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Research

Development of a constitutive model for the plastic deformation and creep of copper and its use in the estimate of the creep life of the copper canister

Kjell Pettersson

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SKI

SKI perspective

Background and purpose of the project

The copper-iron canister for disposal of nuclear waste in the Swedish Programme has a design life exceeding 100,000 years. Whilst the operating temperature and operating stress are modest, the very long design life does require that the likely creep performance of the canister should be investigated. Many studies have been carried out by SKB but these have all involved very short duration tests at relatively high stresses.

Reported creep tests on oxygen free (OF) copper have demonstrated that copper can have extremely low creep ductility. However with the addition of about 50 ppm phosphorus to the copper it appears as if the creep brittleness problem is avoided. OFP copper has consequently been chosen as the canister material. One of the requirements for the copper used in the waste canister is a creep ductility of 10 % [SKB TR-06-09]. The information presented so far by SKB has not demonstrated conclusively that the OFP copper actually have this ductility at the canister operating temperatures.

A previously developed model for the plastic deformation and creep of copper (included as an Appendix to the present report) has been used as the basis for a discussion on the possibility of brittle creep fracture of the copper canister during long term storage of nuclear waste in this project.

Results

Reasonable assumptions on the temperature dependence of the creep brittle process and the fastest such process inaccessible by the SKB experiments on OFP copper still lead to the conclusion that failure by the creep brittle process can not occur despite the fact that stress in the canister can remain at about 100 MPa for thousands of years after an earthquake event.

Effects on SKI work

The study will be a basis for coming SKI research projects and SKI reviews of SKB's RD&D-programme and licence applications.

Project information

Behnaz Aghili has been responsible for the project at SKI.
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Development of a constitutive model for the plastic deformation and creep of copper and its use in the estimate of the creep life of the copper canister

Kjell Pettersson
Matsafe AB
Skeppargatan 84
114 59 Stockholm
Sweden

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Summary

A previously developed model for the plastic deformation and creep of copper (included as an Appendix to the present report) has been used as the basis for a discussion on the possibility of brittle creep fracture of the copper canister during long term storage of nuclear waste. Reported creep tests on oxygen free (OF) copper have demonstrated that copper can have an extremely low creep ductility. However with the addition of about 50 ppm phosphorus to the copper it appears as if the creep brittleness problem is avoided and that type of copper (OFP) has consequently been chosen as the canister material. It is shown in the report that the experiments performed on OFP copper does not exclude the possibility of creep brittleness of OFP copper in the very long term. The plasticity and creep model has been used to estimate creep life under conditions of intergranular creep cracking according to a model formulated by Cocks and Ashby. The estimated life times widely exceed the desing life of the canister. However the observations of creep brittleness in OF copper indicate that the Cocks-Ashby model probably does not apply to the OF copper. Thus additional calculations have been done with the plasitcity and creep model in order to estimate stress as a function of time for the probably most severe loading case of the canister with regard to creep failure, an earth quake shear. Despite the fact that the stress in the canister will remain at the 100 MPa level for thousends of years after an earth quake the low temperature, about 50 ˚C or less, will make the solid state diffusion process assumed to control the brittle cracking process, too slow to lead to any significant brittle creep cracking in the canister.

Sammanfattning

En tidigare utvecklad modell för krypning och plastisk deformation hos koppar (inkluderad som appendix till denna rapport) har använts som utgångspunkt för en diskussion om möjligheten av att sprött krypbrott kan inträffa i kopparkapseln för långtidslagring av högaktivt kärnavfall. Utförda provningar på syrefri koppar (OF) har visat att koppar kan ha en extremt låg krypduktilitet. Emellertid leder en tillsats av ca 50 ppm fosfor till kopparn till att den låga krypduktiliteten försvinner och sådan koppar (OFP) har därför valts för tillverkning av avfallskapslarna. I rapporten visas att de provningar som utförts på OFP-koppar inte utesluter att denna kan ha låg krypduktilitet på mycket lång sikt. Modellen för krypning och plastisk deformation har använts för att uppskatta kryplivslängden vid pågående interkristallin krypspricktillväxt enligt en teori formulerad av Cocks och Ashby. Uppskattade livslängder överstiger med god marginal önskad livslängd för avfallskapseln. Emellertid är det så att de observationer som gjorts av spröda krypbrott i OF-koppar tyder på att Cocks-Ashbyteorin inte gäller för detta material. Därför har ytterligare beräkningar gjorts med modellen för krypning och plastisk deformation för att få en uppskattning av spänningen som funktion av tiden för det troligen mest svårartade belastningsfallet för avfallskapseln med tanke på krypbrott: skjuvning på grund av jordbävning. Trots att spänningen i kapseln kommer att ligga på nivån 100 MPa under tusentals år efter en jordbävning leder den låga temperaturen, 50 ˚C eller lägre, till att risken för sprött krypbrott är försumbar. Orsaken är att mekanismen för det spröda krypbrottet antagits innefatta diffusion av atomer i kopparn. Denna diffusionsprocess går på grund av den låga temperaturen för långsamt för att krypbrottsprocessen skall hinna verka under kapselns livslängd.

1. Introduction

One of the potential problems for copper canisters intended for storage of high level nuclear waste is failure by the growth of creep cracks. The subject has been reviewed recently by Bowyer [1]. In the review Bowyer presents estimates of the life of creep specimens made of oxygen free (OF) copper and compares them with actual observations of creep life. The estimates are based on theories of creep fracture reviewed and developed by Cocks and Ashby [2]. Bowyer has concluded that the form of intergranular creep failure most likely to occur in the stress and temperature range of interest for the waste canister as well as for the creep test specimens is void growth by power-law creep. In this form of failure voids in the grain boundaries are nucleated by some as yet undetermined mechanism and they start to grow because the stress concentrations associated with the voids make the material between voids to creep faster than the interior of the grains. Bowyer has also concluded that the other mechanisms treated by Cocks and Ashby, void growth under boundary diffusion control or under surface diffusion control, are less likely to be important other than in the early stages of void growth.The present writer has reached a similar conclusion presented at an SKI workshop in 2002 [3].

The estimates presented by Bowyer show that the life of the OF creep specimens has generally been much lower than estimated from the Cocks-Ashby theory. The OF creep specimens were available in 2 variants, one with 10 ppm sulphur and one with 6 ppm sulphur [4]. The 10 ppm material failed with very low strains while the 6 ppm failed with about 10 % strain. This is also reflected in the difference between Bowyer's life estimates and the actual life of the creep specimens. For the OF specimens with 10 ppm S the estimated life is typically 100-1000 times higher than the actual life of a specimen while for the 6 ppm variant the life is overpredicted by a factor of 3. These predictions depend on the creep properties and since these may be expressed somewhat differently depending on how one evaluates critical parameters the estimation by this writer overpredicted the life of OF material with 6 ppm sulphur by a factor of about 20 [3]. (In the actual PowerPoint presentation a factor of 2 is given, but it has subsequently been revealed that the estimate is wrong by a factor of 10).

Due to the creep brittle behaviour of the OF material work started on material to which about 50 ppm of phosphorus had been added, the OFP material. This material has so far never been tested under conditions when it has shown creep brittle behaviour and is generally considered to be ductile. When Bowyer applied the Cocks-Ashby theory to creep tests performed on this material there was generally a fairly good agreement between actual and estimated creep life [1]. The question remains however how this material can be expected to behave under conditions relevant to the waste canisters.

2. The Cocks-Ashby theory.

The creep life when intergranular void growth is controlled by power-law creep is given by

$$t_f = t_n + \frac{1}{\beta(n+1)\dot{\varepsilon}_0}\ln\left(\frac{1}{(n+1)f_i}\right)\left(\frac{\sigma_0}{\sigma}\right)^n \qquad (1)$$

where t_n is the time when voids nucleate, β a constant and f_i the initial fraction of the grain boundary covered with voids. The other constants come from the formulation of Norton's law as

$$\dot{\varepsilon} = \dot{\varepsilon}_0\left(\frac{\sigma}{\sigma_0}\right)^n \qquad (2)$$

If Norton's law instead is written as

$$\dot{\varepsilon} = A\sigma^n \qquad (3)$$

then the creep life equation takes the form

$$t_f = t_n + \frac{1}{\beta(n+1)A}\ln\left(\frac{1}{(n+1)f_i}\right)\sigma^{-n} \qquad (4)$$

3. Estimate of the creep life of OFP copper under waste storage conditions

Recently this writer used a model for the creep of OFP copper to extrapolate the creep properties down to 75 and 100 °C [5]. The model is reproduced as Appendix A of the present report [6]. In the extrapolation report [5] it is demonstrated that it is probable that power-law creep is the dominating mechanism also at these lower temperatures. The Norton's law expressions at the two temperatures are

$$\dot{\varepsilon} = 4.79\times10^{-22}\sigma^{6.16} \qquad 100°C$$
$$\dot{\varepsilon} = 3.85\times10^{-23}\sigma^{6.17} \qquad 75°C$$

where the unit for stress is MPa and for strain rate h^{-1}. (In the original report the unit for strain rate was erroneously given as s^{-1})

To get a conservative estimate of the creep life it may be assumed that the nucleation time is 0. Then the constants A and n can be directly inserted in (4) and we get the following estimated creep life at a stress of 50 MPa:

100 °C: 4,800,000 years
75 °C 60,000,000 years.

This is obviously quite a sufficient margin with regard to the needed life time of the waste canisters.

4. Discussion

There is a difference between the life estimates presented by Bowyer [1] and those presented by this writer in 2002 [3]. But a common theme is that both estimates strongly overpredict life of OF copper with 10 ppm S while the overprediction for 6 ppm is only a factor of 3 for Bowyer and about 20 for the present writer. The reason for the difference is that Bowyer has used a simplified formulation based on the assumption of constant load while this writer has used the full expression for constant stress. For the OF specimens with 10 ppm the difference should be very small since they only survived about 1 % strain and the difference between constant load or constant stress is miniscule. For the material with 6 ppm the difference is somewhat greater and it is likely that the estimate presented in Section 3 overpredicts life compared to an estimate. which takes proper account of the fact that the tests were done with constant load. A main point however with the estimates for the 6 ppm material is that theory overpredicts

life by a factor of 3-10. It is now quite reasonable to argue that the OFP material is more like the 6 ppm material than the 10 ppm material and thus we could at most expect theory to overpredict the life of the OFP material by a factor of 10. And if we accept that argument we can safely say that the OFP material has adequate creep fracture properties for use as canister material.

It is also possible to argue in an opposite way. Let us assume that the theory is in fact unreliable and that it leads to overpredictions so that the actual life of the canister material at 75 °C and 50 MPa is only 100000 years i. e. an overprediction by a factor of 600. At what stress would we need to test at 215 °C in order to get intergranular creep failure in 1000 hours if the same mechanism is responsible for failure? The answer is 76 MPa which is in fact lower than actual test stresses. Tests performed at 100, 120 and 140 MPa gave lives of > 5520 h, 7848 h and 1451 hours respectively. Thus the creep specimens were exposed to higher stress than 76 MPa for a longer time than 1000 hours and there was no sign of any creep brittle failure as it surely would have been if a mechanism of the Cocks-Ashby type which gives a 100000 year life at 75 °C had been at work. So any which way one would like to argue there seems to be ovewhelmingly clear that the OFP copper is sufficiently creep ductile in the canister application.

However there is a fundamental flaw in the discussion above. Since the Cocks-Ashby model leads to severe overpredictions of creep life for the OF copper the only thing we can conclude is that the model does not apply to the OF copper. What we know is that the creep brittle mechanism is affected by the amount of sulphur in the material and that the addition of phosphorus apparently leads to a decreased effect of the mechanism. The crucial question is whether or not the mechanism has been eliminated by the addition of phosphorus. One of SKB:s main consultants regarding questions of creep fracture has argued at some length in a KASAM report that the OFP copper has an adequate ductility for use as waste canister material [7]. The problem is that the conclusion is based on a series of creep tests in which the specimens have failed in a ductile manner. Why this may be a problem is illustrated in Figure 1. In the Figure data of the creep lives of OF (OFHC) copper and OFP copper at 215 ˚C have been plotted. It is clear that the creep lives have different stress dependencies for the two materials as would be expected since one of the materials fails with the creep brittle mechanism and the other by ductile failure [2, 8]. For the former mechanism the stress exponent is numerically small, theoretically 1-2 [2, 8] but in the Figure about 6, while for the ductile mechanism

the stress exponent will roughly be the same as the stress exponent in Norton's law, since

$$t_f \cong \varepsilon_f / \dot{\varepsilon} \qquad (5)$$

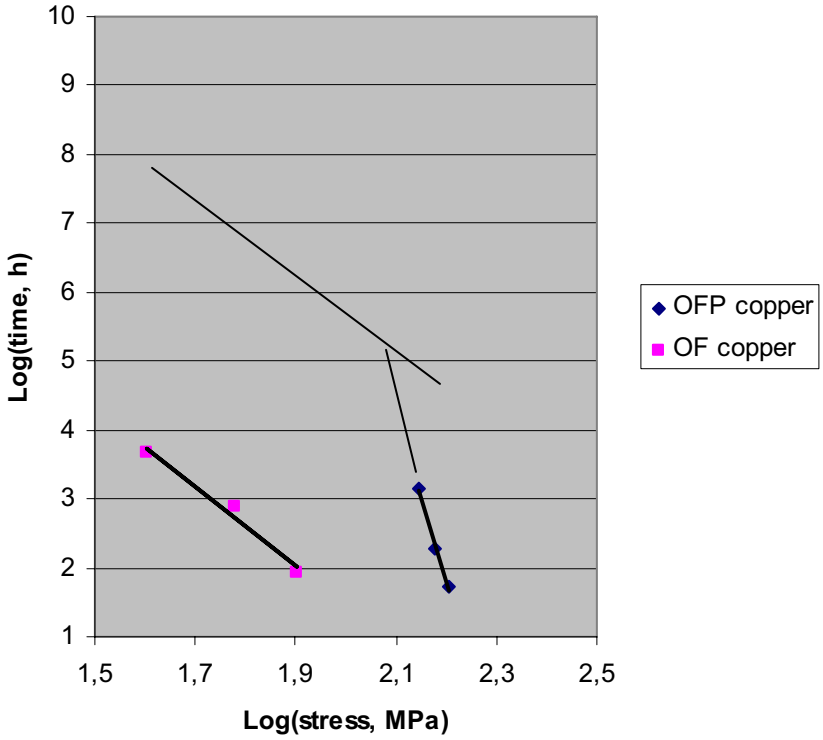

Figure 1. Time to failure as a function of stress for specimens tested in [4].

The thin line parallel with the failure line for OF copper illustrates a hypothetical situation in which the creep brittle mechanism has been decelerated by a factor of 10000, perhaps by the addition of phosphorus. If such a situation existed the creep brittle mechanism would only be accessible by testing for more than 230000 hours at a stress a stress below about 104 MPa. Testing at higher stresses would lead to ductile failures in shorter times. Therefore in our hypothetical situation the creep-brittle mechanism could be at work in OFP copper but it would be inaccessible by practical experiments. If this creep life equation applies to the OFP copper it would have a creep life of just 2000 years at a stress of 50 MPa. It seems quite possible to envisage scenarios in which this level of stress could be present for a long time after an earth quake shear. (An example will be given later.) With regard to creep ductility of the OFP material there is in fact no adequate information available at the present time. It should

be clear from the discussion above that the SKB tests on OFP copper tell us little about the relevant ductility. Thus it is currently uncertain whether or not the OFP copper fulfils the specification of 10 % creep ductility quoted in the SR-Can Report [9].

There is however a possibility to produce data on the creep brittle mechanism and that is to use triaxial stress states in which high tensile stresses can be combined with low shear stresses. Thus the creep brittle mechanism which depends on tensile stress will be accelerated in comparison with the ductile mechanism which depends on shear stress. This has to some extent already been done for the OFP copper by Auerkari and coworkers at VTT [10, 11]. They have tested CT specimens of copper at constant load and for a rather long time. The evaluation of the results have been according to the LICON methodology as described for instance by Bowyer [12]. The LICON methodology relies on a time to failure equation which contains a stress intensification factor H and a deformation mode dependent reference stress. Bowyer notes that the equation is purely empirical and expresses that some of the content of the equation must be taken on trust. One important and promising result of the VTT tests on a CT specimen is the relatively low density of creep cavities in the specimen after the test [10]. Another relevant observation was that the density of cavities was about the same on the surface as in the interior after the test. This latter observation might be an indication that stress triaxiality plays a smaller role than expected for nucleation and growth of creep cavities. Finite element calculations showed that after 64000 h of testing the tensile stress at the crack tip still was about 130 - 360 MPa which would be well above the hypothetical failure line in Figure 1. However in view of the fact that this was a cold worked material and just one test it is not sufficient proof that the OFP copper has no creep brittle behaviour.

The preceding discussion has not taken temperature effects into account. The data in Figure 1 concern 215 ˚C. The VTT experiments were performed at 150 ˚C and the critical temperatures for the copper canister is in the range 20 - 90 ˚C as shown in Figure 2 which is reproduced from [9]. At lower temperatures the creep brittle process will be slower. Since it is a solid state process it is reasonable to assume that it is controlled by a solid state diffusion process with its characteristic activation energy. The lowest activation energy for diffusion in copper is that for boundary diffusion which

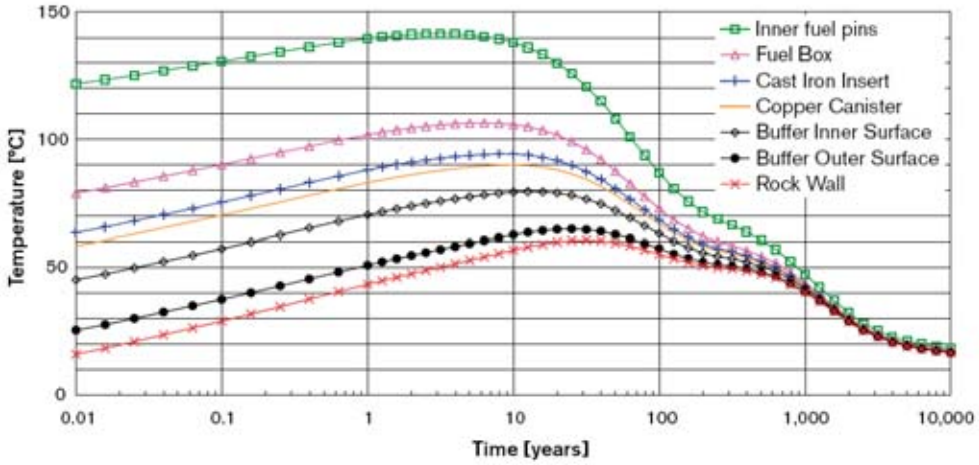

Figure 2 Temperatures in the canister and canister environment for a typical location.

is 104 kJ/mol [13]. If we apply that activation energy to the creep brittle process we can get an estimate of how much longer the creep life would be at constant stress compared to the creep life at 215 ˚C. The data is shown in Table 1.

Table 1

Creep lives relative to the life at 215 ˚C

Temperature ˚C	Creep life relative to life at 215 ˚C
215	1
150	51
100	2710
75	30100
50	486000
25	1.25×10^7

This table indicates that even if the hypothetical situation regarding creep brittlenes depicted in Figure 1 existed, there should be little concern for brittle creep fracture since the creep-brittle process is sufficiently decelerated by the temperature to reduce the risk of brittle fracture. The creep life of 2000 years at 50 MPa in Figure 1 is extended to 2.7 million years at 100 ˚C. As can be seen in Figure 2 the canister only spends about 10 - 100 years at this temperature level.

The margin against creep brittle failure is further reinforced by calculations of the stress level in the canister after an earthquake event using the constitutive model developed within the present work [6]. In the calculations it is assumed that during the earthquake the canister is deformed 5 % in tension in 1 s. The material remains in that deformation state and the stress relaxes by thermally activated deformation and creep for the remainder of the calculation which has been followed for 2000 years. The result is shown in Figure 3.

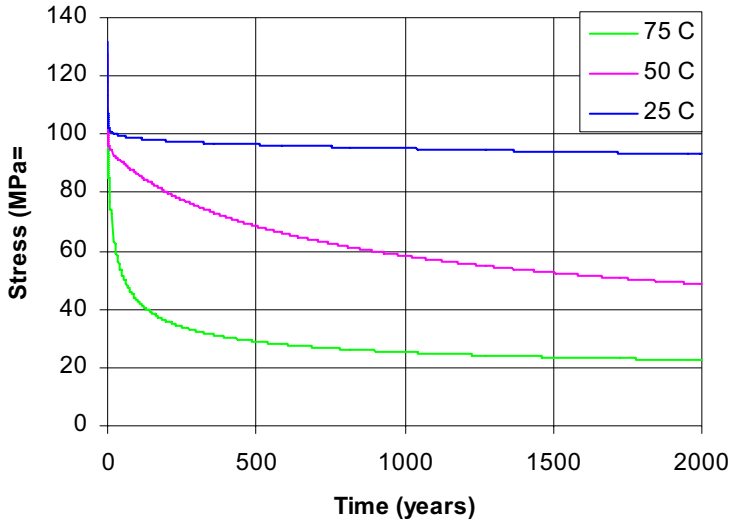

Figure 3 Estimated stress level in canister after an earthquake event at different temperatures.

Due to the scale in Figure 3 the initial stress levels at the start of relaxation is not visible. It is 131, 128 and 124 MPa at 25, 50 and 75 ˚C respectively. The Figure shows that at 75 ˚C the stress quickly goes below 40 MPa, but as can be seen in Figure 2 this takes about the time that the canister spends at that temperature. At 50 ˚C the stress relaxes to about 50 MPa in 2000 years. This is longer than the time the canister spends in this temperature range according to Figure 2. Only at 25 ˚C does stress remain at a high level for a long time. Figure 2 shows that the canister stays at 25 ˚C or lower for a long time. However at the level of 100 MPa the life in the hypothetical situation of Figure 1 would be 30 years at 215 ˚C and with the extension factor shown in Table 1 a predicted life would be in the order of 4×10^8 years.

Another factor of importance after the eaerthquake event is the effect of cavity growth on relaxation. The elastic modulus of copper is about 120000 MPa. It thus only requires

about 0.083 % strain to relax a stress of 100 MPa. Even if the contribution of cavity growth to strain is not quantifiable it is probably fair to say that it provides an additional margin to failure by the creep brittle mechanism.

5. Conclusions

One of the requirements for the copper used in the waste canister is a creep ductility of 10 % [9]. The information presented so far by SKB has not demonstrated conclusively that the OFP copper actually have this ductility at the canister operating temperatures. However reasonable assumptions on the temperature dependence of the creep brittle process and the fastest such process inaccessible by the SKB experiments on OFP copper still lead to the conclusion that failure by the creep brittle process can not occur despite the fact that stress in the canister can remain at about 100 MPa for thousands of years after an earthquake event.

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Further development of a constitutive model for the plastic deformation and creep of copper. SKI project 14.9-020902:02184.

Kjell Pettersson
Matsafe AB.

Summary

A previously available model for the plastic deformation and creep of copper has been developed further. The model is based on the idea that the strain rate at a moment of time is dependent on the applied stress and the microstructural state of the material. In the original version of the model the dislocation density was used as the sole variable to characterize the microstructural state. The strain rate is determined by a thermal activation mechansm dependent on stress and dislocation density. The development of dislocation density is tracked through a differential equation which describes how it changes with strain and time. In the further development of the model the generation of vacancies with plastic deformation is taken into account. The presence of vacancies is potentially important since they increase volume self diffusion and thus contribute to recovery of the dislocation structure. In the original model dislocation core diffusion was dominant at low temperature. Calculations show that if the presence of vacancies is taken into account volume diffusion dominates over core diffusion at low strains. However at higher strains core diffusion is still dominant. This means that volume self diffusion contributes little to the stationary creep rate of copper at temperatures below about 300 °C. An indirect conclusion from this is that the effect of phosphorus on copper creep strength can not be due to interaction between vacancies and phosphorus. The calculations also showed that in order to get agreement between calculated and observed secondary creep rate at different temperatures, it was necessary to assume that the activation energy for core diffusion was lower than the previously published value. In a special case where no agreement between calculation and experiment could be reached this was interpreted as an effect of anisotropy induced by plastic deformation, an effect which probably must be taken into account for a proper modelling of copper.

Vidareutveckling av en konstitutiv ekvation för plastisk deformation och krypning hos koppar..

Kjell Pettersson
Matsafe AB

Sammanfattning.

En tidigare befintlig modell för plastisk deformation av koppar har utvecklats ytterligare. Modellen är baserad på att materialets töjningshastighet vid en viss tidpunkt beror på pålagd spänning och materialets mikrostruktur. I den ursprungliga modellen användes dislokationstätheten som den enda storheten för att karakterisera mikrostrukturen. Töjningshastigheten fås genom termisk aktivering som beror av spänning och dislokationstäthet. Utvecklingen av dislokationstätheten följs med en differentialekvation som beskriver hur denna ändras med tid och töjning. I vidareutvecklingen av modellen tas hänsyn till bildning av vakanser genom den plastiska deformationen. Närvaron av vakanser är potentiellt viktig eftersom dessa ökar volymssjälvdiffusionen och därmed bidrar till återhämtningen av dislokationsstrukturen. I den ursprungliga modellen var diffusion genom dislokationernas kärnor helt dominerande vid låga temperaturer. Beräkningar visar att om man tar hänsyn till bildning av vakanser kommer volymsdiffusion att dominera vid låga töjningar. Vid högre töjningar blir dock kärndiffusion åter dominant. Detta innebär att volymsdiffusion har liten inverkan på sekundärkryphastigheten i koppar vid temperaturer under ca 300 °C. En indirekt slutsats av detta är att fosfors inverkan på kryphållfastheten koppar inte kan bero på växelverkan mella fosforatomer och vakanser. Beräkningarna visade också, att det för att få överensstämmelse mellan beräknad och observerad kryphastighet vid olika temperaturer, var nödvändigt att använda en lägre aktiveringsenergi än den publicerade för diffusion i dislokationskärnorna. I ett speciellt fall där det inte gick att få överensstämmels mellan beräknad och observerad kryphastighet tolkas detta som en effekt av anisotropi framkallad genom plastisk deformation, ett fenomen som man förmodligen måste ta hänsyn till för att få en helt godtagbar modellering av kopparns egenskaper.

1. Introduction

Relatively pure copper will be used as a canister material for the long term storage of high-level nuclear waste. In the development of a safe canister it is necessary to perform numerous structural calculations of stresses and strains of the canister. These calculations will be performed for short term loadings and deformations during manufacture and accident conditions as well as for long term conditions during storage. Another situation in which a good constitutive description is desirable is when creep crack growth is accelerated experimentally by subjecting the material to a triaxial stress state in order to obtain high tensile stresses [10]. Since the level of the stress depends on both strength and creep properties the interpretation of results will depend on how well the stress can be calculated. In a previous report [14] a constitutive model for the plastic deformation and creep of copper was presented. In the present report this model is developed further and a few corrections based on an earlier false interpretation of the data used to calibrate the model are introduced.

2. The model

2.1. Description of the first version of the model.

The model used is a combination of the mechanical model proposed by Bergström for the strain hardening of metals [15] and the recovery creep model as it has been formulated by Lagneborg [16]. The two models are tied together through a rate equation describing the thermally activated motion of dislocations. The rate equation is

$$\dot{\varepsilon} = \dot{\varepsilon}_o \exp - \frac{H(\tau^*)}{kT} \qquad (1)$$

where H(τ*) is a stress dependent activation energy. The preexponential factor $\dot{\varepsilon}_o$ is estimated from the activation frequency ν_o, the number of activation sites per unit volume, and the average area swept after each succesful activation. The mechanism modelled is the cutting of forest dislocations. The volume density of activation sites then is about $\rho^{3/2}$ where ρ is the dislocation density. The area swept at each activation event is about ρ^{-1}. Thus the pre-exponential factor is given by

$$\dot{\varepsilon}_o = \frac{b\rho^{1/2}\nu_o}{\overline{m}} \qquad (2)$$

where $\bar{m}$ is the Taylor factor which relates the shear deformation of the grains to the tensile deformation ε. The activation energy at zero stress has been estimated to be $H_o = 0.5Gb^3$ for cutting of forest dislocations [13]. The activation area is about $b\cdot\rho^{-1/2}$.

This leads to the following expression for the activation energy H where v* is the activation volume, the product of the Burgers vector b and the activation area:

$$H(\tau^*) = H_o - v^* \tau^* = H_o - b^2 \rho^{-1/2} \left(\tau - \tau_o - \alpha G b \sqrt{\rho}\right) \quad (3)$$

I (3) we have assumed that the effective stress τ^* is given as the difference between the applied stress τ and the sum of the friction stress τ_o and the strain hardening term $\alpha Gb\sqrt{\rho}$. In terms of tensile stresses the expression is

$$H(\sigma^*) = H_o - \frac{b^2 \rho^{-1/2}}{\overline{m}} \left(\sigma - \sigma_o - \alpha_m G b \sqrt{\rho}\right) \quad (4)$$

where the subscript m on α denotes that it includes the factor $\bar{m}$. The dislocation density ρ serves as a microstructural parameter which together with the applied stress σ determines the strain rate. For the development of ρ with time and strain we use a combination of the formulations by Bergström and Lagneborg:

$$\frac{d\rho}{d\varepsilon} = \dot{\varepsilon} \frac{d\rho}{d\varepsilon} - M D_s \rho^2 \quad (5)$$

where

$$\frac{d\rho}{d\varepsilon} = \frac{\overline{m}}{bs} - \Omega\rho - A\rho^2 \quad (6)$$

In (6) the first term represents the increase in dislocation density due to dislocation multiplication through loop expansion and generation of new loops in dislocation sources. The second term takes account of the decrease in the efficiency of dislocaton generation with increasing density. Bergström describes it as "remobilization", which can be understood in the following way: instead of all strain being caused by the expansion of loops some of the strain is now caused by some previously stopped dislocations being remobilized, which is a less efficient way of increasing the density. The last term takes account of dynamic recovery: with increasing dislocation density the probability of mutual annihilation of dislocations of opposite sign will increase. The probability will reasonably be proportional to ρ^2. By the same argument the thermal recovery term in eq. 5 will be proportional to ρ^2 and also proportional to the self diffusion coefficient D_S. M is a mobility factor which will be used as an adjustable parameter in the model. The mean free path s is not constant for fcc metals and it has been shown [15] that a reasonable model for its strain dependence is

$$s = s_o + (s_1 - s_o)\exp - k_1 \varepsilon \qquad (7)$$

s_1 is the initial mean free path perhaps related to the grain size and s_o the final value, lower than s_1. This decrease in s with strain leads to an almost linear strain hardening at low strains. The parameter k_1, as well as all the other parameters in the Bergström model, have been determined in uniaxial tensile tests In the present context we want to use ρ as the microstructural parameter and it is thus more appropriate, and also physically reasonable, to make s dependent on ρ rather than on ε. We will thus use

$$s = s_o + (s_1 - s_o)\exp - k_\rho \rho \quad (8)$$

where k_ρ has been fitted so that (8) gives the same strain dependence as (7) in the initial stages of a uniaxial tensile test.

In the temperature range of interest for the modelling creep of the waste canister copper it was assumed in the initial development of the model that the self diffusion was dominated by transport through the dislocation cores. The effective self diffusion coefficient D_{eff} can be written [3]

$$D_{eff} = D_{sv} + \rho a_c D_{sc} \qquad (9)$$

Values for the volume and core self diffusion coefficients D_{sv} and a_cD_{cv} can be found in ref. 5. With the use of these values it can be shown that core diffusion will dominate over volume diffusion at temperatures below 300°C for dislocation densities above those typical of recrystallized and undeformed copper. Therefore the model was limited to core diffusion. As a consequecne of that the thermal recovery term in (5) is proportional to ρ^3.

As a reasonable approach known values of the different parameters were used as far as they were available. In a few cases however these known values may be typical of a particular batch of material rather than being typical of the material. It was on the basis of such considerations that the following list of parameters was considered fixed:

Burgers vector	$b = 2.5 \cdot 10^{-10}$ m
Taylor factor	$\bar{m} = 3.06$
Vibration frequency	$\nu_o = 10^{12}$ s^{-1}
Shear modulus	$G(T) = 42\,100\,[1 - 0.54\,(T - 300)/1356]$ (MPa)
Self diffusion coefficient	$D_s = 1.0 \cdot 10^{-24} \cdot \rho \exp(-14072/T)$

Mean free path parameter $k_\rho = 2.3 \cdot 10^{-15}$ m^2
Final mean free path $s_o = 4.5 \cdot 10^{-6}$ m

All other parameters were used for fitting the equation to data.

As an initial step a stress-strain curve obtained at room temperature was used for fitting. In order to obtain the stress-strain curve in the model, the following equation for the tension test is added to the system of differential ekvations (1) and (5):

$$\dot{\varepsilon}_o = \dot{\varepsilon} + \frac{\dot{\sigma}}{E'} \tag{10}$$

In (10) E' is an apparent elastic modulus which in addition to the elasticity of the specimen includes all other elastic contributions to deformation. No less than six parameters were adjusted in order to fit the model result to the experimental curve. The following values were found to give a very good fit of the model to the stress-strain curve:

$A = 7.0 \times 10^{-15}$ m^4
$\Omega = 0.25$ m^2
$\sigma_o = 20$ Mpa
$\alpha_m = 1.05$
$s_1 = 2.8 \times 10^{-5}$ m
$\rho_o = 2.6 \times 10^{12}$ m^{-2}

ρ_o is of course not strictly a parameter but rather the initial value of the dislocation density ρ. It is obvious that with this large number of adjustable parameters there might be other combinations of parameters which give as good a fit to the experiment as those chosen to obtain the good fit shown in Figure 1.

In a separate calculation a test was simulated in which the extension rate was changed by a factor of 10 for every 2% of strain. As expected the resulting changes in stress increased with strain, a manifestation of the Cottrell-Stokes law. The model used automatically results in Cottrell-Stokes law being followed. The magnitude of the changes agreed well with experimentally determined changes.

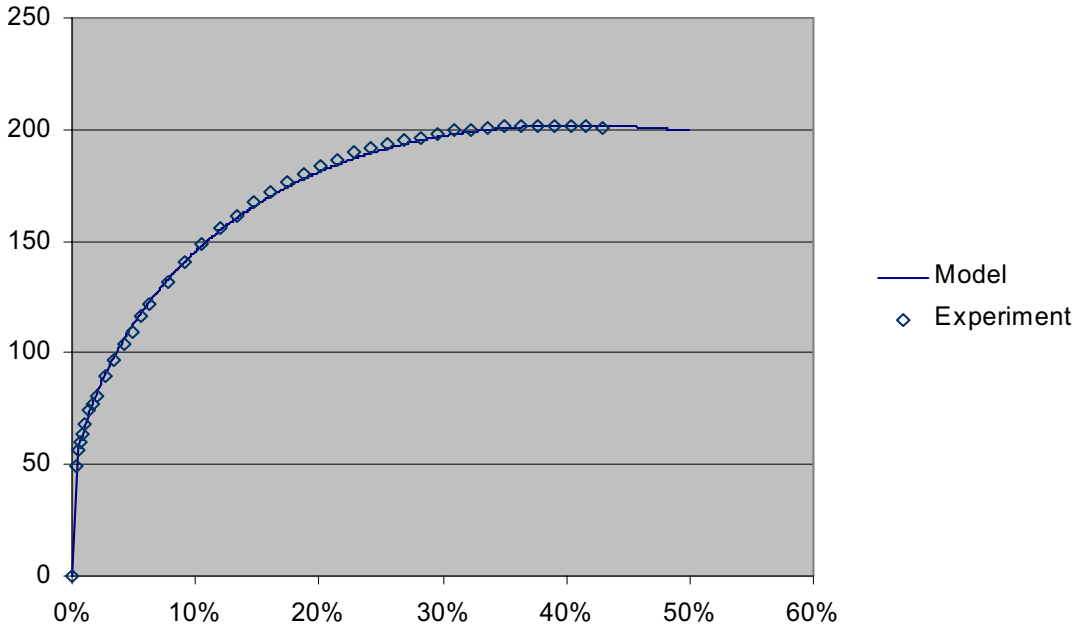

Figure 1. Stress-strain curve for pure copper compared with calculated behaviour.

After fitting the six parameters with the tensile test it only remained to fit the parameter M. For this purpose creep results reported by Lindblom et. al. [17] were used. A test performed at 300 °C at 60 MPa was used. With M = 22 the following result was obtained:

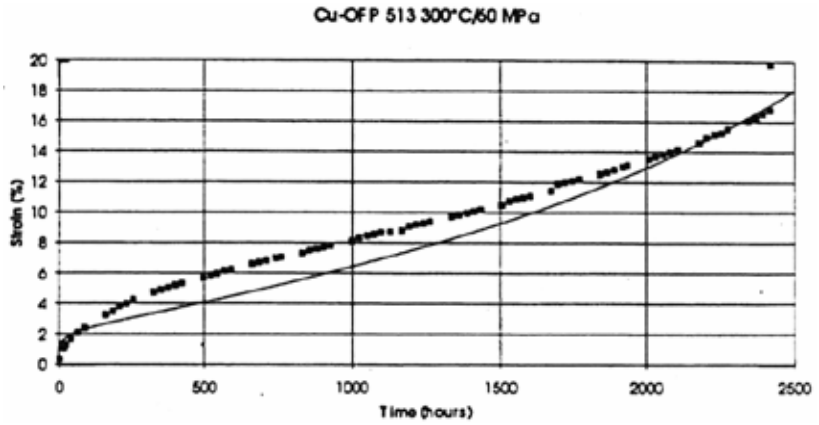

Figure 2. Model calculation compared to creep result from ref. 6. M was adjusted so that the same minimum creep rate as in the test was obtained.

With this value of M the creep deformation at other temperatures and stresses reported in [17] was calculated. The agreement between calculated and reported minimum creep rates was in general quite good. Also calculations of the Norton's law stress exponent agreed well with the experimentally determined values. The main problem with the

model seemed to be that even if the minimum creep rates were predicted well by the model the shape of the creep curves deviated significantly from the experimental curves as can be seen in Figure 2. Another interesting results of the calculations was that they confirmed the expectation that core diffusion would dominate over volume diffusion at the temperatures of interest for copper in the waste canister application.

2.2. Problems with the first version of the model.

One interesting aspect of the creep of copper, as reviewed for instance by Bowyer [18], is the large effect of small amounts of phosphorus on the creep properties. Even though the parameters of the model were developed with data on copper with an about 50 ppm addition of phosphorus it could equally well be used on pure copper with slightly modified parameters. One may ask however what the effect of phosphorus might be. It is clearly not a case of classical solution hardening since it is relatively easy to show that any reasonable solution hardening effect of phosphorus will be insignificant at a concentration of 50 ppm. It has however been suggested that phosphorus might have an effect on the recovery through some interaction with vacancies in the lattice and thus slowing down diffusion. This suggestion also pointed to a shortcoming of the first model since it entirely overlooked the production of vacancies during plastic deformation. In fact vacancies can be expected to be generated in large numbers in the type of deformation on which the model has been based, cutting of forest dislocations. In some of those cuttings jogs will be formed on the moving dislocations, jogs, which when they are dragged along with the gliding dislocation, by necessity will produce vacancies. These vacancies will make volume diffusion relatively much more important at low temperatures as will be discussed subsequently.

It has also recently become clear that the creep data against which the model was calibrated and tested had been misinterpreted. Quite frequently in creep testing, the immediate strain when the load is applied, is not included in the creep curve. This was also the assumption when the results in Figure 2 were tested against the model. Thus in the model calculations, the strain obtained when the loading phase of the calculation was over, was subtracted from the strain calculated for later times. However on careful reading of [17] it becomes clear that it is in fact the total strain which has been plotted. This would however not affect the minimum creep rate calculated so that comparison is not affected.

A more serious problem with the data in [17] is revealed by the Progress Report [19]. It suggests that the 5 mm diameter creep specimens used may have had a cold worked surface layer which has served to increase the strength of the specimens. This is also illustrated by the room temperature yield strength of 92 MPa of the creep specimens

compared to 61-70 for the tensile specimens. In subsequent calculations this will be taken into account by assuming that the creep specimens start with an initially higher dislocation density than the material used in the tensile test. This adjustment is done so that the creep specimens get a room temperature yield strength of 92 MPa in a model calculation.

2.3. How to take vacancy production into account.

The effect of vacancy production has been discussed by Mecking and Estrin [20]. They assume that the vacancy formation energy is αGb^3 where α is a constant and that a certain fraction χ of the plastic deformation energy is used for vacancies. With that assumption the production rate of vacancies is (in atom fraction):

$$\frac{dc_v}{dt} = \frac{\chi\sigma\dot{\varepsilon}}{\alpha G b^3} \times b^3 \qquad (11)$$

The authors assert that 0.1 is a reasonable estimate for χ/α. When the concentration of vacancies increases losses by diffusion to dislocations become significant. A slight modification of the expression proposed by Mecking and Estrin leads to the following expression for vacancy loss:

$$\frac{dc_v}{dt} = -D_v \rho \left(c_v - c_{vo}\right) \qquad (12)$$

where c_{vo} is the equilibrium vacancy concentration and D_v the vacancy diffusion coefficient. The vacancy diffusion coefficient is closely related to the volume self diffusion coefficient in the following way:

$$\begin{aligned} D_s &= D_{so} \exp - \frac{Q_s}{RT} = D_{so} \exp - \frac{Q_v + Q_m}{RT} \\ D_s &= D'_{so} c_v \exp - \frac{Q_m}{RT} = c_v D_v \end{aligned} \qquad (13)$$

Normally c_v has the equilibrium value approximately equal to $\exp(-Q_v/RT)$ but when there is an artificially introduced concentration of vacancies the factor $\exp(-Q_v/RT)$ must be replaced by the actual concentration c_v. As can be seen in eq. 13 this means that the activation energy for volume diffusion is reduced from $Q_v + Q_m$ to just Q_m, the migration energy for vacancies.

Instead of the expression (11) for the generation of vacancies it is possible to derive a more mechanistically based expression from the number of expected dislocation

cuttings as a function of strain rate and current dislocation density. An example of such an expression is

$$\frac{dc_v}{dt} = \overline{m} b \dot{\varepsilon} \sqrt{\rho} \qquad (14)$$

Since it seems reasonable that the vacancy production rate should be dependent on dislocation density eq. 14 has been used in preference to eq. 11 in subsequent calculations. Thus the following differential equation has been added to the previous system of differential equations for calculating creep or stress-strain curves:

$$\frac{dc_v}{dt} = \overline{m} b \dot{\varepsilon} \sqrt{\rho} - D_v \rho \left(c_v - c_{vo} \right) \qquad (15)$$

c_v has been added to ρ as a new state variable which characterizes the microstructure of the material.

2.4 A reoptimization of parameters for the tensile test.

In a study of the stress dependence of strain rate at different temperatures it became clear that the value of σ_o = 20 MPa is somewhat unphysical since no creep would occur below that stress. One way of dealing with that is to make σ_o temperature dependent or to reduce it in magnitude. The information available for determination of σ_o was in addition to the stress-strain curve in Figure 1 data of the yield strength at different temperatures given in the report by Lindblom et. al. In the latter case it was assumed that the material had a higher initial dislocation density than the material resulting in the stress-strain curve of Figure 1. By applying different parameter combinations and different alternatives for σ_o it was found that the most reasonable alternative was a temperature independent σ_o with a value of 10 MPa. With that value the following set of parameters gave the good fit to the tensile data shown in Figure 3:

$A = 2.1 \times 10^{-14}$ m^4
$\Omega = 0.9$ m^2
$\sigma_o = 10$ Mpa
$s_o = 2.5 \times 10^{-6}$ m
$s_1 = 2.8 \times 10^{-5}$ m
$\rho_o = 5.0 \times 10^{12}$ m^{-2}
$k_\rho = 6.0 \times 10^{-15}$ m^2

It may be noted that some of the parameters previously considered fixed are now changed.

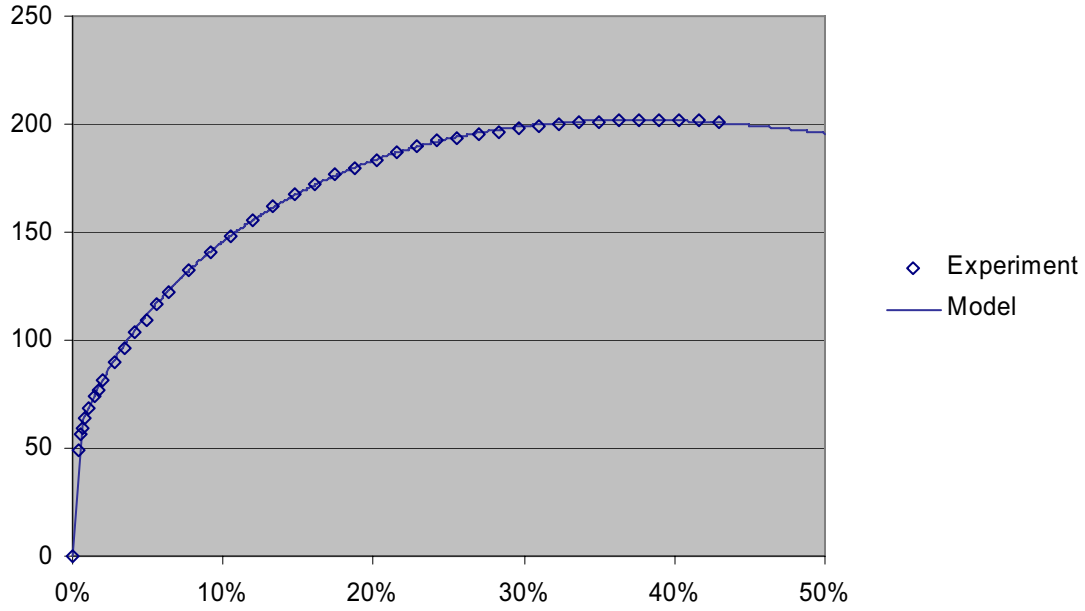

Figure 3. Simulation of the stress-strain curve with the new set of parameters.

2.5. Simulation of creep behaviour.

The first choice to be made is to divide the activation energy of copper self diffusion into a formation energy and a migration energy. The activation energy Q_v for volume self diffusion is given as 197 kJ/mol in [13]. One literature value of the vacancy formation energy is 0.9 eV which translates to 86.4 kJ/mol. Thus the vacancy migration energy should be 110.6 kJ/mol or 13300xR. These values are probably subject to considerable uncertainty. It is certainly possible to find in the literature other values which deviate with up to 10 % from those chosen here. The equations used for vacancy migration and vacancy formation thus were:

$$D_v = 2\times 10^{-4}\exp\left(-\frac{13300}{T}\right)\ \mathrm{m}^2/s$$
$$c_{vo} = 0.1\times\exp\left(-\frac{10392}{T}\right) \qquad (16)$$

In order to get the right magnitude of the yield strength of the creep specimens used by Lindblom et. al. a initial dislocation density of $\rho_o = 1.38\text{x}10^{13}$ m^{-2} had to be assumed. With that value the model was fitted to the creep data at 300 °C by varyinmg the parameter M. A good fit was obtained with M = 6.6 as can be seen in Figure 4.

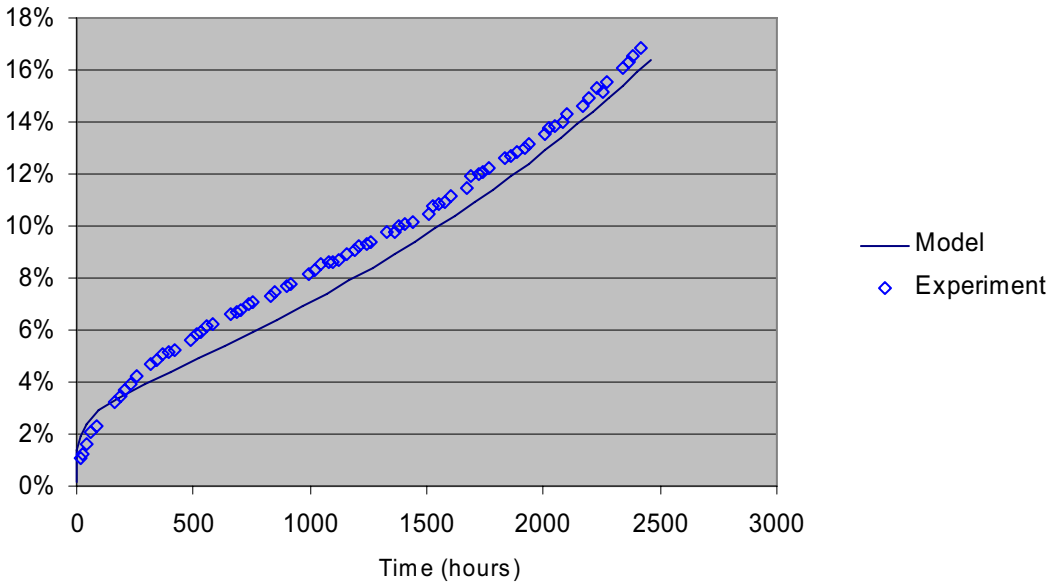

Figure 4. Model fitted to creep data at 60 MPa, 300 °C.

With this value of M creepcurves were calculated for 215 °C, 100 MPa and 200 °C 120 MPa. In both cases the model underpredicted the minimum creep rates at the lower temperatures by a factor of 2 at 215 °C and a factor of 1.65 at 200 °C. An analysis of how the different diffusion components play a role for recovery showed that vacancy production only plays an important role at low strains. At higher strains core diffusion is typically a factor of 10 higher than volume diffusion despite the fact that the vacancy concentration is significantly increased compared to the expected equilibrium vacancy concentration at these temperatures. The dominance of core diffusion indicated that the best way to improve the fit would be to change the activation energy for core diffusion. Whether or not that is a reasonble thing to do depends on the reliability of the published value of core diffusion. The value used here was derived by Frost and Ashby to match the activation energy for low-temperature creep found by Barret and Sherby in 1964 [13]. No actual measurements the activation energy had been done at the time when the book by Frost and Ashby was written. Therefore it does not seem unreasonble to play a little with this value in order to improve the fit between model and experiment.

When the activation energy for core diffusion was reduced from 117 kJ/mol to 98.9 kJ/mol an excellent fit to the data was obtained at all three temperatures. The change in the activation energy was compensated by decreasing the pre-exponential constant $\alpha_c D_{oc}$ so that the core diffusion remained unchanged at 300 °C. The resulting creep curves at 215 and 200 °C are shown in Figures 5 and 6.

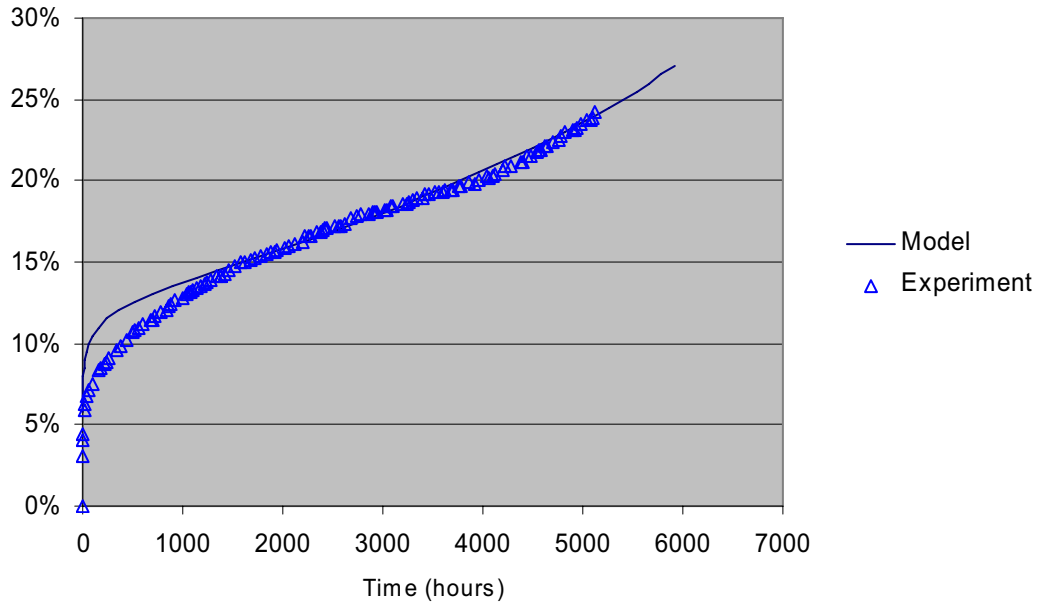

Figure 5. Model and experiment at 215 °C, 100 MPa.

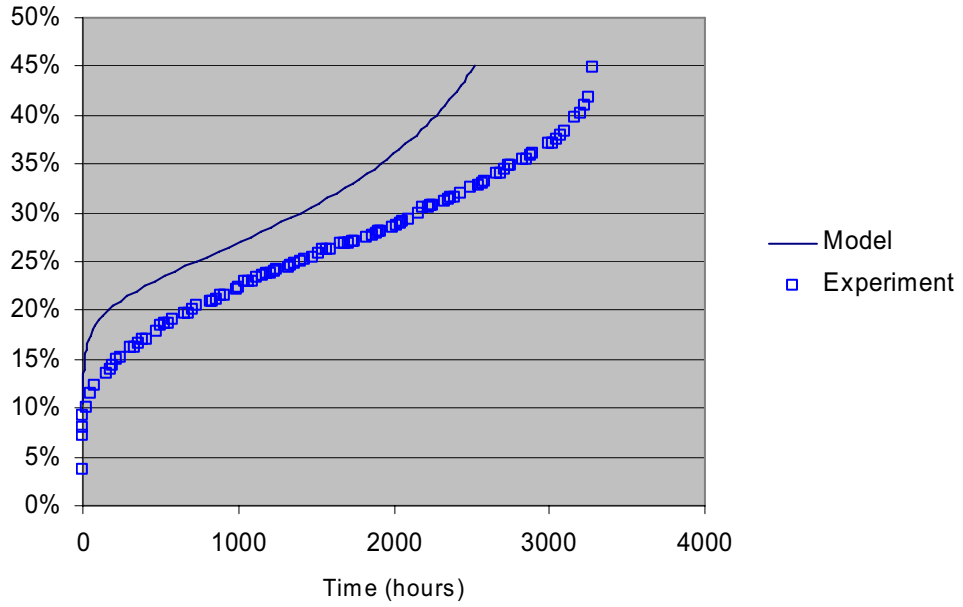

Figure 6. Model and experiment at 200 °C, 120 MPa.

The fit at 200 °C is good with regard to minimum creep rate. However the strain at a given time differs with about 5 %. One possible explanation to this could be that the previously mentioned cold work introduced when the specimens were fabricated might differ between specimens so that this particular specimen was a bit less cold worked than the others.

A comparison was also made with a fourth creep curve. This curve is presented in the report by Holmström et. al.[10]. The material is in this case taken from an extruded prototype canister. It apparently had a significant degree of cold work since in order to get the same strain on loading in the model calculations as in the test, an initial dislocation density of 2.1×10^{13} m^{-2} had to be assumed. But even with this assumption there was a significant deviation between model and test data as can be seen in Figure 7.

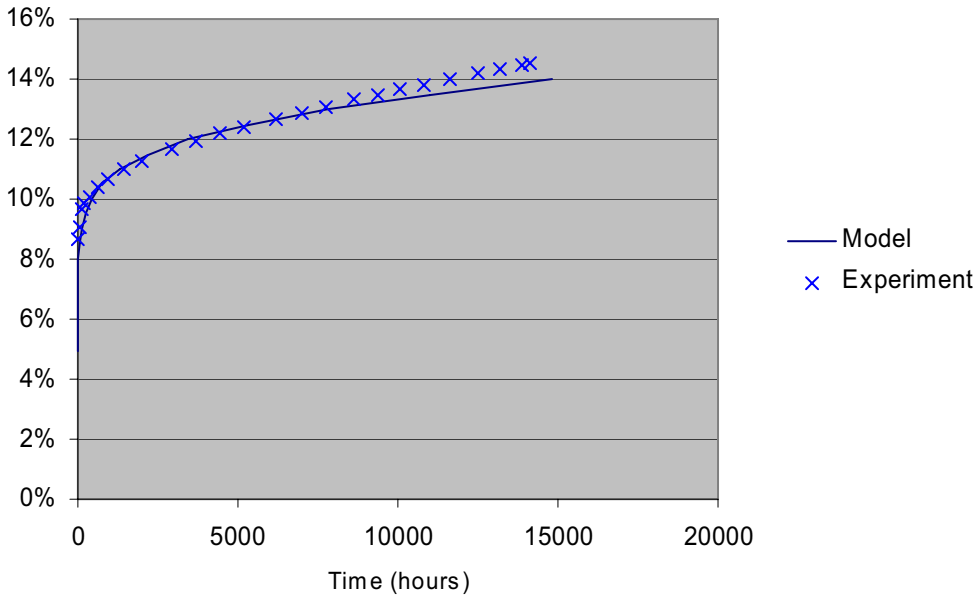

Figure 7. Comparison between creep test results and model calculation for a material from a prototype canister. Note that the strain is plotted as true strain in this figure. In all other figures engineering strain has been used.

There seems to be a significant underprediction of the creep rate. Instead of pointing to a fundamental flaw of the creep model this result may point to a complication that may have to be taken into account for a more accurate modelling of copper deformation behaviour. The point is illustrated by previous experience from work on stress corrosion cracking [21] on the same copper as was used in the tensile test of Figures 1 and 3. One of the problems in performing crack growth tests on this copper using fracture mechanics specimens was that the copper was too soft. Therefore the stress corrosion crack growth tests were performed with specimens made from material pre-deformed to 20 % true strain. The deformation was done by compression in the thickness direction of the initially hot rolled plate. In principle the stress-strain curve after predeformation should follow the part of the stress-strain curve of Figure 1 after 20 % deformation. However as evidenced by the stress-strain curves recorded during slow strain rate stress corrosion cracking tests the mechanical properties of the compressed material were quite different as can be seen in Figure 8.

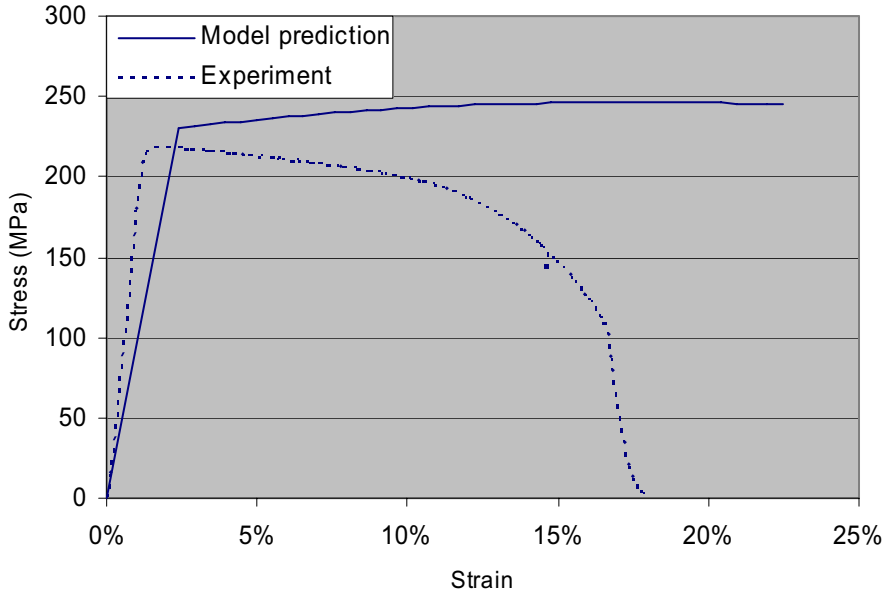

Figure 8. Comparison between the stress-strain curve of pre-compressed copper and model calculation of copper pre-deformed to the same true strain.

There are two explanations to the differences between the two stress-strain curves which are not mutually exclusive. One explanation is that the material is anisotropic to start with. The other explanation is that the strain hardening is anisotropic and also possibly that the cold work results in anisotropy. It may be noted in Figure 8 that the yield strengths are reasonably similar, but the strain hardening behaviours are quite different. The magnitude and importance of these effects should be explored before we put to much confidence into stress and strain calculations for the waste canister.

3. A simple application example.

One of the points of using a model of the type treated in the present report is that it handles situations of changing stresses better and more realistically than the traditional Bailey-Norton relationships complemented by strain hardening or time hardening for treatment of primary creep. In a simple application example it will be shown how the results of the present model compares with results with a time hardening model used in [10]. In the example the bending of a beam with a rectangular cross section by a pure bending moment will be simulated and the strain in the outer fiber will be plotted as a function of time. During the creep time the bending moment will be changed.

The time hardening law can be written:

$$\dot{\varepsilon} = \dot{\varepsilon}_p + \dot{\varepsilon}_s = Bk\sigma^k t^{x-1} + A\sigma^n \qquad (17)$$

where subscripts p and s denote primary and secondary creep rate respectively. The Norton exponent n is chosen as 5 in [10] and for simplicity the same value has been chosen for k. A is given as 5.03×10^{-17} at 150 °C when the units for time and stress are hours and MPa. A best fit to the experimental curve in Figure 6 was obtained with x =0.3 and B = 9×10^{-14}. The advantage with having the same stress dependence for the primary and secondary creep rate is that the stress distribution remains independent of time until the bending moment is changed. One problem with the time hardening law in comparison with the model is that it can not take into account the loading strain and the time independent strain increment when the applied load changes. On that basis the comparison to be performed is rather meaningless but it will be pursued nevertheless.

The beam modeled had a cross section of 0.05x0.05 m^2. The initial bending moment was 3410 Nm which was increased to 4000 Nm after 7500 h. The initial bending moment was chosen so that the stress in the outer fiber would be close to 120 MPa when a steady state stress distribution had been established. In the calculation of the creep strain first a preliminary strain is calculated in the outer fiber based on the current stress. Since the strain varies linearly over the cross section a preliminary strain can be calculated at all other points. In the next step the stress needed to achieve this preliminary strain is calculated. Based on the stress the bending moment over the cross section can be calculated. If this moment does not agree with the applied moment the stress in the outer fiber is adjusted and the calculations are repeated until the calculated bending moment agrees with the applied bending moment. In the case of the time hardening law this iterative process was not really needed since as mentioned above the stress distribution remains constant with time. With the multi-equation creep model a few iterations were needed but basically after a while the stress distribution was more or less constant. It should also be mentioned that a special version of the creep model was used in which the activation energy for core diffusion had been reduced to 87.3 kJ/mol in order to get comparable creep rates to the time hardening model at 150 °C. The result of the calculation of creep of the beam is shown in Figure 8.

As can be seen in the Figure the big difference between the models is the strains obtained when the load is applied or changed. In principle it would be possible to use the loading strain to estimate what happens at the start of loading. It would also be possible to handle different initial loads by just assuming that the strain on loading is the strain given by the stress-strain curve from a tensile test at that particular stress. It is more difficult to handle changes after that some creep has taken place. Then it is no longer possible to use the stress-strain curve

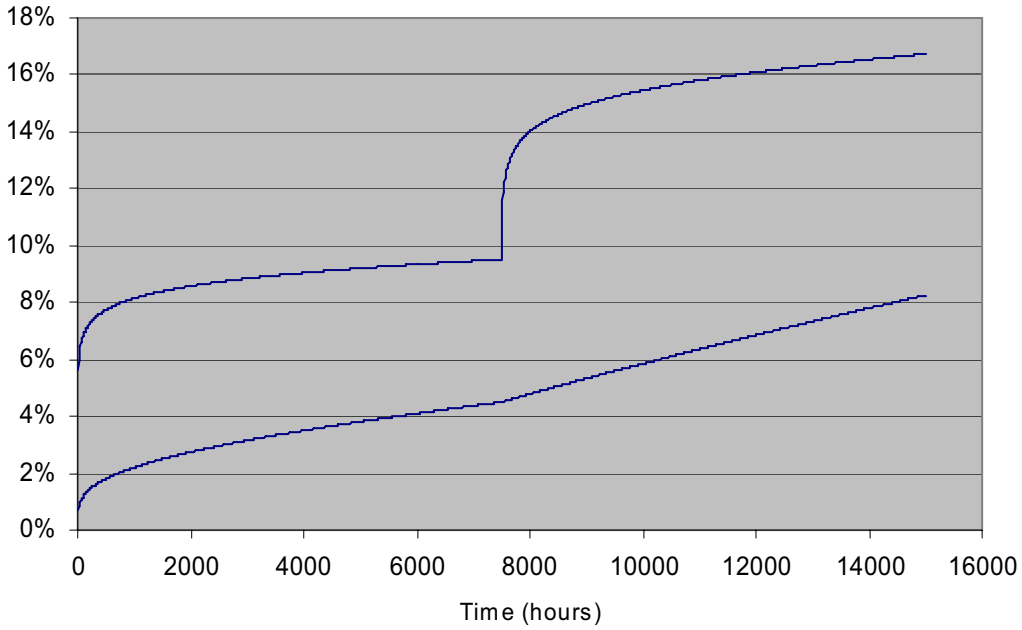

Figure 8. Comparison between results of the two models applied to creep of a beam under a constant bending moment. The lower curve is calculated with the time hardening model.

since the state of the material is different from what it is in the tensile test at that particular strain since some recovery must have occurred. One could of course try to estimate that recovery from the previous deformation history, a procedure which would have to be performed for every point in the material. Such type of work would however in principle be equivalent to working with an equation of state model of the type presented in the present report.

4. Concluding discussion

One of the goals of the present work was to modify the model so that the effect of phosphorus on creep rate could be modeled. The idea was that the small amounts of phosphorus present in the material would have an effect on the recovery of the dislocation structure by interacting with vacancies so that volume diffusion would be slowed down. In the first version of the model dislocation core diffusion was the dominant mechanism of recovery. However that model overlooked the generation of vacancies by plastic deformation and with introduction of vacancy generation into the model it was thought that volume diffusion would make a more significant contribution to recovery and thus also opening a possibility for phosphorus to play a role through its possible effect on vacancy mobility.

However in the calculations made it has been shown that it is only at low strains that volume diffusion dominates over core diffusion and that the minimum creep rate is

obtained in a strain range where core diffusion dominates. Thus it seems that the idea that phosphorus plays a role through its effect on diffusion and thus recovery can be rejected. Other mechanisms by which phosphorus plays a role should be explored. One idea is that phosphorus retards grain boundary sliding. If that is the case the stress concentrations in the grain due to grain boundary sliding will be reduced and these stress concentration will give a smaller contribution to the creep rate of the grains.

Another interesting result from the present work comes from the inability of the model to predict creep rate in a test at 150 °C of a material from an extruded canister. As a possible explanation to this effect has been suggested that worked material is anisotropic and that this anisotropy has to be taken into account when the plastic deformation and creep of copper is modeled. As an example a stress-strain curve of hot rolled copper was shown which had been pre-deformed 20 % in compression. If strain hardening is isotropic and no anisotropy is introduced by the plastic deformation this stress-strain curve should have the appearance of the stress-strain curve after 20 % strain for a material tension tested without any pre-deformation. However that was clearly not the case, which means that some type of anistropic behaviour is involved in the deformation of copper. That must clearly be taken into account in future modelling of copper behaviour.

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STATENS KÄRNKRAFTINSPEKTION
Swedish Nuclear Power Inspectorate

POST/POSTAL ADDRESS SE-106 58 Stockholm
BESÖK/OFFICE Klarabergsviadukten 90
TELEFON/TELEPHONE +46 (0)8 698 84 00
TELEFAX +46 (0)8 661 90 86
E-POST/E-MAIL ski@ski.se
WEBBPLATS/WEB SITE www.ski.se