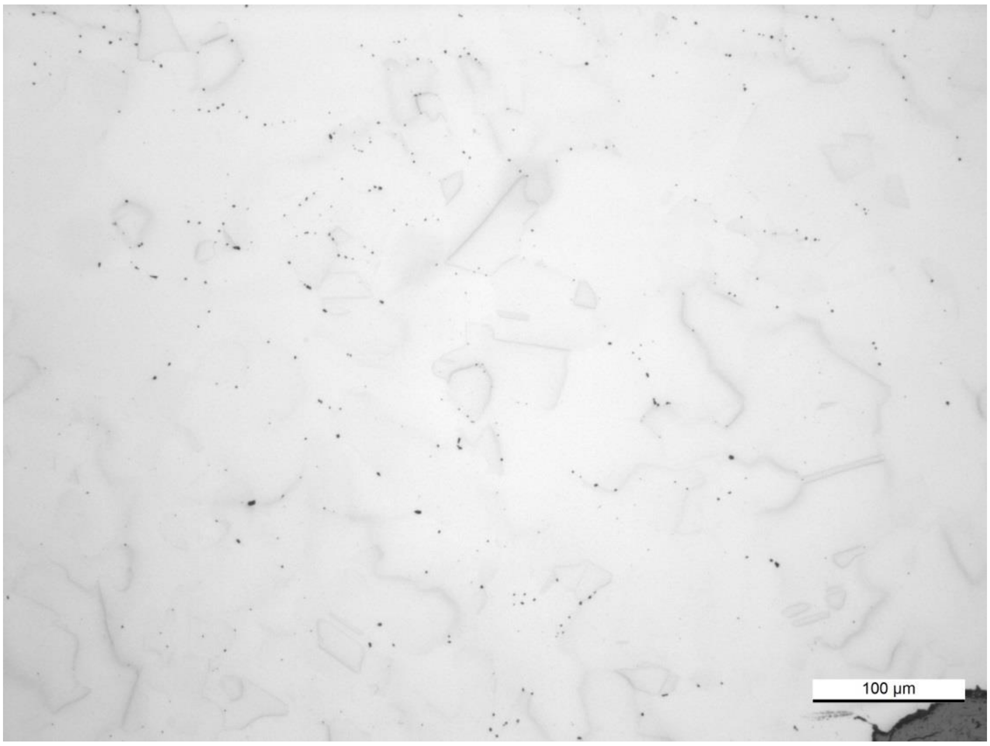

Fig. 6. Grain boundary cavitation near the notch tip of the CT-specimen CS3 after 50 000 h of testing.

Fig. 7. The metallographic section from the middle of the CT-specimen CS3 after 50 000 h of testing with the notch and the joint line hooking on the right and the oxide particle zone 8 mm to the right from the notch tip.

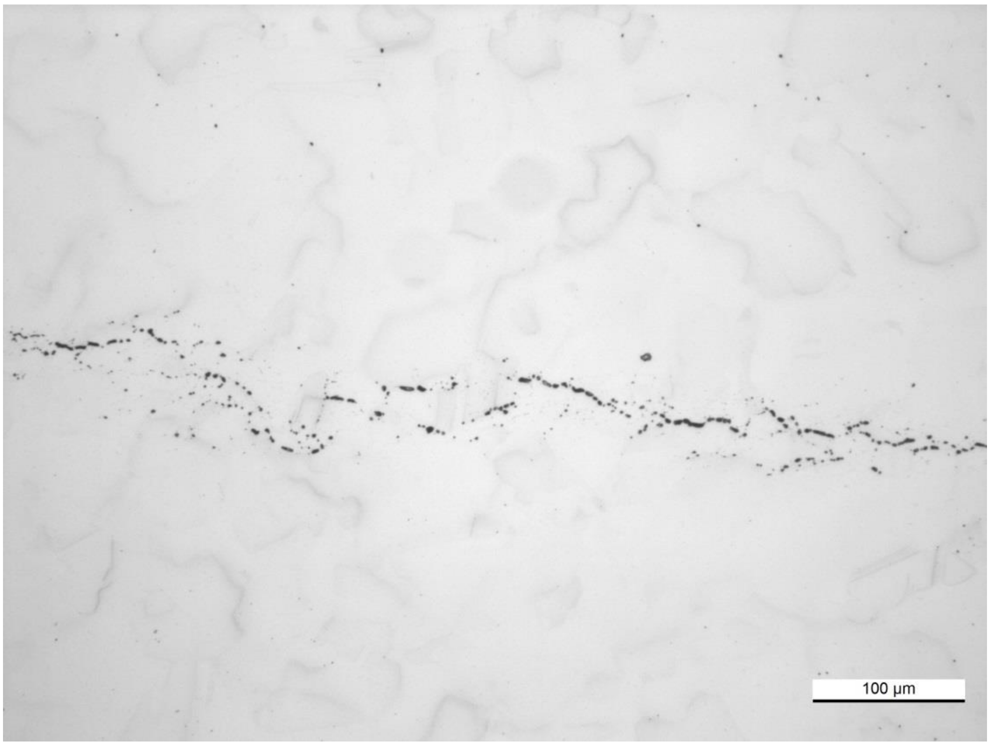

Fig. 8. The cracking in the oxide particle zone in the CT-specimen CS3 after 50 000 h of testing.

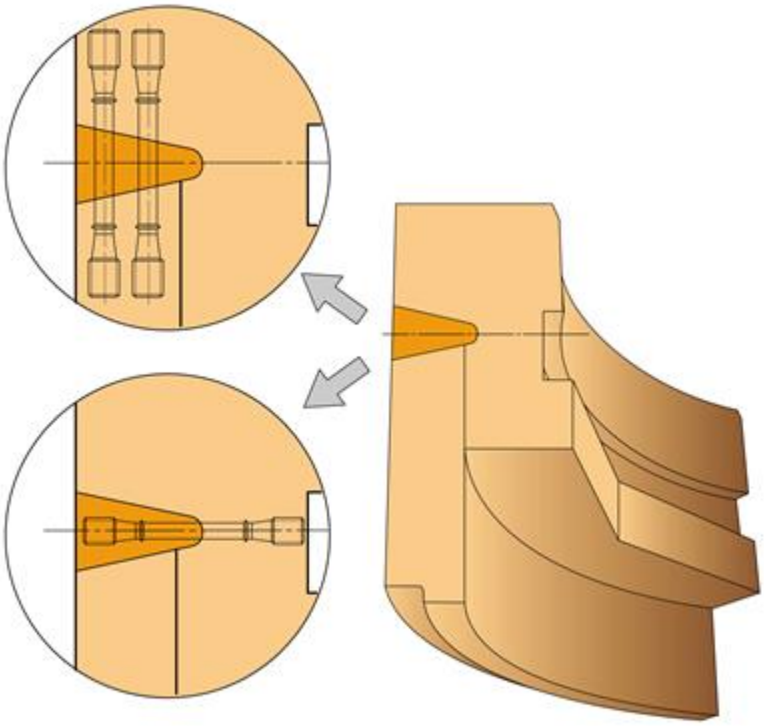

Fig. 9. Uniaxial cross weld specimens removed from the FSW in axial and radial directions.

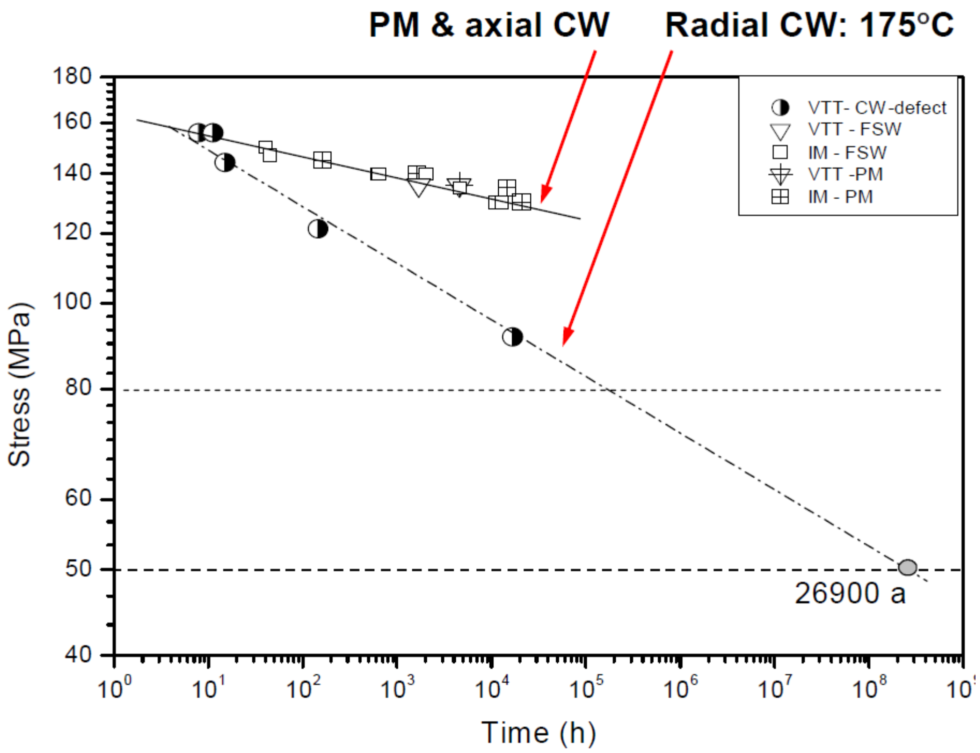

Fig. 10. Radial cross-weld creep rupture data compared with reference data [8].

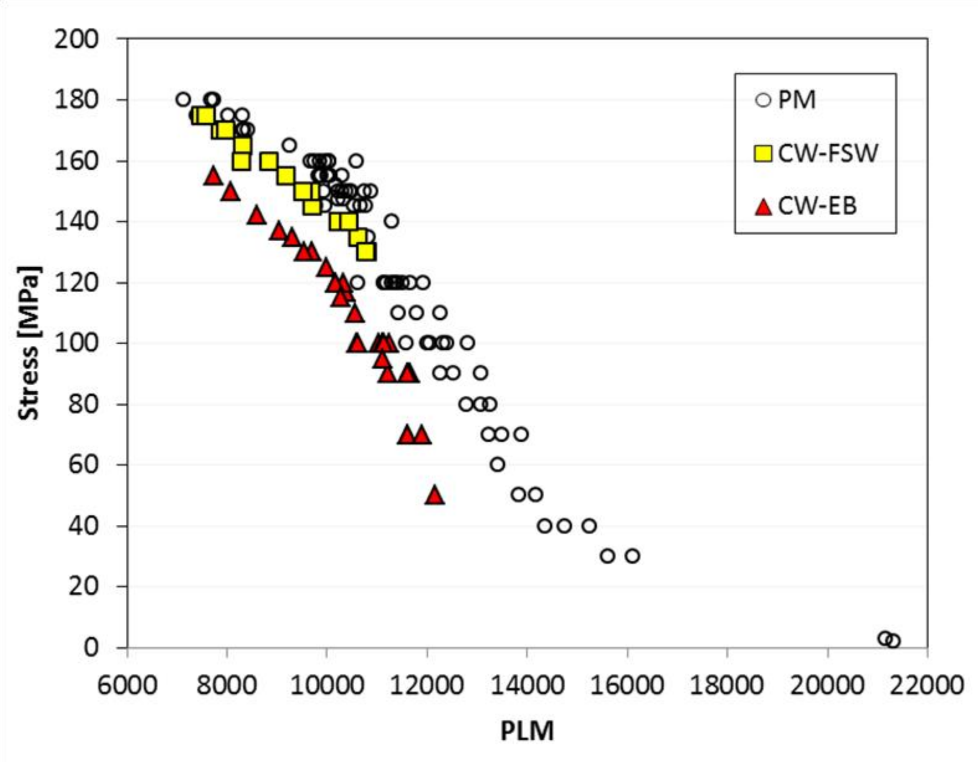

Fig. 11. FSW and EB cross-weld rupture data compared against parent metal (PM) rupture data [9]. PLM = Larson-Miller parameter.

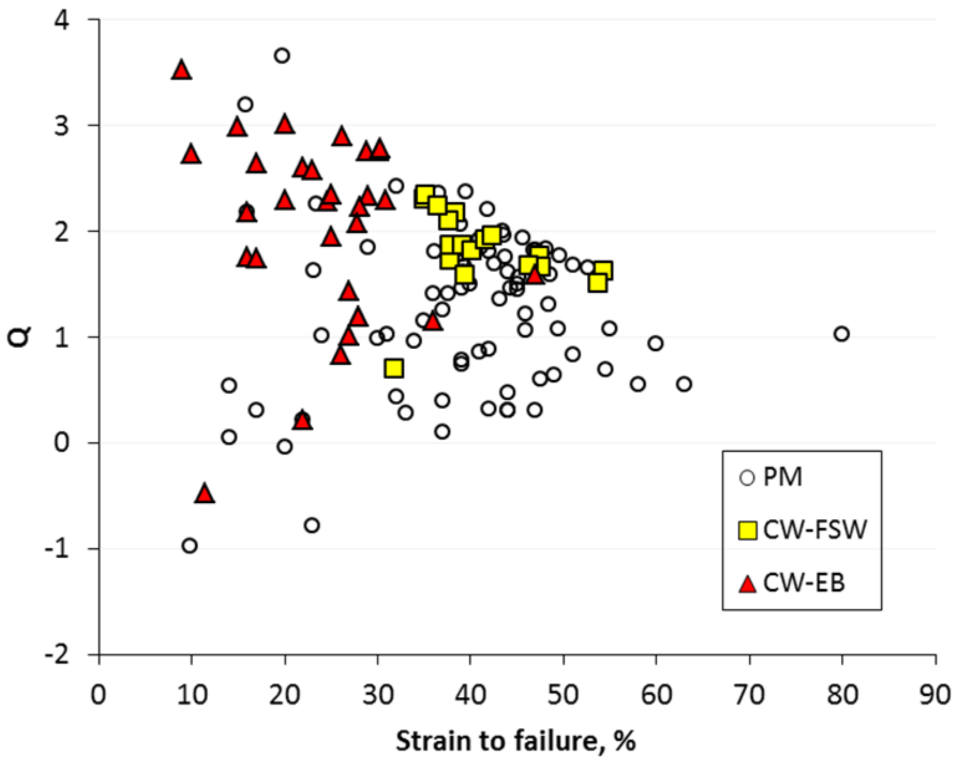

Fig. 12. Parameter Q for localisation of damage as a function of failure strain for FSW and EB cross-welds compared against parent metal (PM) data [9].

5. Canister FE analysis

The effect of the mesh element size was tested, but this had a minor influence in the distribution of the elastic-plastic von Mises stress. A more precise definition of the plastic behaviour has been done on the basis of new tensile test data for OFP copper. As shown in Fig. 3 the LCSP creep model predicted higher strain rates than the initial version of the Kohlrausch relaxation model. When the stress distribution of the canister was calculated, it was seen that the stresses calculated with the LCSP creep model were considerably lower than the stresses calculated with the relaxation model, see Fig. 13.

As mentioned in chapter 3 much of the relaxation test data has been unsatisfactory and need to be repeated. When good quality data has been generated the relaxation models will be re-fitted and the FE analysis will be repeated in 2015.

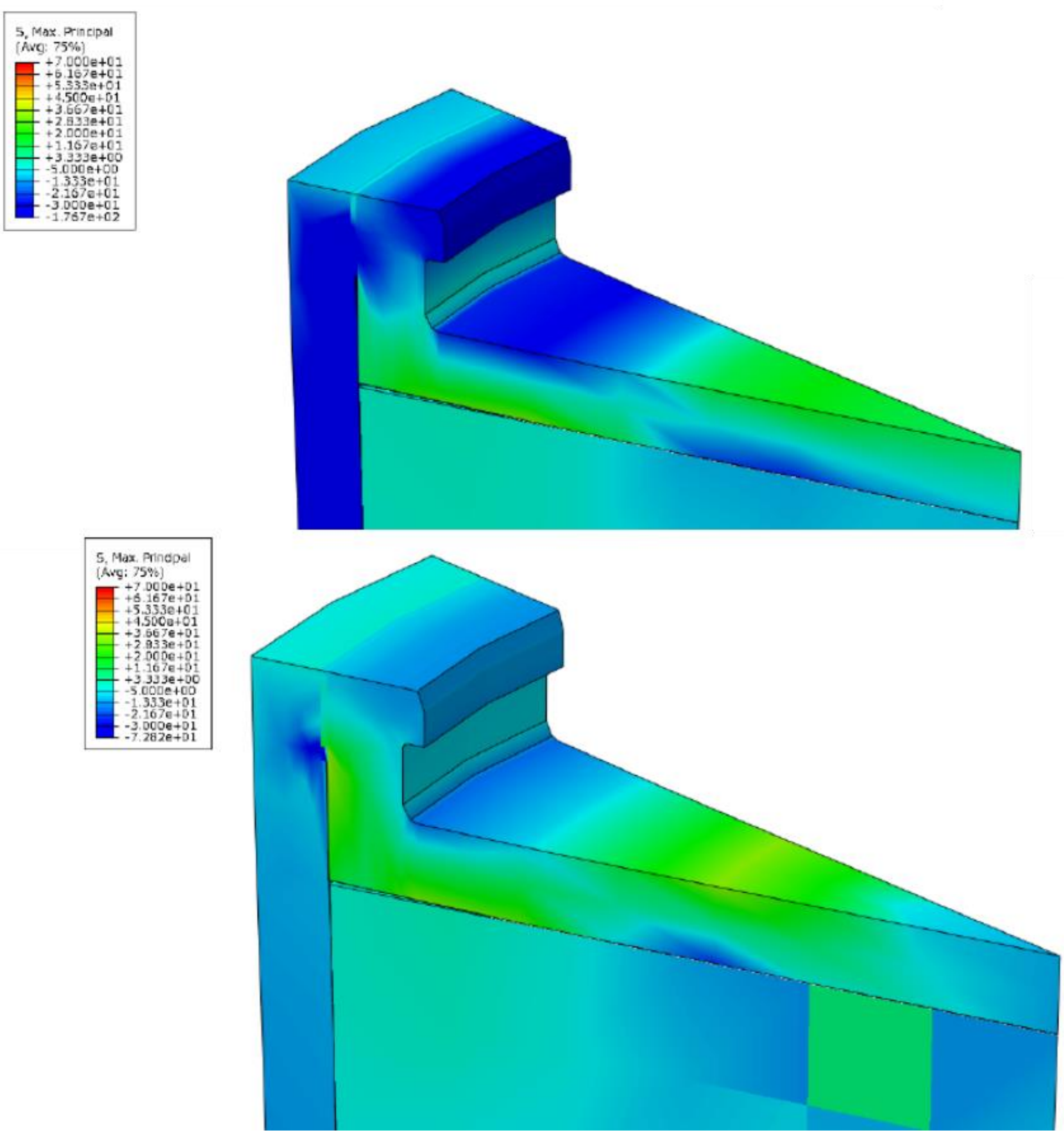

Fig. 13. The maximum principal stress distribution in the canister after 20 hours calculated with the LCSP creep model (top) and the Kohlrausch relaxation model (bottom).

6. Training and development of resources

As a new test method relaxation testing was started in 2014 in order to facilitate the definition of an appropriate relaxation model for the FE analysis. The preparation of the sufficiently long specimens without cold deformation has been a challenge as described already in chapter 3, but wire erosion seems to guarantee good quality specimens. The relaxation testing with constant strain loading in a servo-mechanical machine has been another challenge, and it has proven necessary to use a slower screw-driven servo-mechanical testing machine in the future.

Another new test method which is being prepared is Small Punch testing. A rig for testing at room temperature is being built. Small Punch method is a miniature method which allows small local zones of material to be tested using a coin-shaped specimen with dimensions of diameter 8 mm and thickness of 0.5 mm. It would be interesting to extract a test sample for example from the oxide particle zone.

The PhD thesis of J. Rantala is in progress, but some challenging questions have not yet been answered.

7. Reporting and dissemination

In addition to the normal reporting according to the KYT guidelines a presentation about the creep research of the nuclear waste disposal canister was held at the ECCC2014 Conference in Rome 5-7.5.2014 [10].

8. Conclusions

It is essential to perform the FE analysis of the copper canister by using a material model which consists of models for plasticity, creep and relaxation. Two first elements of these are well covered, but the development of the relaxation model requires better quality data to facilitate the development of a relaxation model. This means in practice that a screw-driven servo-mechanical machine has to be used for the tests instead of a servo-hydraulic machine.

The notched bar test programme is ongoing and has shown comparable results between specimens with square and round cross-section. When plotted by an effective stress the results suggest a strength reduction factor of 0.78.

The long-running CT specimen test has shown cracking at the weld root in the oxide particle zone in spite of the low stress levels far away from the notch tip. This supports the decision to seal the canister in an inert gas atmosphere or vacuum instead of air in order to avoid the formation of oxide particles. Before the quality of the new welds has been demonstrated, as a conservative assumption, a zero strength zone at the joint line hooking and oxide particle zone will be assumed in the FE analysis.

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