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Strain localization in copper canister FSW welds for spent nuclear fuel disposal

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HIGHLIGHTS

• Utilization of shielding gas during FSW of copper reduces the number of oxide particles in the welds considerably.
• Due to differences in the initial state of the lid materials, the new canister weld performed worse than the old weld.
• The strain localization behavior of the welds should be taken into consideration in the manufacturing of the canisters.
• Thermal hydrogen charging of the weld material results in considerable hydrogen uptake at the oxide particles.
• The strain localization behavior was quantified by using Gini coefficients.

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ABSTRACT

Spent nuclear fuel disposal in copper canisters in a deep geologic repository is planned in Finland and Sweden. The purpose of the copper shell is to perform as a ductile corrosion barrier to prevent radioactive substances from leaking into the environment. Therefore, the most important property of the copper shell, besides the good corrosion resistance, is its ductility. The copper canisters are sealed by friction stir welding (FSW), which results in strong welds when compared to the base materials, but the microstructural heterogeneity introduced by the welding may also lead to strain localization. Thus, the strain localization behavior of two different copper canister welds was studied by tensile tests in combination with digital image correlation (DIC). The main difference between the welds is the utilization of shielding gas to reduce oxidation during welding. The shielding gas improves the stability of the welding process, as well as reduces the number of oxide particles in the welds. It is known, that oxide particles are detrimental in copper in the presence of hydrogen. Therefore, the two welds were also thermally hydrogen charged to study hydrogen trapping in the weld material. Thermal desorption measurements (TDS) show that considerable hydrogen uptake occurs in the weld oxide zone, but it did not compromise the ductility of the copper welds in these tests. However, the DIC tests indicate considerably earlier strain localization on the retreating side of the new weld, welded with the shielding gas. This is attributed to differences in the initial state of the lid materials.

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1. Introduction

Spent nuclear fuel encapsulation into 50 mm thick copper canisters and deposition in a deep geologic repository 450 m underground will be the main disposal method of spent nuclear fuel in Finland and Sweden [1–3]. The copper canister provides a ductile corrosion barrier, whereas a cast iron insert inside the canisters, with channels into which the spent nuclear fuel is placed, provides the required mechanical strength against external loads. The canisters are placed upright into holes drilled in the bedrock and surrounded by bentonite clay which, among other functions, performs as a buffer against rock displacement.

The canisters are sealed by friction stir welding (FSW) a lid onto the tubular body of the canisters. While it is known that FSW of copper leads to mechanical properties near to those of the base materials [4–6], the variation in cross-weld microstructure and residual stresses may result in strain localization during the long disposal period of the canisters. The copper canister should
withstand the strains without rupture to prevent radioactive substances leaking into the environment. Thus, the most important property of the copper shell, besides the good corrosion resistance, is its ductility. Strain localization, on the other hand, may compromise the deformation capability of the copper shell locally.

The loads in the repository conditions are mostly compressive. However, tensile stresses may be introduced by uneven bentonite swelling and shift of the bedrock [3]. Creep of the copper shell, until it is in contact with the cast iron insert, will also introduce some local tensile stresses due to geometrical effects. The capability of the copper shell to withstand these strains without rupture is affected by the extremely low strain rates, exposure to elevated temperatures, radiation, and possibly by corrosion reactions. One of the uncertainties mentioned by the Swedish Nuclear Safety Authority (SSM) [7] is hydrogen absorption due to, for example, corrosion reactions [8,9] and radiation-induced hydrolysis of water [10]. It has been shown that hydrogen causes strain localization by activating more slip locally in deformation of single crystal copper [11] and polycrystalline copper may suffer from reduced creep ductility due to opening of grain boundaries under electrolytic hydrogen charging [12]. Hydrogen may also affect void generation in copper by stabilizing vacancy clusters [13]. The solubility of hydrogen in copper is rather low [14,15], but oxide particles in the FSW welds [4] may increase hydrogen uptake in the weld material. In addition, copper is known to suffer from hydrogen embrittlement when oxide particles are present in the material [16]. This may locally compromise the ductility.

In this paper, strain localization in two different copper canister FSW welds, one welded without shielding gas and one with shielding gas to prevent oxidation during welding, was studied by digital image correlation (DIC) [17] in combination with tensile testing. DIC brings several advantages when studying localized deformations in comparison to traditional methods, such as strain gauges, extensometers or a manual grid method, as it enables full-field measurement of local displacements from the whole surface of interest over the whole duration of the test. DIC measurements may be performed in 2D by using one camera or in 3D by using several cameras. For a tensile test 2D measurement is often considered adequate. Once the images are captured, they can be reanalyzed as many times as needed, and the local displacement data may be used to reveal different aspects of the underlying behavior of the material. In addition to studying strain localization in the two welds with DIC, the same two welds were thermally hydrogen charged to study hydrogen trapping in the oxide zone of the welds, and the effect of the trapped hydrogen on the deformation of the welds.

2. Experimental

2.1. Material, welding of the canisters, and tensile specimen preparation

The studied material was phosphorous-alloyed oxygen-free copper (Cu-OFP) intended for use in spent nuclear fuel disposal canisters [3]. The material consists of 99.99 wt.-% copper with addition of 50 ppm phosphorus to improve the creep resistance [18,19]. The chemical composition was measured by the manufacturer to be within specification. A measurement of the composition is presented in Table 1. The lids of the canisters are produced by forging and the tubes by extrusion or pierce-and-draw method. Two canister FSW welds were studied in terms of strain localization. The main known difference between the welds is the utilization of the shielding gas. The old weld, designated as FSWL69, was welded without shielding gas, whereas the new weld, designated as FSWL109, was welded in a protective shielding gas to prevent oxidation during welding. The welding process was shielded externally by argon and internally (inside the canister) by nitrogen. The original weld designs of the manufacturer are used to enable full traceability of the results.

The welds were prepared by the Swedish Nuclear Fuel and Waste Management Co. (SKB) over several years, according to the welding routine at that time. There may be small differences in the welding process, but the welding tool design and the welding parameters have remained for the most part the same. The welding speed was 86 mm/min and the default tool rotation was 400 rpm. The welding temperature for both welds was maintained at 850 °C by controlling the tool rotation [20]. The welding is performed clock-wise when looking at the canister from the top and the tool rotation is also clock-wise, as indicated in Fig. 1. This means that the retreating side (RS) of the weld is located on the lid side and the advancing side (AS) of the weld is located on the tube side of the canister.

Table 1

<table>
<thead>
<tr>
<th>Cu</th>
<th>Ag</th>
<th>As</th>
<th>Bi</th>
<th>Cd</th>
<th>Fe</th>
<th>Hg</th>
<th>Mn</th>
<th>Ni</th>
<th>O</th>
<th>Pb</th>
<th>Sb</th>
<th>Se</th>
<th>Sn</th>
<th>Te</th>
<th>Zn</th>
</tr>
</thead>
<tbody>
<tr>
<td>bulk (min. 99.992 wt.-%)</td>
<td>13.9</td>
<td>0.92</td>
<td>0.13</td>
<td>&lt;0.003</td>
<td>0.6</td>
<td><strong>0.5</strong></td>
<td>&lt;0.1</td>
<td>0.6</td>
<td>1.8</td>
<td>51</td>
<td>0.26</td>
<td>5.0</td>
<td>0.06</td>
<td>0.1</td>
<td>0.08</td>
</tr>
</tbody>
</table>

Fig. 1 shows the canister geometry, welding direction, location of the FSW weld in the canister structure, as well as location of the tensile specimens and thermal desorption spectroscopy (TDS) samples cut across the welds. All the specimens were cut with electro-discharge machining (EDM) to minimize unwanted deformation on the sample surfaces. Dimensions of the gauge section of the tensile specimens were 3.0 × 30.0 × 53.6 mm. The cross-section size was chosen to result in maximum load, close to the maximum capability of the tensile testing machine. Prior to testing the tensile specimens were patterned with an optimized DIC pattern [21] similarly to as described in ref. [22] to improve the precision of the DIC measurements. The tensile specimens in this study were cut from segments of the canister welds provided by the manufacturer. In FSWL69, the tensile specimens were cut at about 76° and 100° of weld travel, and in FSWL109 the tensile specimens were cut at about 127° of weld travel. Four specimens were cut from both welds.

2.2. Microstructural observations

The welds were studied by optical macroscopy for the general view and by scanning electron microscopy (SEM) for details in the microstructure. Prior to optical observations, the cross-sections of the welds were wet ground and polished down to 1 μm diamond paste, followed by etching in FeCl₃+HCl solution to reveal the microstructure. Several optical macrographs were taken to cover the whole area of the weld. The images were then combined to create a single image. This introduces some distortions in the dimensions of the weld macrograph, but it is assumed that these errors are negligible.

In addition to the macrographs, the welds were studied locally with electron backscatter diffraction (EBSD) to understand the strain localization behavior. The SEM used for these measurements is a Zeiss Merlin VP Compact field emission gun FEG-SEM equipped
5.1.1 by the method of ref. [24]. The evaluation gives a lower bound on the energy weighted dislocation density. The actual dislocation density is most likely higher since z-direction of the geometry is not included in the EBSD data (it is only approximated by the method), and some dislocations may form dipoles, which do not affect the geometry of the crystals significantly.

2.3. Hardness measurements

Microhardness of the two welds was characterized with an automated CSM Instruments microhardness tester by using 4.903 N load (HV 0.5/500 gf), 9.807 N/min loading rate, and 10 s dwell time. The samples were cut with EDM, and prepared with conventional wet grinding down to 4000 grit SiC paper and diamond paste polishing down to 1 μm to remove deformation from the surface. Arrays of 33 x 24 measurement points were taken over the welds with spacing of 2 mm, which equals to an area of 64 x 46 mm. The instrumented Vickers hardness (HVIR) was determined automatically from the load-displacement curves by using the Oliver-Pharr method [25]. The HVIR values and location of the measurement points were then imported to Matlab for data visualization. The hardness maps were smoothed by median filtering over 3 x 3 measurement points and by interpolating between the median-filtered data points.

2.4. Hydrogen charging and measurement

There are two common methods for hydrogen charging: electrochemical and thermal hydrogen charging. Electrochemical hydrogen charging was not possible in this instance due to the large size of the tensile specimens. Thus, thermal charging was selected to study hydrogen trapping at the oxide particles of the welds. Two tensile specimens, one from each weld, and additional left-over pieces, located on the right-hand side of the tensile specimen in Fig. 1, from each weld were thermally hydrogen charged at 250 °C, 100 MPa, for 10 days. It is known, that the solubility of hydrogen in the copper lattice is rather low, and thermal charging at 250 °C would not result in considerable hydrogen uptake at the copper lattice, but it would still most likely result in hydrogen uptake at the oxide particles.

The left-over pieces were used for thermal desorption spectroscopy (TDS) analysis and the tensile specimens were tested similarly to other specimens to study the effect of trapped hydrogen on the ductility of the FSW welds. Several TDS samples, named S1–S5 for both welds, were cut from the left-over pieces from the location shown with dashed lines in Fig. 1 to study hydrogen trapping near the outside surface of the canister. More detailed location of these samples is presented along the results for hydrogen uptake. A typical size of the TDS samples was 0.9 x 3.8 x 10−15 mm. Before measurements, the TDS specimens were mechanically polished down to 1200 grit SiC paper, washed in acetone in an ultrasonic bath, and dried under flow of pure helium gas. The TDS apparatus is based on the mass-spectrometry measurement of hydrogen desorption from the specimen in ultra-high vacuum (UHV) chamber under constant heating rate. With vacuum better than 5 x 10−9 mbar, the apparatus allows to measure small quantities of hydrogen in metals down to 0.1 at.ppm. The temperature range of the TDS measurements covers room temperature to 1200 °C.

2.5. Tensile testing with DIC

Tensile testing with 2D optical displacement measurement through digital image correlation (DIC) was performed with a screw-driven Zwick/Roell 2020 tensile testing machine and a Strain-Master DIC system by LaVision. The tests were performed at room temperature with two constant extension rates: 0.001 mm/min and 1 mm/min, which for the gauge length of 53.6 mm, corresponds to strain rates of 3.1 x 10−4 1/s and 3.1 x 10−3 1/s. The slow strain rate, which corresponds to minimum of the tensile testing machine, led to test duration of about two to three weeks depending on the specimen and the fast strain rate led to test duration of about 30–40 min. The strain rates in the real repository conditions are expected to be much lower than the strain rates in this study, still.

Prior to testing, the tensile specimens were patterned with an optimized copper oxide pattern by utilizing a photolithographic method, to increase the image contrast, which results in improved DIC data quality and precision [22]. A similar patterning method as presented in ref. [22] was used, with the exception that the...
specimens were not heated during oxidation of the pattern to avoid hydrogen escape from the hydrogen charged tensile specimens. This was possible by using a liver-of-sulphur (K₂SO₄) solution for the oxidation of the pattern, instead of a mixture of sodium hydroxide (NaOH) and sodium chloride (NaClO₂) as used in ref. [22]. The patterning sequence was also different from ref. [22] as the oxidation of the surface in this study was done after only application and development of the photoresist mask. Pattern preparation included wet grinding the specimen surface down to 2000 grit SiC paper, application of a photoresist layer on the wet grinded and cleaned surface by spray can (MicroSpray Positive was used), spinning the specimen to provide an evenly thick photoresist layer, curing the photoresist with hair dryer for 1–2 min, exposing the photoresist to UV light for 7–8 min (this requires the use of a glass mask on top of the specimen with the pattern inscribed on it), developing the photoresist in an NaOH based developing solution for about 1 min, and finally oxidation of the pattern in a liver-of-sulphur (K₂SO₄) solution for about 1–2 min. The remaining photoresist was then removed with acetone.

The images were taken with a LaVision Imager pro X camera equipped with a Nikon Micro-Nikkor 105 mm lens. Dynamic range of the camera is 14 bits and the size of the camera sensor is 1600 × 1200 pixels. The images were taken with a field of view of roughly 600 pixels horizontally and 1600 pixels vertically. The resulting physical size of the pixels with this set-up was about 60 μm/pixel. Images were taken with 0.004 Hz for the slow strain rate tests and 4 Hz for the fast tests. This corresponds to 0.08 mm cross-head displacement between each image for both strain rates. The specimens were illuminated by indirect diffused light to minimize detrimental reflections from the copper surface [22]. A spotlight was placed behind the specimen and the light was reflected from two curved curtains, made of projector screen canvas, located in front of the specimen. Monitoring of the specimen was performed from an opening between the curtains. After the tests, digital image correlation was performed with LaVision DaVis software version 8.4.0. The calculation settings are shown in Table 2. Guidelines for performing mechanical testing with DIC may be found in ref. [23].

2.6. Data analysis and visualization

Displacement data and original images obtained with DaVis software were transferred to Matlab for further analysis and visualization. The vertical engineering strain components were calculated by using central finite differences on raw displacement field data. The edges of the displacement data were omitted to avoid erroneous strains from edge effects. No out-of-plane compensation was performed, which results in small errors in strains when necking of the specimen occurs. Since the plastic strains in these experiments are in the order of 50% nominal strain, this error is acceptable: the estimated error is 0.05% maximum apparent strain from out-of-plane component of the displacement field at final fracture zone.

Strain profiles and strain maps were created to show local cumulative engineering strains in time or at a specific point in time, respectively. The color scheme [26] of the strain maps maintains a monotonically increasing perception of intensity, and when converted to black and white, maintains greyscale values with monotonically increasing brightness, whereas the hue variations visualize small differences in the strain field. Local strain rates and Gini coefficients [27] were calculated to characterize the onset and rate of strain localization similarly to ref. [6]. Local strain rates were used instead of cumulative strains or incremental displacements for detection of strain localization, since strain rate is by definition more sensitive to rate of change, and assuming that the time step between the images is sufficiently small, the strain rate values are not affected by the increment of the images. Local strain rates were calculated from the displacement data in vertical direction at each grid point. The local strain rate data was then further refined by utilizing Gini coefficients to extract a single value for the extent of strain localization along a line extending from top to bottom of the gauge section in different x-coordinates across the tensile specimen at each point in time. Gini coefficient indicates the mean of the difference between every possible pair of data points at a given time, divided by the mean size [28]:

\[ g = \frac{\sum_{i=1}^{n-1} \sum_{j=1}^{n} |x_i - x_j|}{2n \sum_{i=1}^{n} x_i} \]  

where \( x_i \) and \( x_j \) = data points along a line extending from top to bottom of the gauge section.

n = number of data points along that line.

Gini coefficient is commonly used as a measure of inequality within a nation's income distribution [27]. While it is not an established method for evaluation of strain localization, it was found to reliably react to variations in strain, and as a global measure of the whole distribution, it is not susceptible to outliers in the data. It can be interpreted as an indication of how evenly strain is distributed on the gauge section of a tensile specimen. The Gini coefficient becomes zero when incremental deformation is perfectly evenly distributed over the whole gauge length and theoretically it should reach the value of one, when deformation is entirely localized between two grid points and no further deformation occurs outside of this section. In practice, the value of one is never reached because the grid spacing is smaller than the spatial resolution of the displacement measurement and the value of zero is never reached because of noise in the data.

Additionally, local “true” stress-strain curves were calculated by combining the force data from the tensile testing machine and the DIC data under the assumption that the stress remains uniform across each cross-section of the specimen. The area reduction of each cross-section was calculated from the y-component of local strain measured by DIC by assuming that the volume of each section remains constant. Simultaneously, the local strains were converted from engineering strains to true strains. The mean strain across each cross-section was then taken as the local value for strain. Note that the assumption of uniform stress across each cross-section is only an approximation, and not even a very good approximation in some parts of the specimen, especially when strain localization occurs. Better estimates of the local stress state may be obtained by a data-driven method [29], but the application of such method is not currently viable for large plastic strains such as in these tests.

<table>
<thead>
<tr>
<th>Table 2</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Calculation settings in DIC.</strong></td>
</tr>
<tr>
<td>Subset size</td>
</tr>
<tr>
<td>Step size</td>
</tr>
<tr>
<td>Correlation method</td>
</tr>
<tr>
<td>Minimum number of valid pixels</td>
</tr>
<tr>
<td>Pyramid levels</td>
</tr>
<tr>
<td>Maximum iterations</td>
</tr>
<tr>
<td>Epsilon</td>
</tr>
<tr>
<td>Threshold for correlation value</td>
</tr>
<tr>
<td>Threshold for confidence margin</td>
</tr>
<tr>
<td>Subset shape</td>
</tr>
<tr>
<td>High accuracy interpolation</td>
</tr>
<tr>
<td>Outlier and smoothing filters</td>
</tr>
<tr>
<td>Fit function</td>
</tr>
</tbody>
</table>
3. Results

3.1. Microstructure of the welds

The microstructure of friction stir welded copper is dynamically recrystallized. Therefore, the grain size of the weld is small in comparison to the base materials. Fig. 2 shows both weld microstructures examined with an optical microscope after polishing and etching. Different zones of the welds are marked and highlighted with dashed lines. The zones become less apparent towards the surface of the welds. The resulting grain size varies in different weld zones depending on the heat input and strain rate during welding. The surface of the weld exhibits significantly larger grains than the root of the weld, caused by the higher heat input from the larger diameter of the tapered welding probe near the surface, and due to the proximity of the rotating shoulder. The root of the weld, on the other hand, experiences less heat, which results in less grain growth. There is a difference between the retreating side (RS) and advancing side (AS) of the welds, as well. Especially in FSWL109, the grain size gradient is more pronounced on the retreating side of the weld. The advancing side, on the other hand, contains a fairly visible shear line, where the flow of material originally split during welding. This line is not as visible in FSWL69. The most significant difference between the two welds is the lack of a clearly visible oxide zone within the stir zone (SZ) of FSWL109. Only a faint meandering line, originating most likely from the original butting surfaces, may be observed.

Fig. 3 shows larger magnifications of the root area and weld boundary area on the retreating/lid side of both welds. These magnified areas are marked with 1 and 2 in Fig. 2, for both welds. In FSWL109, the weld boundary (Fig. 3 a) is difficult to find, whereas in FSWL109 it is easy to locate (Fig. 3 b). EBSD maps, the location of which is marked with red dashed lines, were measured over the boundary to study the grain size transition. In FSWL69, the original joint line transforms into an oxide zone, as shown for the root of the weld in Fig. 3 c). Part of this oxide zone is also visible in Fig. 3 a) on the right-hand side of the image. The oxide particles form a meandering dispersed line or zone, typical to FSW of copper [4]. The dispersion of the particles is affected by the tool design, and in some cases the zone may resemble more like a line of oxide particles than a zone. Dispersion is more efficient towards the surface of the weld. The small particles of size <1 μm in diameter are located on the grain boundaries [4], and thus they may pin grain growth. The particles are difficult to find with conventional methods, but the existence of a line of small grains may reveal their location due to this pinning effect. The exact location of these particles may be revealed by hydrogen annealing at 850 °C, in which hydrogen reacts with the particles forming water, or by electrolytic polishing [4]. Unfortunately, both of these methods remove the particles and deform the surrounding copper lattice. Hydrogen annealing may also lead to grain growth. Thus, direct observation of these particles is difficult.

A similar oxide zone or clearly visible shear line are not visible in FSWL109 in Fig. 3 b) and d). Only a very faint line, originating most likely from the original butting surfaces, may be observed in right-hand side of Fig. 3 b). However, there are some voids near the tip of the weld (Fig. 3 d). This may be explained by poor downward motion of the welded material, which results in poor forging force near the tip of the welding probe due to the current FSW tool geometry [30]. However, in terms of lack of an oxide zone, less visible shear line, and small grain size throughout the weld, the microstructure of FSWL109 weld material seems more uniform than that of FSWL69 weld material.

3.2. Hardness maps

Hardness maps were measured over the two welds to study possible strength mismatch between different weld zones. The hardness maps, shown in Fig. 4 for both welds, are similar in their general appearance. Hardness increases towards the tip of the weld and higher hardness values are observed on the advancing side of the weld than on the retreating side of the weld, in general. This may be explained by lower heat input during welding in the corresponding weld zones. Lower heat input leads to smaller grain size in the dynamically recrystallized stir zone towards the tip of the weld and less recovery of residual stresses and strains outside of the stir zone on the advancing side of the weld. The surface of the weld experiences the highest heat input, which is also visible as lower hardness. However, the lid material of FSWL109 exhibits the lowest hardness values down to 46 HV10 at the bottom right corner of the map and 50 HV10 at the top left corner of the map. Hardness of the lid material of FSWL69 in the same locations is 58 HV10. The average hardness on a line extending from top to bottom of the left-hand side of both maps is 58 HV10 and 54 HV10 for FSWL69 and FSWL109, respectively. These observations indicate a softer lid material of FSWL109.

Hardness may be taken as a measure of resistance to dislocation movement. Since the studied material is almost pure copper, resistance to dislocation movement results mostly from grain boundaries and dislocation density. The oxide particles in FSWL69 may also play a minor role. When taking this into account, it is interesting that similar or even lower hardness values are observed
for some parts of the welds, when compared to the base materials, even if the grain size in the welds is considerably smaller. This means that a drop in dislocation density must occur in the recrystallized weld zone, which is in line with common understanding of dynamic recrystallization. This phenomenon may be observed from the EBSD data, presented in the next chapter, by the evaluation of geometrically necessary dislocation density and measurement of the grain size. When looking at the location of the EBSD maps, marked in Fig. 4 by red dashed lines, a hardness drop is observed over the weld boundary on the retreating side of FSWL69 coming from the lid material towards the weld material, even if the grain size decreases over that boundary. The hardness drops from about 59 HV<sub>IT</sub> to 55 HV<sub>IT</sub> over the boundary in that location. This means that a simultaneous drop in dislocation density must occur.

In FSWL109, a hardness increase from 57 HV<sub>IT</sub> to 60 HV<sub>IT</sub> is observed in the same location coming from the lid material towards the weld material. Due to the limited spatial resolution of the hardness measurements (as data points were taken every 2 mm and smoothing was applied), the hardness transition is blurred in the maps. In reality a sharper transition in grain size and dislocation density occurs, as indicated by the EBSD measurements in the next chapter.

### 3.3. EBSD of the weld boundary

The microstructural observations show a sharper grain size gradient in FSWL109 on the retreating side of the weld when compared to FSWL69. Therefore, EBSD was performed across the weld boundary about 27.5 mm inwards from the surface of the welds from the location indicated in Figs. 2–4. The five consecutive EBSD maps shown in Fig. 5 cover an area of about 9.3 × 1.4 mm in total. The inverse pole figure maps (IPF Z) show random texture, which is typical to FSW of copper [4]. The grain size in FSWL109 changes from more than 200 μm in the lid to about 60 μm in the weld within one EBSD map, which covers a width of 1.8 mm. In FSWL69, grain size changes from about 150 μm in the lid to about 80 μm in the weld much more gradually over several EBSD maps. As a result, the grain size gradient in FSWL109 is considerably sharper than that of FSWL69. The grain size in the stir zone of the welds at the same depth from the surface was also measured. In FSWL69, the...
average grain size across the oxide zone is $78 \text{ \mu m} \pm 6 \text{ \mu m}$. In FSWL109, the average grain size in the same location is $71 \text{ \mu m} \pm 1.5 \text{ \mu m}$.

In addition to grain size, the geometrically necessary dislocation density was calculated from the same EBSD data according to the method of ref. [24]. The dislocation density in FSWL69 decreases towards the weld material, as expected from the hardness drop at the weld boundary. An opposite behavior is observed in FSWL109 as dislocation density increases towards the weld material. This also seems to match with the hardness increase on the retreating side of FSWL109, but the decrease of grain size also naturally affects this. It should be noted, that the absolute values of dislocation densities measured from the EBSD data may be affected, for example, by the quality of the polishing of the samples. Thus, comparison of absolute values measured from different EBSD samples may be problematic, but the trend in change of dislocation density measured from one EBSD sample, is still a reliable indication of differences in the microstructure. Therefore, the smaller grain size and higher dislocation density of FSWL109 may suggest that it was a slightly colder weld, even if the temperature control should maintain the welding temperature at $850^\circ \text{C}$. This may be due to, for example cooling effect of the shielding gas.

3.4. Hydrogen uptake

Hydrogen trapping and uptake was measured by TDS from samples named S1–S5, cut across the two FSW welds from locations shown in Fig. 6 b). The left-over piece shown in Fig. 6 b) (from cutting of the tensile specimens), corresponds to a section of the canister surface shown in Fig. 1 on the right-hand side of the tensile specimen. The samples S1–S5 were cut from different weld zones to study trapping of hydrogen in the different zones. However, due to the homogeneous nature of copper FSW welds, the exact location of these zones is not easily recognizable by the naked eye, and only approximate locations could be selected. The location and size of samples S1–S5 is the same for both welds, even if FSWL109 does not contain an oxide zone. Fig. 6 a) shows the corresponding measured average hydrogen content in wt.ppm in each TDS sample.
cut across the two welds. The TDS specimens represent weld material near the outside surface of the canister, which may experience some hydrogen uptake due to, for example corrosion reactions in the repository conditions. However, the mechanisms of hydrogen uptake will be different from thermal charging used in this study.

Thermal hydrogen charging at 250 °C results in significant increase of hydrogen content in the oxide zone of FSWL69 (sample S3), but it has little effect on the base materials (samples S1 and S5). The maximum allowed hydrogen content in Cu-OPF is 0.6 wt.ppm and the composition measurement in Table 1 indicates the presence of 0.5 wt.ppm hydrogen in the as-received state. Thus, hydrogen is most likely trapped at the oxide particle interfaces, since hydrogen uptake outside of the stir zone was negligible. Similarly, hydrogen uptake in FSWL109 as a whole is almost non-existent, which indicates a markedly reduced number of oxide particles in the weld.

It is known, that hydrogen bubble formation may occur in the copper lattice if the temperature in hydrogen charging is high enough to increase the solubility of hydrogen in the copper lattice considerably (approximately >600 °C). As the material is cooled down, the dissolved hydrogen does not fit in the lattice anymore and bubble formation occurs [14]. However, it is evident from the TDS measurements that this did not occur in the current tests. Hydrogen bubble formation would show as increased hydrogen uptake throughout the weld and base materials, but this did not occur. Therefore, the lack of hydrogen uptake in the base materials and FSWL109 as a whole indicate that no hydrogen bubble formation occurred in the material. Direct observation of these hydrogen bubbles, if they were present in the material, would require the use of transmission electron microscopy (TEM) [14].

Most of the hydrogen in FSWL69 was trapped in the oxide zone (sample S3). Some hydrogen was also trapped in the thermomechanically affected zone (TMAZ) on the advancing side of the weld (sample S2) and weld material on the retreating side of the weld (sample S4). These results indicate that most of the oxide particles, which originate from the original butting surfaces and oxidation during welding, stay in the oxide zone, and that only some oxide particles are dispersed in the rest of the weld. The hydrogen desorption curves are shown in Fig. 7. Hydrogen samples S3 and S2 of FSWL69 weld exhibit several sudden spikes on the low temperature side of the peaks. The spikes are an indication of molecular hydrogen trapped at the oxide particles near the surface of the sample, which burst due to increasing internal pressure as the temperature increases. There are a couple of similar spikes in hydrogen sample S3 of FSWL109, but the number and magnitude of these spikes is significantly lower. The hydrogen desorption profiles of hydrogen charged FSWL109 are similar to the as-supplied weld material.

3.5. Tensile properties

In total eight tensile tests, four for each weld, were performed with two different strain rates and hydrogen charging of both weld materials. Naming of the tensile specimens, fracture location, strain rate, engineering tensile properties, and Gini coefficient onset and slope values are presented in Table 3. Specimens -3H were hydrogen charged (simultaneously with the left-over pieces from cutting of the very same specimens for TDS), and specimens -4 were tested with the higher strain rate. The corresponding engineering stress-strain curves are shown in Fig. 8. The curves match well for the same weld tested with the same strain rate. The welds tested with the same strain rate exhibit similar yield strength and ultimate tensile strength. Specimens FSWL69-4 and FSWL109-4 exhibit higher strength values due to the higher strain rate, but it did not affect the elongations markedly. The yield strength and ultimate tensile strength of these two specimens are similar to those of the base materials previously tested in ref. [6]. However, the most important property of copper in this instance is its ductility. In this regard, it is notable that the elongations of FSWL109 are considerably lower, not meeting the requirement of 40% engineering strain for elongation to fracture [3], in comparison to FSWL69 which fractured at elongations similar to those of the base materials [50–55% [6]]. This indicates earlier strain localization in the newer FSWL109 weld. Thermal hydrogen charging (specimens named -3H), on the other hand, did not compromise the macroscopic ductility of these welds, even if hydrogen trapping oxide particles were present in the FSWL69 weld material.

3.6. Strain localization behavior studied by DIC

Strain localization behavior of the welds was studied with the aid of digital image correlation (DIC). The strain maps, presented in Fig. 9 for most of the tensile specimens, show the peak local strains before the final fracture. In each of the strain maps, the left-hand side is towards the weld root and the right-hand side is towards the surface of the weld, similarly to Fig. 1. Approximate location of the welds is marked with white dashed lines. In FSWL69 fracture occurred either in the oxide zone or at the weld boundary on the lid side of the weld. In FSWL109, fracture occurred always on the base material side of the weld boundary on the lid side, just outside the weld.

The most important finding is that the FSWL109 weld deformed less than the old FSWL69 weld. This may be observed in the strain maps, as well as in the strain profiles presented in Fig. 10. The strain profiles show the peak strains before the final fracture similarly to the strain maps, but it is easier to read the strain values from the profiles. The difference in total elongation to fracture between

Fig. 7. TDS profiles of samples S1–S5 for a) FSWL69 and b) FSWL109, as well as a desorption profile for the as-supplied FSWL69 weld material.
FSWL109 and FSWL69 is explained by local strains outside of the final fracture zone. In FSWL109, local strains outside of the final fracture zone are consistently about 25–30% local engineering strain. However, in FSWL69 they are almost 50% for a wide area. Thus, the lower total elongation to fracture of FSWL109 is the result of lower local strains outside of the final fracture zone. The peak strains, on the other hand, are similar in each of the tensile specimens, indicating similar local ductility. It should be noted, that care should be taken when comparing peak strains obtained by DIC, because their magnitude is affected by the spatial resolution of the DIC measurement and timing of the final image in relation to the fracture event. In this instance, similar test conditions and the same calculation parameters were used for each of the tests, which minimizes this effect.

The onset and extent of strain localization over the whole duration of the tensile test was characterized with the aid of Gini coefficients, calculated for the spatial variation of local strain rates. These are plotted in Fig. 11, using the engineering strain calculated from the elongation of the whole gauge section as the index for time, while overlaying several Gini coefficients for each point in time, corresponding to different x-coordinates across the gauge section. These plots thus illustrate the temporal evolution of the strain distribution on the whole tensile specimen surface. The Gini coefficient initially remains constant, as long as strain is distributed evenly in the tensile specimen. It then increases as the distribution of strain over the gauge section becomes uneven when strain localization occurs. The beginning of strain localization may be evaluated by taking the slope of the increasing Gini coefficient as shown in Fig. 11 a). The obtained onset and slope values are presented in Table 3 for each of the tests.

The Gini coefficients clearly show that the low local strains in FSWL109, in comparison to FSWL69, are the result of earlier onset of strain localization in FSWL109. The onset of strain localization occurs at about 20% elongation of the gauge length in FSWL109, whereas in FSWL69 it occurs at about 30% elongation of the gauge length. These values are roughly 10 %–points earlier than the uniform elongations of both welds, where necking is generally considered to occur. As strains start to localize, deformations outside of the strain localization zone gradually cease. This shows as gradual increase of the Gini coefficient. The transition to steady increase of Gini coefficient (or steady slope of the curve) seems to occur roughly at the same time as uniform elongations occur. It should be noted, that all the specimens, which fractured at the weld boundary show higher Gini slope values. This is explained by the location of fracture near the end of the tensile specimen. A tri-axial stress state forms in the necking region, and it is further enhanced by the tensile specimen geometry near the end of the tensile specimen. Thus, the rate of strain localization was slightly higher after necking occurred in specimens, which fractured at the weld boundary than which fractured at the oxide zone.

Table 3
Naming of the tensile specimens, fracture location, strain rate, engineering tensile properties, as well as Gini coefficient onset and slope values.

<table>
<thead>
<tr>
<th>Specimen</th>
<th>Fracture location</th>
<th>Strain rate (1/s)</th>
<th>Yield strength R_{p0.2} (MPa)</th>
<th>Ultimate tensile strength R_m (MPa)</th>
<th>Uniform elongation</th>
<th>Total elongation to fracture</th>
<th>Gini onset</th>
<th>Gini slope</th>
</tr>
</thead>
<tbody>
<tr>
<td>FSWL69-1</td>
<td>Oxide zone</td>
<td>3.1 x 10^{-7}</td>
<td>80</td>
<td>184</td>
<td>39%</td>
<td>49%</td>
<td>30%</td>
<td>0.064</td>
</tr>
<tr>
<td>FSWL69-2</td>
<td>Weld boundary</td>
<td>3.1 x 10^{-7}</td>
<td>83</td>
<td>182</td>
<td>39%</td>
<td>45%</td>
<td>30%</td>
<td>0.195</td>
</tr>
<tr>
<td>FSWL69-3H</td>
<td>Weld boundary</td>
<td>3.1 x 10^{-7}</td>
<td>80</td>
<td>182</td>
<td>39%</td>
<td>46%</td>
<td>25%</td>
<td>0.109</td>
</tr>
<tr>
<td>FSWL69-4</td>
<td>Oxide zone</td>
<td>3.1 x 10^{-7}</td>
<td>87</td>
<td>200</td>
<td>39%</td>
<td>49%</td>
<td>28%</td>
<td>0.058</td>
</tr>
<tr>
<td>FSWL109-1</td>
<td>Weld boundary</td>
<td>3.1 x 10^{-7}</td>
<td>80</td>
<td>175</td>
<td>31%</td>
<td>35%</td>
<td>20%</td>
<td>0.158</td>
</tr>
<tr>
<td>FSWL109-2</td>
<td>Weld boundary</td>
<td>3.1 x 10^{-7}</td>
<td>80</td>
<td>178</td>
<td>32%</td>
<td>36%</td>
<td>20%</td>
<td>0.136</td>
</tr>
<tr>
<td>FSWL109-3H</td>
<td>Weld boundary</td>
<td>3.1 x 10^{-7}</td>
<td>81</td>
<td>180</td>
<td>32%</td>
<td>35%</td>
<td>18%</td>
<td>0.106</td>
</tr>
<tr>
<td>FSWL109-4</td>
<td>Weld boundary</td>
<td>3.1 x 10^{-4}</td>
<td>93</td>
<td>200</td>
<td>33%</td>
<td>38%</td>
<td>18%</td>
<td>0.100</td>
</tr>
</tbody>
</table>

3.7. Local stress-strain curves

Local “true” stress-strain curves were calculated by combining the force data of the tensile testing machine and the local strain data from DIC, under the assumption that stress remains uniform across each originally horizontal cross-section of the tensile specimen. Note that this is only an approximation, and the real stress state differs from this approximation, especially when strain localization occurs. The local stress–strain curves are shown in Fig. 12a) and b) for two of the FSWL69 specimens, which fractured at the oxide zone. The local peak stress–strain curves were extracted from each of the tests and they are shown in Fig. 12 c). The local stress increases as long as fracture occurs except for FSWL109-4. This may be explained by partial fracture of the gauge section before the final fracture.

4. Discussion

Localization of plastic deformation in these welds is potentially of concern for their use in spent nuclear fuel disposal canisters. The primary purpose of the copper shell is to perform as a ductile corrosion barrier, whereas a cast iron insert inside the shell provides the principal structural strength against external loads. In case the loads are high enough to cause deformation of the cast iron insert, the copper shell should accommodate that by deforming itself. Therefore, the most important property of copper, besides the corrosion resistance, is its ductility, whereas tensile strength is not as important.

The ductility may be locally compromised by strain localization due to, for example microstructural heterogeneity and residual stresses introduced by welding of the lid to the tubular body of the canisters. The lids are welded by FSW, which results in dynamic
recrystallization of the weld and small grain size in comparison to
the base materials. The small grain size does not necessarily mean
that the weld material is considerably stronger than the base ma-
terials due to recovery of residual stresses and strains in the
recrystallized weld, but if such difference exists, it is likely to cause
strain localization. Therefore, hardness over the welds was
measured. On average hardness of both weld materials is similar.
Hardness increases from the surface towards the tip of the welds,
and higher hardness is observed on the advancing side than on the
retreating side of the welds. However, there is a difference between
the lid materials, the lid material of the new weld being softer than
the lid material of the old weld. The grain size is also larger in the lid
material of the new weld. The lids were manufactured at different
times by different manufacturers and the forging process may have
been as well slightly different. Therefore, it seems that the initial
state of the lid materials was different.

Oxide particles will be present in the FSW welds if welding is
performed without shielding gas [4] and copper is known to suffer
from hydrogen embrittlement at elevated temperatures [16] in the
presence of oxide particles. Therefore, the welds were thermally
hydrogen charged to study the possibility of hydrogen embrittle-
ment in the weld oxide zone. It was con

confir

firmed, that thermal
hydrogen charging results in considerable hydrogen uptake at the
oxide zone. Hydrogen is most likely trapped at the oxide particle
interfaces, since hydrogen uptake outside the oxide zone was
negligible. The hydrogen charged FSW welds were tensile tested
similar to the other specimens, but in this instance the trapped
hydrogen did not compromise the macroscopic ductility of these
FSW welds. Diffusible hydrogen has been shown to affect the
deformation of single crystal copper by activating localized slip
[11], but if this mechanism was active in the current tests, less
macroscopic deformation would be expected. On the other hand,
according to a recent modeling study hydrogen stabilizes vacancies in copper [13] and by doing so, it may as well affect the creep cavity nucleation [18,19]. This may be relevant in the repository conditions, but hydrogen embrittlement of the oxide zone seems unlikely. In these tests embrittlement did not occur since the temperature in thermal hydrogen charging (250 °C) was not high enough to induce water vapor formation at the oxide particles. The temperature of the copper shell is not expected to exceed 100 °C, and therefore it seems unlikely that similar hydrogen embrittlement of the FSW welds would occur. It is more likely that the cast iron insert may suffer from hydrogen embrittlement than the copper shell due to higher sensitivity of cast iron to hydrogen [31]. The FSW welding method has recently been modified by shielding gas to reduce oxidation during welding. It is apparent that the new weld is improved in sense of less oxide particles and more uniform microstructure within the weld itself, but the DIC results indicate stronger strain localization at the weld boundary on the retreating side of the weld on the lid side of the canister. The main known difference between the old and the new FSW welds is the utilization of shielding gas, but as concluded above it seems that the initial state of the lid materials was also different. The combination of a softer lid material with larger grain size, smaller grain size in the weld, and sharp grain size gradient between the two, conditions for enhanced strain localization at the weld boundary of the new weld are created. This leads to considerably lower elongation to fracture, not meeting the requirements of 40% engineering strain [3] when measuring over the weld. The weld material could probably withstand more strain than that of the old weld as the microstructure is more uniform, but the steep grain size gradient on the weld boundary causes strain localization to occur elsewhere earlier. To minimize the risk of strain localization, the strength mismatch and grain size gradient between the weld and the base materials should be minimized when manufacturing the real canisters. It should also be taken into consideration that the weld microstructure may vary along the long weld. The tensile specimens in this study were cut from segments relatively close to each other, but cutting the specimens from near the end of the weld cycle may yield different results.

All the specimens that fractured at the weld boundary, fractured on the base material side of the boundary. However, in conditions relevant to the geologic repository, with creep occurring in the material, fracture may occur on the weld side, since in creep conditions grain boundary sliding becomes a significant factor [19] and the weld has a smaller grain size. These tests were not slow enough and the temperature was not high enough for this to occur. It should be noted, that the weld boundary was located near the end of the tensile specimens. Therefore, end-effects from the specimen geometry affected the local stress state at the final fracture zone. This shows as increased rate of strain localization after necking occurred, as indicted by the Gini coefficient slope values. Longer tensile specimens would help in avoiding this problem, but in this instance it is not possible to manufacture longer specimens due to the canister geometry.

Gini coefficients were found useful for characterizing the strain

![Fig. 10. Strain profiles of a-c) FSWL69 and d-f) FSWL109 before final fracture. The lower total elongation to fracture of FSWL109 in comparison to FSWL69 is the result of lower local strains outside of the final fracture zone. Peak strains are similar in each specimen. The coordinates refer to initial positions of the DIC grid points in the beginning of the tensile test, and therefore they do not match with the coordinates in the strain maps.](image-url)
Fig. 11. Gini coefficients of a-c) FSWL69 and d-f) FSWL109 for the whole duration of the test. Gini coefficient is plotted versus the engineering strain of the whole gauge length. The onset and slope of Gini coefficients were determined to quantify the beginning and rate of strain localization.

Fig. 12. Local “true” stress-strain curves for a) FSWL69-1 and b) FSWL69-4. c) Local “true” peak stress-strain curves for each of the specimens.
localization behavior in these tests. The absolute values of Gini coefficients are affected by the noise in the data, and therefore a direct comparison of the Gini coefficient values may not be meaningful, unless all the test conditions are similar and computational factors are the same. In this study, the test conditions were as similar as possible and the same calculation settings were used for each of the tests. When this is taken into account, Gini coefficients are a robust, yet sensitive measure of strain localization in a tensile test observed by DIC. As a global measure of strain localization the values are not susceptible to outliers in the data.

5. Summary and conclusions

This paper compares the deformation and strain localization of two FSW welds of copper canisters in the context of spent nuclear fuel disposal. The main known difference between the FSW welds is the utilization of shielding gas during welding, although the initial condition of the lid materials was most likely also different. The strain localization behavior was monitored by DIC and quantified by Gini coefficients. In addition, both welds were thermally hydrogen charged to study hydrogen trapping by oxide particles in the welds. The results can be summarized as follows:

- The old weld fractured at the oxide zone or at the weld boundary on the lid side of the canister. Elongation over the weld was similar, independent of the fracture location.
- The new weld fractured always at the weld boundary on the lid side of the canister.
- The new weld deformed significantly less than the old weld due to earlier strain localization.
- The hardness of the lid material of the new weld is lower and the grain size is larger. This indicates a difference in the forging of the lids.
- Thus, the earlier strain localization in the new weld was caused by the weaker lid material and a steep grain size gradient between the weld and the lid.
- Oxide particles in the weld trap considerable amounts of hydrogen, but the effect of trapped hydrogen on the macroscopic plastic deformation in these tests was negligible.
- Shielding gas reduced the number of oxide particles in the weld considerably, which also resulted in markedly reduced hydrogen uptake.
- Gini coefficients, calculated for the spatial variation of local strain rates, are useful in characterizing strain localization in a tensile test monitored by DIC.

Declarations of interest

None.

Data availability

The raw/processed data required to reproduce these findings cannot be shared at this time due to technical or time limitations.

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