Fundamental Models and Testing of Creep in Copper

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Academic thesis, which with the approval of KTH Royal Institute of Technology, will be presented for public review in fulfilment of the requirements for a Doctorate of Engineering in Materials Science and Engineering. Public review: KTH Royal Institute of Technology, Brinellvägen 8, room Kollegiesalen, at 10:00 on 12th June 2018.
To my parents, husband
and my daughter, Vilja

献给妈妈爸爸

烨哲和橙
Abstract

Many sustainable technologies for energy production, for example, generation IV nuclear system, demand the use of materials operating at elevated temperatures for long duration of up to 60 years. Requirements that are even more stringent are found for creep exposed copper canisters for disposal of spent nuclear waste. The canisters should stay intact for thousands of years. Traditional design procedures that involve empirical extrapolation of creep data are no longer reliable for such extended times. Instead physically based material models have to be used.

The final stage of creep before rupture, tertiary creep has been handled with empirical methods with adjustable parameters in the past, which makes it difficult to safely identify the controlling mechanisms. A physically based model has been developed for copper taking the substructure, cavitation and necking into account.

To improve the understanding of the important contribution from particles to the creep strength an earlier formulated model has analyzed and further developed. The model has successfully been able to describe the temperature and stress dependence of precipitation hardening for copper-cobalt alloys, where this contribution totally dominates the creep strength.

Multiaxial stress states are crucial for practically all high temperature applications. Fundamental material models have been extended for such conditions. These models have been compared with strain and stress controlled tests for notched specimens that have been performed.

Keywords: Copper; Creep tests; Multiaxial stress state; Finite element method; Basic modelling; Tertiary creep; Precipitation hardening
Sammanfattning

Många system för att begränsa klimatpåverkan som till exempel kärnkraft av generation IV typ, kräver användning av material som arbetar vid förhöjda temperatur under lång tid på upp till 60 år. Ännu strängare krav finns för krypexponerade kopparbehållare för använt kärnavfall som ska vara intakta i tusentals år. Traditionella designprocedurer som involverar extrapolering av uppmätta krypdata med empiriska metoder är inte längre tillförlitliga för sådana tidsperioder. I stället måste fysikaliskt baserade materialmodeller användas.

Det sista skedet av krypning före brott, tertiärkryp har tidigare hanterats med empiriska metoder med justerbara parametrar, vilket gör det svårt att säkert identifiera de styrande mekanismerna. En fysikaliskt baserad modell har utvecklats för koppar som tar hänsyn till substrukturen, kavitation och midjebildning.

För att förbättra förståelsen för det viktiga bidraget från partiklar till kryphållfastheten har en tidigare framtagen modell analyserats och vidareutvecklats. Modellen har framgångsrikt kunnat beskriva temperatur- och spänningsberoendet hos utskiljningshärdningen för koppar-koboltlegeringar, där detta bidrag dominerar kryphållfastheten helt.


Nyckelord: Koppar; Krypprov; Fleraxliga spänningsstillstånd; Finite element metod; Grundläggande modellering; Tertiärkryp; Partikelhärdning
Appended papers and the author’s contribution

**Paper I**
Slow Strain Rate Tensile Tests on Notched Specimens of Copper.  
**Fangfei Sui** and Rolf Sandström  
Contribution: literature survey, planning and performing the experiments, major part of modelling, data analysis and major part of the writing.

**Paper II**
Creep Tests on Notched Specimens of Copper.  
**Fangfei Sui,** Rolf Sandström and Rui Wu  
Submitted for publication  
Contribution: literature survey, major part of modelling, data analysis, major part of the writing.

**Paper III**
Basic Modelling of Tertiary Creep of Copper.  
**Fangfei Sui** and Rolf Sandström  
Contribution: literature survey, major part of modelling and data analysis, major part of the writing.
**Paper IV**

Fundamental Modelling of Mechanisms Contributing to Tertiary Creep in Copper at 215 and 250 °C.

**Fangfei Sui** and Rolf Sandström

Accepted by ASME PVP Pressure Vessels & Piping Conference 2018

Contribution: literature survey, major part of the modelling and data analysis, major part of the writing.

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**Paper V**

Creep Strength Contribution due to Precipitation Hardening in Copper Cobalt Alloys.

**Fangfei Sui** and Rolf Sandström

Submitted for publication

Contribution: literature survey, collecting and analyzing experimental data, part of modelling and major part of writing.
Conference contributions

Part of the work has been presented at the following conferences:


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With four parameters I can fit an elephant, and with five I can make him wiggle his trunk (John von Neumann, “father of the computer”)

1 Introduction

1.1 Creep

Creep is a time dependent slow plastic deformation that occurs when a material is subjected to a mechanical stress even when it is constant. Theoretically, creep can happen at all temperatures [1]. In metals, it takes place primarily at elevated temperatures above 0.4 \( T_m \), where \( T_m \) is the absolute melting temperature. During creep strain tests, the change of strain is recorded as a function of time. Figure 1 shows a typical creep curve for constant stress conditions. The initial strain represents the elastic strain plus the initial plastic strain that take place on loading. Three regions of the creep curve can be distinguished. In the primary creep region, the creep rate decreases with time. During secondary creep region, the creep rate stays constant at a minimum value over a range of strain, which is also denoted as steady state or stationary creep. In tertiary creep, microstructural changes take place that are referred to as creep damage, increasing the creep rate and eventually leading to rupture.
INTRODUCTION

Figure 1 Typical creep curve [2]

Creep deformation in polycrystalline metals takes place as a result of motion of dislocations, although in special cases at very high temperatures, it can be controlled by diffusion. So the appearance of a common creep curve can be explained with the help of dislocation mechanisms. In primary creep, generation of dislocation by work hardening dominates the deformation. With increasing strain, the dislocation density is raised and dislocations start to combine and annihilate each other, which is referred to as recovery. Eventually, balance between work hardening and recovery is obtained and then the steady state creep is reached. The acceleration of the creep rate in tertiary creep is due to a number of influencing factors, including localized necking, formation of creep cavities and acceleration of recovery.

In many applications under elevated temperature, the components lifetimes are limited by creep properties of materials. This concern has given rise to a large amount of work on developing alloys with superior creep resistance. In addition, modelling work on predicting creep life in terms of both creep deformation and creep rupture is vital.
1.2 Fundamental models

In the past, modelling mechanical properties have been generally described with expressions that have been fitted to the test data. Such models are here referred to as empirical. Empirical models have been widely used to analyze and extrapolate creep experimental data. Given the fact that most empirical models contain two or more adjustable parameters, they can usually describe the experiments accurately. However, the empirical models do not automatically provide any basic understanding of the studied phenomena. In addition, they are difficult to extrapolate to new test conditions, because the fitted parameters are then not automatically valid any longer.

In the present thesis, the focus will be on fundamental modelling of creep properties. What is meant here with fundamental modelling will be clearly defined, since it might be interpreted differently in different contexts. The following criteria are assumed to be satisfied [3].

- The models should be derived from fundamental physical knowledge about the controlling mechanisms.

- All parameters in the models should be well defined. It should be known how the parameter values could be derived.

- Introduction of “hand waving” and “fudge” parameters should be avoided.

- No fitting of parameters to experimental mechanical property data should be involved.

- The results in the form of analytical expressions should be sufficiently precise to be possible to use technically.

In the context of the present thesis, fundamental models are
the same as basic models and the terms are used interchangeably.

The significance of the absence of adjustable parameters should be noticed. It is common that models are partially or fully derived based on physical principles. If a few adjustable parameters are involved, many models can often give an apparently accurate representation of test data independently of their physical background. It is then very difficult to judge whether a specific model represents the controlling mechanisms. This has for example been illustrated for representations of creep curves [4].

For safe extrapolation, it is crucial to understand the complex time dependent chemical and mechanical degradation mechanisms and model the relevant damage mechanisms. The disadvantages of empirical methods can be avoided if models with physical basis are used. With the help of fundamental models, the underlying controlling mechanisms can be well understood. With precise representation of tests data, a safer extrapolation can be expected. The extrapolation by many orders in time can be feasible if fundamental models are used which is illustrated in [4]. In other words, the fundamental models are predictive for new conditions if the underlying assumptions are still valid.

A good example of a fundamental modelling approach is from Sauzay et al. [5] [6]. They have proposed models by taking the appropriate damage mechanisms into account for austenitic stainless steels. They assume that at high stresses, necking is the main damage mechanism and intergranular damage mechanism dominates the low stress condition. By combining the models based on necking for high stress and intergranular damage for low stress, good representation of results has been obtained.

It is quite a challenging task to formulate fundamental models according to the requirements specified above. Fortunately, in recent years a number of fundamental creep models have been
developed by the research group. The first basic step has been to establish a valid dislocation model that describes work hardening and recovery of dislocations [4, 7, 8]. With this model, expressions for the primary and secondary creep have been formulated as well as for stress strain curves [7, 9]. The significance of the substructure has been analyzed. It plays an important role for the effect of cold work on the creep rate as well as for a dislocation back stress that is built up [8, 10]. The contributions from slowly and fast diffusing elements to solid solution hardening have been determined [11-13]. A survey is given in Ref. [14]. Grain boundary sliding, nucleation and growth of cavities along grain boundary have been modelled [15-18]. By applying the models for cavitation, creep rupture and creep ductility have been studied. The fundamental models have been applied to copper, austenitic stainless steels and aluminum alloys [8, 19, 20]. Mechanical properties including creep rupture strength and creep elongation have been precisely represented. Some of these models will be presented in the thesis.

These models have given the possibility to quantitatively predict property values and compare the predictions with experiments. In addition, a number of phenomena have been possible to be explained that were poorly understood in the past. One such example is the influence of cold work on the creep strength. Previously, no quantitative model was available in the literature. By taking the role of substructure into account, it has for example been possible to understand why 24% cold work in copper increases the creep life by six orders of magnitude [10]. Another example is that many creep curves for cases where the creep exponent is high, have about the same appearance as if the creep exponent is about five. The explanation is that the rapidly increasing true stress with strain is balanced by a dislocation back stress from the substructure [8]. A third example is grain boundary sliding and cavity nucleation. Well based models are now available that can describe the nucleation in the substructure and around
particles. For example, the well-known linear relation between the number of nucleated cavities and the creep strain can be fully quantified [16, 21].

1.3 Copper canisters for disposal of spent nuclear fuel

Safe and long-term management of radioactive waste from nuclear technologies has been a crucial problem for the whole world. Extensive studies have been conducted over the last decades regarding this subject. A final agreement has been reached that underground geological disposal is the mostly preferred solution for high level waste and spent nuclear fuel [22]. Deep geological disposal would be located at places where the geological conditions are stable so that the waste can be isolated from humans and the environment until it is no longer hazardous. This method has been selected because geological disposal can provide a multiple barrier protection system from both local geology and engineered materials. Based on this principle, a final repository 500 meters underground has been proposed in Sweden, named the KBS-3 concept as shown in Figure 2. In this repository the spent nuclear fuel is planned to be enclosed in copper canisters with cast iron insert, surrounded by clay. The multiple barrier protection is from copper canisters, clay and bedrock [23].
Figure 2 Illustration of the KBS-3 concept

The research interest of the thesis is the copper material for the canisters. It is selected as the protective engineered materials due to its excellent corrosion resistance and high ductility. Each canister is five meters long with a diameter of one meter. It is composed of three parts, the lid, tube and base. The lid and the base are sealed to the tube by friction stir welding \[24\]. Sometimes the tube and base is made in one piece and then the welding of the base is not required. Figure 3 gives a sketch of the size of the copper canister and the bentonite clay compared with a human.
Figure 3 Sketch of the size of copper canister and bentonite clay

The service condition of copper canisters is believed to be complex. Firstly, during disposal the spent nuclear fuel will release heat while decaying, increasing the temperature in the repository up to a maximum of 100 °C. Since the canisters are buried 500 meters underground, there is hydrostatic pressure on the canisters. In addition, the clay will swell when it comes into contact with underground water. The swelled clay will also exert a pressure on the canister. The pressure will gradually increase until the clay is water saturated, after that the full pressure is reached.

There are critical points on the copper canister. At the connections between lid/base and tube there exist slits about 75 mm in length and 0.15-0.33 mm in width. Considering the large scale of canister and existence of tiny slits, large stress concentration will appear. The top section of the canister is exposed to horizontal and vertical loads that give rise to complex multiaxial stresses. The canisters are under raised temperature and complex stresses, which will result in creep. To simulate the service condition of copper canisters, it is essential to perform tests under multiaxial stress state. The period of pressure increasing can
be simulated by slow strain rate tensile (SSRT) tests. After the full pressure is reached, creep deformation commences and creep tests should be conducted for such a situation.

The repository is planned to be functional for thousands of years until the radioactive waste is no longer hazardous, making the designed lifetime of the copper canister to be 100000 years. To predict the damage during such a long period, it is critical that the deformation controlling mechanisms are fully understood. Usual empirical models are no longer valid for such a design life. Fundamental models based only on physical phenomena are needed.

1.4 Aim of the work

The main theme of the current thesis is to study the creep properties of copper through both experiments and modelling. Fundamental models will be developed and applied to gain insight of the deformation and damage mechanisms of copper canisters.

The first part of the thesis is to simulate the deformation of copper canister by performing SSRT tests and creep tests under multiaxial stress state. As a new approach, fundamental models are adapted and applied to multiaxial stress states. Experimental data is used to verify the validity of the fundamental models for interpreting both load and strain controlled tests under multiaxial stress states.

A wide range of empirical models for handling tertiary creep are available in the literature, but no fundamental models, where all the relevant mechanisms are taken into account in spite of its technical significance for creep damage and creep rupture. Basic models for both primary and secondary creep for copper have been available for a while. In the thesis, tertiary creep of copper is treated as well.
INTRODUCTION

There is an extensive literature on the influence of precipitation hardening on the creep strength. In most papers, a constant threshold stress is considered. This is at variance with experimental observations that show a pronounced temperature and stress dependence except for some oxide dispersion strengthened alloys. Using principles formulated previously in the research group, a model that avoids these limitations is formulated. The validity of the temperature and stress dependence of the precipitation hardening in the model is verified for published experimental data of Cu-Co alloys.
2 Prediction of long term data

As the environment pollution is getting more severe and world population is expanding, the demand for energy is growing. Considering the large production of energy and low carbon dioxide emissions, nuclear energy is needed as a prominent supply to satisfy the energy demand of mankind. It was been developed initially in the 1950s with the early prototype reactor, which is called first generation nuclear system. Figure 4 shows the generations of nuclear energy systems, with regard to different types of reactors and other features.

![Figure 4: Generations of nuclear power system](image)

There is no doubt that the material technology is a key factor to the success of new nuclear systems. Although different reactor systems are associated with different demands, high temperature components are always expected to meet the complicated service condition e.g. high temperature and harsh irradiation. Major factors to be concerned in selection of materials for structural components used in the reactor systems are as following [26-29]:

- High temperature mechanical properties: the components materials need to exhibit good properties under high temperature, such as high creep resistance, high ductility and
creep-fatigue resistance.

- Property stability under corrosion: the components materials properties, including tensile strength, fracture toughness, ductility, creep resistance, etc. are expected to be stable under irradiation, stress and corrosive atmosphere.

- Long-term stability: the components are designed to be used for at least 60 years, during which the properties should remain reliable.

These demands apply in practice to high temperature plants in general, for example for advanced fossil fired power plants and units for productions of hydro carbons.

In practice, to meet these requirements, existing reactor materials are needed to be optimized and new materials have to be developed. One of the most important requirements for newly developed materials is to improve the high temperature properties. One of the main challenges for high temperature material for new generation nuclear reactors is the demonstration of their lifetime for up to 60 years. In view of this fact, both long-term creep tests data and reliable models for extrapolation are needed.

### 2.1 Empirical models for creep rupture

In the past, design against creep when selecting component material for example for power plants was based on the allowable creep strength (the stress causing failure in $10^5$ h at the in service creep conditions) [30]. Since it is difficult to acquire a large number of long-term data, the allowable creep strength was usually estimated from shorter term data where the tests lasted for up to 30000 h. Extrapolation of short-term experimental curves (log creep rupture time versus log creep rupture stress) based on power law relationship was used to predict long-term creep lifetime. Various time temperature parameter methods have been
widely used, such as Larson-Miller and Manson-Haferd [31, 32]. The master curve equation is in the following form,

\[ P = b_0 + b_1 (\log \sigma) + b_2 (\log \sigma)^2 + \ldots + b_k (\log \sigma)^k + e_i \]  

(1)

where \( \sigma \) is the stress, \( b_0, b_1, b_2, b_k \) the regression coefficients, \( k \) the degree of regression equation and \( e_i \) the error term. The parameter \( P \) differs in the various methods.

For the Larson-Miller method [31], \( P \) is given by

\[ P_{LM} = T(C_{LM} + \log t_r) \]  

(2)

where \( T \) is the absolute temperature, \( t_r \) the time to rupture, and \( C_{LM} \) is a material constant. This method has been widely used due to its simplicity. However, it has been reported that the best fit \( C_{LM} \) value was often too large and overestimated the creep strength. To get a more conservative result, a value of 20 for \( C_{LM} \) is used for ordinary steels [33].

For Manson-Haferd method, the corresponding expression is [32]

\[ P_{MH} = (\log t_r - \log t_a) / (T - T_a) \]  

(3)

where \( \log t_a \) and \( T_a \) are constants that are fitted to the experimental data. Since it contains two adjustable parameters, it can describe creep rupture curves with strong curvature.

Wilshire [34] proposed an approach using the commonly adopted Larson-Miller methodology to predict the lifetime of high Cr steels. In essence, the ultimate tensile strength values and activation energy for matrix diffusion were introduced. By normalizing the applied stress in creep test through the ultimate tensile strength values at creep test temperatures for each heat, the multi-batch creep fracture data can be extrapolated. The stress and temperature dependence of time to rupture is given as
(σ / σ_{TS}) = \exp(-k(t_f \exp(-Q_c / RT))^u)

(4)

where $k$ and $u$ are constants and $Q_c$ the activation energy. The obtained allowable creep strength values at different temperatures were compared with available long-term data, emphasizing the use of shorter term test data of $t_r < 30000$ h.

Region splitting analysis method was proposed by Kimura et al. [35]. Because of the microstructure degradation at low stress long-term tests at high temperature, the creep strength would decrease dramatically. They proposed that short-term data should be excluded when parametric methods are used to predict long-term performance. Multi region analysis method was also proposed to analyze creep rupture data for some ferritic and austenitic steels [36]. Regions denoted as H, L and L₂ with different stress exponent $n$ and different activation energy $Q$ were introduced.

The methods mentioned above are used to predict the creep lifetime with creep fracture data. There are many other models representing creep versus time curves from simple phenomenological equations. To predict creep performance at new conditions, how the adjustable parameters vary with stress and temperatures are described. One of these equations is the Omega model [37, 38], which contains two parts, describing primary and tertiary creep respectively,

$\varepsilon = \ln(\sqrt[n]{\omega_1 \omega_2 t + 1} / \omega_2)$

(5)

$\varepsilon = -\ln(-\sqrt[n]{\omega_3 \omega_4 t + 1} / \omega_4)$

(6)

With the combination of eqs. (5) and (6), the whole creep curve can be represented. This method has been used to describe the creep curves and predict creep rupture of many creep resistant steels.

However, even if the best fitting procedure is chosen, the
achieved accuracy is not satisfying enough [39, 40]. For some materials like 9 to 12 Cr steels and austenitic stainless steels, creep rate shows different stress dependences at high and low stresses. When extrapolating the creep lifetime using these methods, it is likely to underestimate the creep rate and overestimate the allowable creep rupture strength. Large difference in lifetime as a factor of 10 has been found in some cases by comparing the extrapolated data with available long-term experimental results. In fact, the estimated allowable creep strength has been reduced progressively as new long-term data have been obtained. Considering the disadvantages of above-mentioned methods, it is not reliable to predict the long-term lifetime by using the simple traditional statistical extrapolations by more than a factor of three in time [5].

2.2 Empirical models for creep deformation

The creep lifetime is not only determined by creep rupture mechanisms but also by creep deformation. In metals and alloys, the stress and temperature dependence of the secondary creep rate is represented by combined power-law and Arrhenius equations originally proposed by Mukherjee et al. [41] in the form

$$
\dot{\varepsilon} = A \frac{D_{0sd}Gb}{k_BT} \left( \frac{\sigma}{G} \right)^n \exp\left( -\frac{Q_c}{RT} \right)
$$

(7)

where $\dot{\varepsilon}$ is the steady state creep rate, $A$ a constant, $n$ the stress exponent, $\sigma$ the applied stress, $G$ the shear modulus, $b$ the length of the Burgers vector, $k_B$ the Boltzmann constant and $R$ the gas constant. $Q_c$ is the creep activation energy, which is close to the activation energy for self-diffusion in pure metals, $Q_{sd}$. $D_{0sd}$ is the pre-exponential factor in the Arrhenius equation for diffusion.

The stress exponent $n$ can be determined experimentally from the gradient of $\log \dot{\varepsilon} / \log \sigma$ relationship at constant $T$. The $n$ value
usually lies in a range of 4 to 7 for pure metals and many alloys, conveniently termed as “five power law” [42]. In the past, it has been assumed that the stress exponent values reflect the operating dislocation mechanism. For intermediate stress exponents ranging from 4 to 7, the controlled mechanism is believed to be climb. Viscous glide controlled creep has been assumed to give a stress exponent around 3. When the creep exponent is approaching unity at low stresses, diffusion creep is assumed to become the main mechanism although there are limited data to support this assumption. At high stresses and usually associated with low temperature, power-law breakdown takes place with an increasing stress exponent. However, these common assumptions on whether the stress exponent gives a reliable prediction of the controlling mechanism have been questioned [14, 43]. In eq. (7), two parameters \( A \) and \( n \) are given as fitting parameters to the experimental data. In addition, it can only be used to describe power-law regime with a constant \( n \). For power-law breakdown creep, a different model is needed.

2.3 Summary of empirical models

In summary, all the above-mentioned models have a simple mathematical form and contain two or more adjustable parameters. These models have been widely used although a quantitative physical basis is often absent. However, the accuracy of extrapolation is limited when using these models [44]. The disadvantages of empirical models are particularly notable when it is used for material design for new generation nuclear reactor, where the design life is up to 60 years. To make an extrapolation for such a long period in a reasonably safe way, both accurate long duration tests data sets and suitable procedures are needed. Therefore, although there remains a significant challenge, the basic and quantitative modelling based on only physical phenomenon without fitting parameters is in great demand.
3 Materials and testing

3.1 Material and specimens

The canisters were planned to be made of oxygen free pure copper (CuOF). However, extensive experiments showed that the pure copper could have very low creep ductility [45]. By adding 50-60 ppm phosphorus, the creep ductility of copper increased dramatically [46, 47]. Since then the canisters have been made of oxygen free copper with 50 ppm phosphorus (Cu-OFP) although the effect mechanism of phosphorus is still under discussion [48]. The testing materials were cut from full-scale copper canister tube and lid provided by Svensk Kärnänslehantering AB (Swedish Nuclear Fuel and Waste Management Company, SKB). As-received material is shown in Figure 5.

To simulate the in-service condition of copper canisters, multiaxial stress state was considered. Practically, tests of large scale components are difficult and expensive to perform. A simplified way to introduce multiaxial stress state is to subject circumferentially notched bars to axial load [49, 50]. Various studies have been carried out with notched bars to examine the influence of multiaxial stress state [51-53].

Double notched Cu-OFP cylinder bars were used for both SSRT and creep tests. The specimens for both test sets had the same geometry with a gauge length of 47 mm, total length of 129 mm and three different notch geometries, see Figure 6. The notch acuity was defined as the notch root radius (a) divided by the notch throat radius (R). Different notch acuities represent different stress states. The only difference in these two sets specimens was that the knife edges were not used in the SSRT testing. Appearance of specimens for SSRT tests are given in Figure 7.
MATERIALS AND TESTING

Figure 5 Image of tube material as received from SKB

Figure 6 Specimen geometry used in SSRT and creep tests
3.2 Slow strain rate tensile tests

SSRT tests were strain rate controlled. In the current study, two different strain rates (10^{-6} and 10^{-7} s^{-1}) were tested at two temperatures (75 and 125 °C) for each notch acuity. In total, 16 SSRT tests were conducted and all tests went to rupture.

3.3 Creep tests

The tensile creep tests were performed in air under constant load at 75 °C. For each notch acuity, creep tests were conducted at different net section stresses. In total, 20 creep tests were conducted, most of which went to rupture. Some were interrupted due to unforeseen long test duration. Details of the creep tests are shown in Table 1.
Table 1 Details of the performed creep tests

<table>
<thead>
<tr>
<th>Notch acuity</th>
<th>Net section stresses (MPa)</th>
<th>Interrupted</th>
<th>Ruptured</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>170, 175, 180</td>
</tr>
<tr>
<td>0</td>
<td>170, 180</td>
<td>195, 200, 215</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>170, 180, 200</td>
<td>215, 225, 230</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>170, 180, 200, 215, 230</td>
<td>245</td>
<td></td>
</tr>
</tbody>
</table>

3.4 Measurements

3.4.1 Reduction in area

To determine the ductility of the specimens, area reduction of the ruptured notches was measured. Fracture images were taken by scanning electron microscopy (SEM, Hitachi TM3000, Japan) and the radii of the specimen were measured with the built-in measuring tools. The images and data were analyzed in Photoshop CS5. Since the fracture area was in varying contrast, it was automatically selected by the “selected tool” and then adjusted manually. The exact fracture area was highlighted, see Figure 8.

Figure 8 Illustration of the measured area reduction in SEM micrograph
3.4.2 Necking profile

High resolution photos were taken of the ruptured specimens at the necking position at a high magnification. The radii of the creep specimen at different positions were measured using a Kappa measuring microscope at Swerea Kimab. The photo of necking specimen and necking radii data were then loaded into Techdig digitizing software. To describe the necking profile accurately, data points were collected particularly at the position near rupture. The data was then fitted into curves and compared with a FEM modelled necking profile. Figure 9 shows the data points taken from the specimen in the Techdig software.

Figure 9 Illustration of determined necking profile by Techdig
4 Fundamental models

Analysis of experimental creep data has traditionally been made with empirical models involving a number of parameters that are fitted to experimental data. However, the empirical models do not always give insight of the controlling mechanisms. With such models, it can be difficult to extrapolate results to new test conditions or longer time. In recent years, basic models for prediction of creep properties have been proposed. They are based on physical mechanisms and do not include adjustable parameters.

By understanding the detailed controlling mechanism, a safer extrapolation to longer time can be expected. The basic models cover dislocation, particle and solid solution hardening, cavity nucleation and growth, ductile and brittle rupture.

4.1 Dislocation model

To describe the plastic deformation, the key quantity is the dislocation density and its variation with time. According to recovery creep theory, the work hardening gives rise to an increasing density of dislocations. At the same time, the recovery due to the annihilation of dislocations reduces the dislocation density. Two kinds of recovery, dynamic and static are considered. The distinction of these two recovery mechanisms is important for examples for describing tertiary creep which will be discussed in a later section. The terminology concerning recovery is not fully consistent in the literature. Here dynamic recovery is considered to be strain dependent while static recovery is time dependent [54]. Dynamic recovery will occur as long as straining takes place by the rearrangement of dislocation into lower energy configurations [55]. Static recovery describes how dislocations of opposite sign attract each other and eventually annihilate. Dynamic recovery also takes place when dislocations pass through cell boundaries and remove dislocation locks [56, 57].
To describe how the dislocation density develops during creep, the contributions from three processes are considered [4, 7].

\[
\frac{d \rho}{d \varepsilon} = \frac{m}{b c_L} \rho^{1/2} - \omega \rho - 2 \tau_L M \rho^2 / \dot{\varepsilon} 
\]  

(8)

A detailed derivation of eq. (8) and its constants can be found in [58]. The three terms on the right hand side of the equation represent work hardening, dynamic recovery and static recovery, respectively. In eq. (8), \( \rho \) is the dislocation density, \( \varepsilon \) the strain, \( m \) the Taylor factor, \( b \) Burger's vector, \( c_L \) a work hardening constant, and \( \omega \) the dynamic recovery rate constant. \( \tau_L \) is the dislocation line tension, \( \dot{\varepsilon} \) the strain rate. \( M \) is the creep mobility and is given by

\[
M(T, \sigma) = \frac{D_{so} b}{k_B T} e^{\frac{\sigma b^3}{k_B T}} e^{-\frac{Q}{RT} \left[ 1 - \left( \frac{\sigma}{\sigma_{\text{max}}} \right)^2 \right]}
\]  

(9)

where \( D_{so} \) the pre-exponential coefficient for self-diffusion, \( Q \) the activation energy for self-diffusion and \( \sigma_{\text{max}} \) the max back stress which is taken as the tensile strength at room temperature. The main part of eq. (9) is the climb mobility derived by Hirth and Lothe [59]. The final term in the square brackets was originally formulated to take glide into account [60]. Recently, it has been shown that it can be obtained if the influence of strain induced vacancies on the climb rate is considered [14, 19].

The climb mobility is an important quantity; it was first derived for CuOF. For Cu-OFP, the influence of phosphorus content should be taken into account [60].

\[
M_{\text{OFP}}(T, \sigma) = \int_0^1 \frac{D_{so} b}{k_B T} e^{\frac{(\sigma - \sigma_{\text{break}}) b^3}{k_B T}} e^{-\frac{Q}{RT} \left[ 1 - \left( \frac{\sigma - \sigma_{\text{break}}}{\sigma_{\text{max}}} \right)^2 \right]}
\]  

(10)

A minimum stress \( \sigma_{\text{break}} \) is required to make the dislocations break away from the atmospheres of phosphorus atoms (Cottrell atmospheres). Phosphorus also influences the activation energy.
This is taken into account with the factor $f_Q$

$$f_Q = e^{-U_p^{\text{max}} / RT} \quad (11)$$

$U_p^{\text{max}}$ is the maximum interaction energy between a phosphorus solute and a dislocation.

Eq. (8) is a general equation for the development of the dislocation density during plastic deformation in copper. Its validity has also been demonstrated for aluminum alloys [14, 61] and stainless steels [62]. It will be illustrated in the following sections that the dislocation models can be used to describe stress strain curves from constant strain rate tests as well as creep strain versus time curves from creep tests at constant load.

### 4.2 Stress strain curves

When computing stress strain curves, the static recovery can be neglected since the contribution is negligibly small. Eq. (8) is simplified as

$$\frac{d \rho}{d \varepsilon} = \frac{m}{bc_L} \rho^{1/2} - \omega \rho \quad (12)$$

The dislocations give rise to a stress that is given by Taylor’s equation

$$\sigma = \sigma_y + m\alpha_T Gb \sqrt{\rho} \quad (13)$$

where $\alpha_T$ is a constant, $G$ the shear modulus, and $\sigma_y$ the yield strength. Using eqs. (12) and (13), the differential equation for stress strain curves can be derived

$$\frac{d \sigma}{d \varepsilon} = \frac{\omega}{2} (K + \sigma_y - \sigma) \quad (14)$$
where

\[ K = \frac{m^2 \alpha_L G}{c_L \omega} \]  

(15)

Integrating eq. (14) gives

\[ \sigma = \sigma_y(T, \dot{\varepsilon}) + (\sigma_{\text{max}} - \sigma_y(T, \dot{\varepsilon}))(1 - e^{-\omega \varepsilon/2}) \]  

(16)

where \( \sigma_{\text{max}} = K + \sigma_y \) is the maximum stress in the flow curve. The stress strain curves can be modelled by eq. (16), which has the form of the Voce equation [63].

### 4.3 Primary and secondary creep

When computing creep curves, the dynamic recovery term will be temporarily ignored to avoid complex expressions. Eq. (8) can then be simplified to

\[ \frac{d \rho}{d \varepsilon} = \frac{m}{b c_L} \rho^{\frac{3}{2}} - 2 \tau L M \rho^2 / \dot{\varepsilon} \]  

(17)

During stationary conditions that characterize secondary creep with a constant strain rate, the strain derivative of the dislocation density is zero. Combining with Taylor’s equation, it gives an expression for the secondary strain rate as

\[ \dot{\varepsilon}_{\text{stat}} = \frac{2 b c_L \tau L M(T, \sigma)}{m} \left( \frac{\sigma}{\alpha_L m G b} \right)^3 \]  

(18)

Eq.(16) represents tests at constant strain rate and eq. (18) tests at constant load. Since the maximum stress in the flow curve is related to the stationary creep stress at the given strain rate, to derive an expression for creep curves during primary creep, the two equations are combined. By extracting \( \sigma_{\text{max}} \) from eq. (16) and inserting the expression into eq.(18), we find an equation that
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covers both primary and secondary creep.

\[
\frac{d\varepsilon}{dt} = h(\sigma_y(T, \dot{\varepsilon}) + \frac{\sigma - \sigma_y(T, \dot{\varepsilon})}{1 - e^{-\omega \varepsilon/2}}, T) \tag{19}
\]

Eq. (19) is used to represent primary and secondary creep.

4.4 Tertiary creep

Modelling of tertiary creep is different from the model of primary and secondary creep in the previous section. The dynamic recovery term cannot be ignored. In addition, the role of substructure should be taken into account. In copper and many other metallic materials, a cell structure is formed during deformation and most dislocations lie in the cell boundaries [54, 64]. In the modelling of tertiary creep, a distinction of balanced and unbalanced dislocation was made for the dislocations on cell walls [10, 65]. Balanced dislocations are characterized by the presence of dislocations with opposite Burgers vector on the same slip system. On the other hand for the unbalanced dislocations, dislocations with the opposite Burgers vector are not available. According to this distinction, the unbalanced dislocations are not subject to static recovery. Both unbalanced and balanced dislocations are subject to dynamic recovery. Their densities satisfy the following equations,

\[
\frac{d\rho_{\text{bnd}}}{d\varepsilon} = k_{\text{bnd}} \frac{m\rho_{\text{bnd}}^{1/2}}{b c_L} - \omega \rho_{\text{bnd}} - 2\tau_L M \rho_{\text{bnd}}^2 / \dot{\varepsilon} \tag{20}
\]

\[
\frac{d\rho_{\text{bnde}}}{d\varepsilon} = k_{\text{bnde}} \frac{m\rho_{\text{bnde}}^{1/2}}{b c_L} - \omega \rho_{\text{bnde}} \tag{21}
\]

where \(\rho_{\text{bnd}}\) and \(\rho_{\text{bnde}}\) are the balanced and unbalanced dislocation density in the cell walls. The cell structure configures low energy, where the dislocations give a lower contribution to the strength. Taking this into account, parameters \(k_{\text{bnd}}\) and \(k_{\text{bnde}}\) are introduced
to control the amount of balanced and unbalanced dislocations.

A back stress is introduced in the model as the extra hardening from the unbalanced dislocations in the cell walls [8]. This stress compensates for the sharp increase in true applied stress

$$\sigma_{appl} = \sigma_{appl0} e^\varepsilon$$
during secondary creep at high stress exponents, where $\sigma_{appl0}$ is the applied nominal stress. The magnitude of the back stress equals the dislocation stress minus the nominal applied stress where $\sigma_{disl}$ is given by

$$\sigma_{disl} = \frac{m\alpha T G b}{2} \sqrt{\rho_{bnd} + \rho_{bnde}}$$  \hspace{1cm} (22)

The effective creep stress is the true applied stress minus the accompanying back stress

$$\sigma_{creep} = \sigma_{appl} - \sigma_{back}$$  \hspace{1cm} (23)

From eq. (20), an expression for the stationary creep rate can be obtained. In this expression, the creep stress in eq. (23) should be applied

$$\dot{\varepsilon}_{sec} = 2\tau_L M(T, \sigma_{appl} - \sigma_{back}) \rho_{bnd}^{3/2} / \left( k_{bnd} \frac{m}{bcL} - \omega \rho_{bnd}^{1/2} \right)$$  \hspace{1cm} (24)

If we insert the expression for the back stress into eq. (23), we find that

$$\sigma_{creep} = \sigma_{appl} + \sigma_{appl0} - \sigma_{disl}$$  \hspace{1cm} (25)

We now generalize eq. (24) by assuming that it is not only valid for secondary creep but also for the influence of the changes of the dislocation density for the whole creep phase, provided eq. (25) is applied [10]

$$\dot{\varepsilon} = 2\tau_L M(T, \sigma_{appl} + \sigma_{appl0} - \sigma_{disl}) \rho_{bnd}^{3/2} / \left( k_{bnd} \frac{m}{bcL} - \omega \rho_{bnd}^{1/2} \right)$$  \hspace{1cm} (26)
Eq. (26) is the formulation for the whole creep curve including tertiary creep.

4.5 Precipitation hardening

Precipitation hardening is believed to be the most potent way of increasing the creep strength of high temperature alloys [66]. The hardening mechanisms have, however, not been fully understood. An internal stress is introduced to take the effect of particles into account. The internal stress is also referred to back stress or threshold stress. But here, the threshold is assumed to be a constant stress independent of temperature and applied stress. Many times the Orowan stress has been used to estimate the internal stress,

$$\sigma_O = \frac{mCGb}{\lambda}$$

(27)

where $\lambda$ is the interparticle spacing and $C$ a constant equal to 0.8 [67]. However, the estimated stress usually largely overestimates the contribution. The reason is that the stress required for climb is small and much less than expected. A distinction is made between the local and general climb. During local climb the dislocation climbs at the particle/matrix interface, and the dislocation segments between the particles are in their slip plane. For general climb these conditions are relaxed and the dislocations only touch the particles. Many attempts have been made in the past to estimate the threshold stress for climb across particles [68-72]. It has been shown convincingly that general climb gives much lower threshold stresses than local climb. Consequently, it is only necessary to consider general climb, since it will be the controlling mechanism. In the beginning, the threshold stress was estimated to be quite high, but the values were gradually reduced with new researches [68-72]. The best estimated climb stress $\sigma_{cl\text{min}}$ is [71, 72]
\[
\frac{\sigma_{clmin}}{\sigma_0} = \frac{\alpha}{\alpha + 2C}
\]  
(28)

where

\[
\alpha = \frac{2 \bar{r}}{3 \lambda} = \sqrt{\frac{2f}{3\pi}}
\]  
(29)

where \( \bar{r} \) is the mean radius of the particles and \( f \) the volume fraction of precipitate. It gave a threshold stress as low as \( 0.03 \sigma_0 \). However, it is too low to explain the influence of particles on creep strength. In addition, a constant internal gives a stress exponent that decreases with increasing applied stress, which is inconsistent with the experimental results for most materials.

In the thesis, to model the precipitation hardening, an alternative assumption of controlling mechanism was made. Instead of a threshold stress, it was assumed that the time it takes for a dislocation to climb across a particle is the controlling mechanism of whether a dislocation will climb a particle or not. The maximum particle size where there is enough time for dislocations to climb across particles is referred to as the critical radius \( r_{crit} \). Particles with radius below the critical radius will not contribute to the creep strength, since there is sufficient time for the dislocations to climb. Only particles larger than critical radius contribute to creep strength. To calculate the contribution, the Orowan mechanism was used for these larger particles, see eq. (27). In the past, the planar square lattice particle spacing \( \bar{r} \sqrt{2\pi/3f} \) was used for calculating \( \lambda \). It is believed now that the Friedel spacing \( \lambda_F \) can describe the actual spacing of particles along the dislocation line in a better way than the planar square lattice particle spacing (See Paper V).
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\[
\left( \frac{\lambda}{\lambda_F} \right)^3 = \frac{\sigma_{c_{\text{clmin}}}}{\sigma_O}
\]  

(30)

If climb takes less time than glide, climb will not have any significant influence on the creep strength. So the critical radius can be determined when the glide time equals the climb time,

\[ t_{\text{climb}} = t_{\text{glide}} \]  

(31)

where the time for climb can be calculated by the critical radius divided by climb velocity, which is proportional to the climb mobility in eq. (9)

\[ t_{\text{climb}} = \frac{r_{\text{crit}}}{M_{\text{cl}}(T, \sigma)b\sigma} \]  

(32)

The glide time can be calculated by the interparticle spacing \( \lambda \) divided by glide velocity, which relates to creep rate in eq. (18).

\[ t_{\text{glide}} = \frac{\lambda b \rho}{\hat{\varepsilon}m} \]  

(33)

When the expressions in eqs. (32) and (33) are equal, the critical radius can be derived,

\[ r_{\text{crit}} = M_{\text{cl}}(T, \sigma)b^2\sigma\lambda_F \frac{\rho}{\hat{\varepsilon}_{\text{sec}}m} \]  

(34)

The critical interparticle spacing is determined by the particle size distribution. The number of particles per unit area \( N_A \) can be described by

\[ N_A = N_{A0}e^{-k(\bar{r}-\eta_0)} \]  

(35)

where \( N_{A0} = 1/\lambda^2 \) and \( k = 1/(\bar{r}-\eta_0) \). Often satisfactory observations at very small particle sizes are not available. This is
taken into account by introducing the small quantity $r_0$. The
average interparticle spacing for the particles larger than critical
radius is

$$\lambda_{\text{crit}} = \sqrt{N_0 A_0 e^{-k(r_{\text{crit}} - r_0)/2}}$$  \hspace{1cm} (36)

The particle hardening strength can be calculated by Orowan
mechanism

$$\sigma_{\text{partcreep}} = \frac{C G b m}{\lambda_{\text{crit}}} = \sigma_0 e^{-k(r_{\text{crit}} - r_0)/2}$$  \hspace{1cm} (37)

The expression has been used to predict the total creep
strength of austenitic stainless steels. A very good agreement with
experimental data was obtained [11, 20, 62].

4.6 Cavitation

There are a number of mechanisms describing cavity formation,
like grain boundary sliding (GBS) and local high stress [21, 73].
However, it is difficult to explain the experimental stress strain
dependences of cavity formation without assuming that GBS is the
dominating mechanism [16]. The ratio of the GBS displacement
rate and the creep strain rate is a constant, denoted as $C_s$, which is
approximately 50 μm for copper [21]. It is very well established
that both the GBS distance and the number of nucleated cavities
are proportional to the creep strain [74]. It has been experimentally proved that $C_s$ is approximately constant over a
wide range of test conditions (from 125 to 600°C at different strain
rates) [21].

It is commonly assumed that cavities are nucleated at
particles, but particles do not give any significant contribution to
cavitation in pure copper, since there are very few particles.
Instead it has been proposed that the possible nucleation sites of
cavities in copper are the intersections of sub-boundaries with sub-
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grain corners on the other side of a sliding grain boundary. This has been validated with thermodynamic analysis [21]. The derived nucleation rate for cavities is [16]

\[
\frac{dn_{\text{cav}}}{dt} = \frac{0.9 C_s \dot{\epsilon}}{d_{\text{sub}}^3}
\]  

(38)

where \(n_{\text{cav}}\) is the number of cavities, and \(d_{\text{sub}}\) the subgrain size.

The cavity starts to grow when a critical size is exceeded. Diffusion controls the growth of cavities. The growth rate is proportional to the stress. The traditional diffusion controlled cavity growth models grossly overestimated the growth rate. A constrained growth was proposed by Dyson [75]. He suggested that it is not feasible if the cavity growth rate is larger than the creep rate. The constrained cavity growth rate is [76]

\[
\frac{dR_{\text{cav}}}{dt} = 2D_0K_f (\sigma_{\text{red}} - \sigma_0) \frac{1}{R_{\text{cav}}^2}
\]  

(39)

where \(R_{\text{cav}}\) is the cavity radius, \(\sigma_0\) the sinter stress, \(D_0\) a parameter related to the grain boundary diffusion coefficient, \(K_f\) a factor dependent on the cavitated area fraction \(A_{\text{cav}}\). \(\sigma_{\text{red}}\) is the reduced stress. \(\sigma_{\text{red}}\) can be computed with a differential equation [17]

\[
2\pi D_0K_f (\sigma_{\text{red}} - \sigma_0)/L^2 R_{\text{cav}} + \dot{\epsilon}(\sigma_{\text{red}}) = \dot{\epsilon}(\sigma_{\text{appl}})
\]  

(40)

where \(L\) is the cavity spacing. The grain boundary cavitated area fraction \(A_{\text{cav}}\) is given by [47]

\[
A_{\text{cav}} = \int_0^t \frac{dn_{\text{cav}}}{dt'} (t') \pi R_{\text{cav}}^2(t,t') dt'
\]  

(41)

where \(R_{\text{cav}}(t,t')\) is the radius of the cavity at time \(t\) that was formed at time \(t'\). It is believed that rupture takes place when the \(A_{\text{cav}}\) value is larger than 25% [20].
4.7 Parameter values in the models

All the parameter values used in models mentioned above are summarized in Table 2.
Table 2 Parameters used in the thesis

<table>
<thead>
<tr>
<th>Parameter description</th>
<th>Parameter</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Burgers’ vector</td>
<td>$b$</td>
<td>$2.56 \times 10^{-10}$ m</td>
<td></td>
</tr>
<tr>
<td>Taylor factor</td>
<td>$m$</td>
<td>3.06</td>
<td></td>
</tr>
<tr>
<td>Dynamic recovery constant</td>
<td>$\omega$</td>
<td>14.7</td>
<td>[4]</td>
</tr>
<tr>
<td>Mobility</td>
<td>$M$</td>
<td></td>
<td>[48]</td>
</tr>
<tr>
<td>Dislocation line tension</td>
<td>$\tau_L$</td>
<td>$Gb^2/2 = 7.94 \times 10^{-16}$ MN</td>
<td>[77]</td>
</tr>
<tr>
<td>Boltzmann’s constant</td>
<td>$k_B$</td>
<td>$1.381 \times 10^{-23}$ J/grad</td>
<td></td>
</tr>
<tr>
<td>Constant in Taylor’s equation</td>
<td>$\alpha_T$</td>
<td>$(1-v/2)/2\pi(1-v)=0.19 \text{ with Poisson's ratio } v=0.308$</td>
<td>[78, 79]</td>
</tr>
<tr>
<td>Shear modulus</td>
<td>$G$</td>
<td>$G=45400(1-7.1\times10^{-4}(T-20))$ MPa, $T$ in °C</td>
<td>[80]</td>
</tr>
<tr>
<td>Work hardening constant</td>
<td>$k_{bnd}$</td>
<td>1.6-1.7</td>
<td>[8]</td>
</tr>
<tr>
<td>Work hardening constant</td>
<td>$k_{bnde}$</td>
<td>1.6-1.7</td>
<td>[8]</td>
</tr>
<tr>
<td>Work hardening constant</td>
<td>$C_L$</td>
<td>30</td>
<td>[7]</td>
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<td>Symbol</td>
<td>Value</td>
<td>Reference</td>
</tr>
<tr>
<td>-------------------------------</td>
<td>--------</td>
<td>-------------------------------</td>
<td>-----------</td>
</tr>
<tr>
<td>Grain size</td>
<td>$d$</td>
<td>$1 \times 10^{-4}$ m</td>
<td>[4]</td>
</tr>
<tr>
<td>Cavity radius</td>
<td>$r_h$</td>
<td>$1 \times 10^{-6}$ m</td>
<td>[47]</td>
</tr>
<tr>
<td>GBS parameter</td>
<td>$C_s$</td>
<td>50 µm</td>
<td>[18]</td>
</tr>
<tr>
<td>Subgrain size</td>
<td>$d_{sub}$</td>
<td>$\frac{KGb}{\sigma_{appl0}}$</td>
<td>with $K=11$ for copper</td>
</tr>
<tr>
<td>Parameter in the cavitation growth equation</td>
<td>$K_l$</td>
<td>$\frac{1}{-2 \log A_{cav} - (1 - A_{cav})(3 - A_{cav})}$</td>
<td>[17] where $A_{cav}$ is given in eq.(41)</td>
</tr>
<tr>
<td>Grain boundary diffusion coefficient</td>
<td>$\delta D_{GB}$</td>
<td>$\delta D_{GB} \exp(-\frac{Q_{GB}}{RT})$</td>
<td>with $\delta=2b$, $D_{GB0}=2.4 \times 10^{-6}$ m²s⁻¹</td>
</tr>
<tr>
<td>Cavity spacing</td>
<td>$L$</td>
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<td>[18]</td>
</tr>
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<td>Atomic volume</td>
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<td></td>
</tr>
<tr>
<td>Grain boundary diffusion parameter</td>
<td>$D_0$</td>
<td>$\delta D_{GB}\Omega/k_B T$</td>
<td>[17]</td>
</tr>
<tr>
<td>Particle radius</td>
<td>$r_0$</td>
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</tr>
<tr>
<td>Orowan equation constant</td>
<td>$C$</td>
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<td>[67]</td>
</tr>
</tbody>
</table>
5 Finite element method

5.1 Notched specimens

The finite element method (FEM) has been widely used for investigating the stress distribution and damage evolution around the specimen notches [82-84]. In order to study the distribution of deformation in the notched specimens of copper, FEM was performed using the commercial software Comsol Multiphysics. Both creep and SSRT tests were simulated by subjecting the notched specimens to an axial load. The notched specimens exhibit two-dimension axial symmetry as well as a symmetry plane in the middle of the specimen. For this reason the specimen geometry was simplified to a rectangle with width equal to the specimen radius and height equal to half of the specimen length and only one notch was involved. The mesh was physics-controlled with extra fine size, see Figure 10. The minimum mesh size that lies around the notch is $2 \times 10^{-6}$ m.

To model the SSRT tests, elasto-plastic stress analysis was performed. Since the ductility of all the specimens were over 10%, a model with large plastic strain was selected. Eq. (16) was used as isotropic hardening function in computation.

To model the creep tests, a time dependent creep model was used. Eq. (18) was implemented to describe the secondary creep rate.
Figure 10 Specimen geometry and mesh used in FEM for notch acuity 0.5, 2 and 5

5.2 Reference stress

Reference stress is often used to evaluate the rupture life of complex components under multiaxial stress state [52]. There are various methods to estimate the value of the reference stress. The models proposed by Hayhurst et al. [85] and Nix et al. [86] have been widely used. They took into account the relative contribution from each stress component governing creep life, including maximum principal stress, hydrostatic stress and von Mises stress.

In the present thesis, to determine the reference stress for notched specimens, FEM analysis was performed with perfect plasticity conditions [87]. It was a two-dimensional elasto-plastic computation. The isotropic hardening equation was then selected as perfect plasticity, where there is no kinematic hardening term.
The reference stress was computed in steps. First, the stress was raised until a region with the yield stress was formed across the specimen. Then, the stress was further increased until the computation no longer converged. The reference stress was taken as the maximum von Mises stress in the band when the computations still converged.

5.3 Necking

Necking is defined as a loss of stability for small variation of cross section area [88, 89]. At the last few percent of tensile creep life, the dominant damage mechanism is necking [90].

The creep necking phenomenon was investigated in the present thesis with the aid of FEM computation. Since the basic tertiary creep model contains multiple variables, it cannot be used as constitutive equations in FEM in a straightforward manner. Only the secondary creep stage could be simulated.

During FEM computation, severe necking was obtained. The computed profile was used to compare with experimental necking profile, as shown in Figure 11. The computed results reproduce experimental radii within about 10%.
**Figure 11** Comparison of experimental necking profile with FEM results for Cu-OFP at 75 °C, 175 MPa
6 Summary of appended papers

This thesis contains both experimental and modelling work. In papers I and II, SSRT and creep tests were conducted to simulate the deformation of copper canister under service conditions. The basic models for stress strain curves and for primary and secondary creep described in Sections 4.1, 4.2 and 4.3 were used to reproduce the experimental curves. Papers III and IV focused on the modelling of tertiary creep at both low and higher temperatures based on the basic models described in Section 4.4. Paper V studied the precipitation hardening with the help of the model in Section 4.5.

In summary, the thesis demonstrates that the basic models can be used to describe both stress and strain controlled deformation under both uniaxial and multiaxial conditions. Tertiary creep can be represented with fundamental models. In addition, the mechanism of precipitation hardening can be accurately simulated.

Paper I Slow strain rate tensile tests on notched specimens of copper

SSRT tests were performed for notched specimens of copper at 75 °C and 125 °C with strain rates of $10^{-6}$ and $10^{-7}$ s$^{-1}$. FEM computation was conducted to interpret the test results. The basic models described in Section 4.1 and 4.2 were adapted and used in FEM as constitutive equations. The modelled stress strain curves were compared with the ones obtained in experiments. A satisfying representation of experimental curves can be seen in Figure 12, indicating that the basic models developed for uniaxial strain controlled case can be a basis for describing multiaxial deformation.
**Figure 12** Comparisons of FEM modelled stress strain curves with experimental results under different test conditions

**Paper II Creep tests on notched specimens of copper**

Creep tests were performed for notched specimens of copper at 75 °C under constant load with the net section stresses ranging from 170 MPa to 245 MPa (details can be seen in Table 1). FEM computation was conducted to interpret the test results. The basic models described in Section 4.1 and 4.3 were used in FEM as constitutive equations. The modelled time creep strain curves were compared with the experimental ones. A reasonable representation of the experimental curves was obtained although discrepancy exists (Figure 13), demonstrating that the basic models derived for
uniaxial stress controlled case can be used to describe multiaxial creep deformation.

**Figure 13** Comparisons of FEM modelled time strain curves with experimental results under different test conditions

**Paper III Basic modelling of tertiary creep of copper**

Creep damage mechanisms including microstructure degradation, cavitation and necking instability have been discussed for a long time in the literature [91, 92]. There are numerous empirical models describing cumulative creep damage, few of which have taken all the relevant mechanisms into account. In the present paper, a model for tertiary creep has been formulated. The model was developed based on previous basic models for secondary creep and took the contribution of substructure into account. It was
concluded that the largest contribution to tertiary creep of copper came from dislocation structure. Other contributions considered were from cavitation and necking. The basic models described in Section 4.1 and 4.4 were used to reproduce tertiary creep of copper at low temperatures. A comparison was made between the modelled creep curves and experimental curves in Figure 14 and a well representation was obtained. In conclusion, the tertiary creep of copper at low temperatures can be modelled with basic models.

**Figure 14** Comparisons of modelled creep curves with experimental results under different test conditions

**Paper IV** Fundamental modelling of mechanisms contributing to tertiary creep in copper at 215 and 250 °C

This is an extension of the work of Paper III. It has been suspected
that the creep mechanisms are different in low temperature and higher temperature creep. It is of interest to verify if the model for tertiary creep at low temperature can be used at higher temperatures. The models in Section 4.1 and 4.4 were then applied for creep of copper at 215 and 250 °C. The modelled creep curve was compared with experimental curve as given in Figure 15. A similar result as Paper III was obtained, indicating that the model is applicable at both low and higher temperatures.

Figure 15 Comparison of modelled creep curves with experimental result at 250 °C, 120MPa

Paper V Creep strength contribution due to precipitation hardening in Copper-Cobalt alloys

Precipitation hardening models in Section 4.5 were used in the paper to describe the influence of Cu-Co particles on the creep rate. Cu-Co alloy was chosen because it is a simple system for investigating the effect of particles on creep strength, which can
easily avoid the influence of solid solution hardening. The modelled results were compared with experimental data. Figure 16 shows a good agreement of the modelled creep rate with experimental results.

**Figure 16** Modelling of stationary creep rate for Cu-Co alloys and pure copper compared with experimental data
7 Conclusions

This thesis studied the creep properties of copper in experimental and theoretical contexts. From the experimental results, some conclusions have been drawn:

- Cu-OFP was found to show notch strengthening. Creep rupture strength increased with the increasing sharpness of notches.

- Ductile creep rupture mechanism was observed for all ruptured specimens from both slow strain rate tensile tests and creep tests. Very limited creep cavitation was observed.

- Severe necking was seen for all ruptured specimens. The stress triaxiality increases with notch acuity. As a consequence, specimens with sharper notches yield lower ductility.

The experimental results were used to compare with physically based basic models without any fitting parameters. Main conclusions have been drawn, as follows:

- In a unique effort, constitutive equations based on fundamental materials models have been applied in FEM to describe the deformation of specimens that have been exposed to multiaxial stresses. Both stress and strain controlled plastic deformations of copper have been analyzed in this way with a reasonable success.

- A fundamental model for tertiary creep of copper has been formulated. It is believed to be the first fundamental model for tertiary creep where all the relevant creep damage mechanisms are taken into account, in the case of copper dislocation recovery,
CONCLUSIONS

cavitation and necking.

- A crucial factor is to consider the role of the substructure.

- The model predicts that necking does not take place until the very end of creep life. This is in agreement with published data for CrMo-steels.

- Cavitation is not a critical contribution for creep damage of copper at both low (75 °C) and higher (215 and 250 °C) temperatures. The modelled and experimental (75°C) cavitated area fraction was small enough to be ignored.

- An extended model for the contribution of precipitation to the creep strength is presented where the limiting factor for precipitation hardening is the time it takes for a dislocation to climb across a particle. To verify the model, it has been compared with published experimental data for Cu-Co alloys. This system is of particular interest, since the precipitates make by far the largest contribution to the creep strength. For the first time, it has been verified that the temperature and stress dependence of the internal stress from the particles can be well predicted by a model without using adjustable parameters. In addition, the model can at least approximately describe the influence of alloy composition and heat treatment for the Cu-Co alloys.
FUTURE WORK

8 Future work

The thesis is part of the effort to develop basic models for creep and other mechanical properties. This is certainly vital for disposal of nuclear waste, where the waste package must be safe for thousands of years. Without basic models, predictions over such extended periods are not meaningful. This issue is also of major importance for new generation of nuclear reactors where the design lifetime is 60 years. Fundamental models also dramatically improve the understanding of the controlling mechanisms. Although it is of critical importance for future technical application, fundamental modelling has only been performed to a limited extent so far and only a limited number of research groups are active in this field internationally. The need for further work is therefore immense. Some suggestions for future work are given:

- Existing fundamental models should be analyzed for a wider range of materials and test conditions.
  - Basic modelling of cavitation has mainly been studied for copper and austenitic stainless steels. Other creep resistant steels are of great interest to investigate.
  - Creep ductility is vital in most high temperature applications but the mechanisms are poorly understood for many alloy systems.

- New fundamental models describing other mechanical properties should be developed.
  - Very few fundamental models have been formulated for low cycle fatigue that is a concern in many high temperature applications.
  - Creep crack growth is also an area where more basic modelling is needed.
Acknowledgement

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Fangfei Sui

Stockholm, April 2018
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Paper I
Slow strain rate tensile tests on notched specimens of copper

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Abstract

In this study, slow strain rate tensile tests have been performed on phosphorus alloyed copper under uniaxial and multiaxial stress states at 75 and 125 °C with two strain rates of $10^{-6}$ and $10^{-7}$ s$^{-1}$. Multiaxial stress states have been introduced by incorporating three different notch geometries on the uniaxial specimens. It has shown that the presence of the notches decreased the strength and ductility of copper. Ductility exhaustion was likely to be the dominant rupture mechanism. Finite element analysis was conducted to compare with the experimental results with a physically based model for stress strain curves without fitting parameters. The model could successfully describe the experimental data, and it could predict the dependence of acuity, temperature and strain rate in the multiaxial tests.

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1. Introduction

Deep geological disposal is believed to be the most appropriate solution for long term management of spent nuclear fuel. In Sweden, a specific concept named KBS-3 is proposed. The spent nuclear fuel will be enclosed in tightly sealed copper canisters surrounded by clay, and buried 500 m down in the bedrock as the final repository [1,2]. The three barriers, copper canister, clay and bedrock, will serve with the principal purpose of delaying the transport of radionuclides. Copper is selected because of its excellent corrosion resistance in reducing ground water and thereby isolating the spent fuel from the surrounding environment. In the repository, the copper canister will also be subjected to mechanical loading at temperatures up to 100 °C. The clay will swell when it comes into contact with underground water. The swelled clay will exert a pressure on the canister. During the swelling process, the load on the canister will gradually increase and it may take centuries before full pressure is reached [3]. In the swelling stage the loading is essentially strain controlled whereas stress control will be activated after full pressure has been reached. In this study, the gradual loading is simulated with slow strain rate tensile (SSRT) tests and full loading with creep tests. In order to describe the role of the deformation of the copper canister, both SSRT and creep tests should be carried out.

Many uniaxial tests (both SSRT and creep) on copper have previously been performed [4–7]. For investigating the role of the complex stress conditions in the repository, it is natural to perform tests also under multiaxial stress state [8–10]. A series of multiaxial creep tests have been conducted [11,12]. However, literatures provide limited information about the performance of copper during SSRT tests under multiaxial stress state.

The purpose of the present work is to perform SSRT tests for copper under multiaxial states. The tests results are used to verify that the basic models which have been successfully developed for uniaxial tests can be used to describe multiaxial tests as well.

2. Material and testing

The canisters used for storage of the spent nuclear fuel are made of oxygen free copper alloyed with 50 ppm phosphorus (Cu-OFP). According to previous research, pure copper was detected to sometimes have extra low creep ductility [13], and phosphorus was added to give copper acceptable creep ductility [14–16]. In order to simulate the multiaxial stress state, double notched Cu-OFP cylinder bars are used in the present tests. The chemical composition of the test material is given in Table 1.

By changing the notch geometry, various stress states can be obtained [17]. Multiaxial SSRT tests were carried out with three kinds of notch geometries. Plain copper bars were also tested as reference. The geometries of the specimens are presented in Fig. 1 and Table 2. To indicate different notch geometries, notch acuity is defined as the notch root radius divided by the notch throat radius. All the specimens were 7.98 mm in diameter, 47 mm in gauge length and 2.82 mm in notch throat radius. The notch root radii of the specimens were varied in the range from 0.564 to 5.64 mm for achieving different notch acuities.

The specimens were tested at two temperatures, 75 and 125 °C,
Table 1
Chemical composition of the Cu-OFP cylinder bars, ppm.

<table>
<thead>
<tr>
<th>Cu (%)</th>
<th>Ag</th>
<th>As</th>
<th>Bi</th>
<th>Cd</th>
<th>Ni</th>
<th>Zn</th>
<th>Fe</th>
<th>Mn</th>
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<td>99.992</td>
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<td>13.2</td>
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<td>0.85–0.87</td>
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<td>0.003</td>
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<td>&lt; 1.1–1.2</td>
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<tr>
<td>1.6–2.4</td>
<td>Sb</td>
<td>S</td>
<td>Pb</td>
<td>Se</td>
<td>Sn</td>
<td>H</td>
<td>Te</td>
<td>P</td>
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<td>0.06</td>
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<td></td>
<td></td>
<td></td>
<td></td>
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<td>5.3–5.6</td>
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<td>0.26–0.29</td>
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Table 2
Specimen geometry and test conditions.

<table>
<thead>
<tr>
<th>Gauge length (mm)</th>
<th>Stress state</th>
<th>Notch root radius (mm)</th>
<th>Notch throat radius (mm)</th>
<th>Notch acuity</th>
<th>Test temperatures (°C)</th>
<th>Strain rate (s⁻¹)</th>
</tr>
</thead>
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<tr>
<td>Ø798 × 47</td>
<td>Uniaxial</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>75, 125</td>
<td>1 × 10⁻⁶, 1 × 10⁻⁷</td>
</tr>
<tr>
<td></td>
<td>Multiaxial</td>
<td>5.64</td>
<td>2.82</td>
<td>0.5</td>
<td>75, 125</td>
<td>1 × 10⁻⁶, 1 × 10⁻⁷</td>
</tr>
<tr>
<td></td>
<td>Multiaxial</td>
<td>1.14</td>
<td>2.82</td>
<td>2</td>
<td>75, 125</td>
<td>1 × 10⁻⁶, 1 × 10⁻⁷</td>
</tr>
<tr>
<td></td>
<td>Multiaxial</td>
<td>0.564</td>
<td>2.82</td>
<td>5</td>
<td>75, 125</td>
<td>1 × 10⁻⁶, 1 × 10⁻⁷</td>
</tr>
</tbody>
</table>
Since the dynamic and static recovery are controlled by the same dislocation mechanisms, an assumption was made in Ref [6] that their maximum rates are the same. This assumption determines the relation between stress strain curves and creep data, which will be discussed below. When the maximum rates of the dynamic and static recovery are equal, an expression for the maximum dislocation density can be derived. Taylor’s equation is used to transfer the dislocation density to strength. With the help of Taylor’s equation, the maximum stress \( \sigma_{\text{max}} \) can be obtained.

\[
\sigma_{\text{max}} = \sigma_y + \frac{m \alpha G b}{2 (1 - \nu) M (T, \sigma_{\text{max}})} \]  

(5)

where \( \sigma_y \) is the yield strength, \( \alpha \) a constant in Taylor’s equation, \( G \) the shear modulus, \( \dot{\varepsilon} \) is the stationary strain rate given in Eq. (6).

\[
\dot{\varepsilon} = \frac{2 \beta c M (T, \sigma)}{m} \]  

(6)

To describe the stress strain curves, the work hardening and dynamic recovery are considered. In fact, the last term on the right hand side of Eq. (1) is usually negligible in this case. By integrating Eq. (1) and using the dislocation mobility for Cu-OFP, the expression for stress strain curves can be obtained.

\[
\sigma = \sigma_y (T, \dot{\varepsilon}) + K (1 - e^{-\eta \dot{\varepsilon}^m}) 
\]

(7)

where

\[
K = \sigma_{\text{max}} - \sigma_y (T, \dot{\varepsilon})
\]

(8)

The elastic contribution is unimportant in the present case and has therefore been neglected. The value of the yield strength \( \sigma_y (T, \dot{\varepsilon}) \) depends on the temperature and strain rate and is related to a reference condition \( (\dot{\varepsilon} = \dot{\varepsilon}_0 = 1 \times 10^{-4} \text{ s}^{-1} \) and \( T = T_0 = 20 \text{ °C} \) for Cu-OFP). This relation is given in Eq. (9), which was originally derived in [6].

\[
\sigma_y (T, \dot{\varepsilon}) = \sigma_y (T_0, \dot{\varepsilon}_0) \frac{G (T)}{G (T_0)} \left( \frac{\dot{\varepsilon}}{\dot{\varepsilon}_0} \right)^{(1/\eta)}
\]

(9)

\( \eta \) is the stress exponent, which depends on the stress and temperature. It can be determined from the stationary creep rate from Eq. (6). For example, \( \eta \) values are 86, 66, at 75 and 125 °C for \( \dot{\varepsilon} = \dot{\varepsilon}_0 \) for Cu-OFP.

The model for stress strain flow curves, Eq. (7), has been used to compare with the SSRT tests under uniaxial and multiaxial stress states in the present work.

Constants used in the model are listed in Table 3.

### 3.2. Finite element implementation

The finite element (FE) analysis in this study was conducted using Comsol Multiphysics 4.4a. A two dimensional elasto-plastic stress analysis with a large plastic strain model has been performed. In the plasticity model, the first term in Eq. (7) \( \sigma_y (T, \dot{\varepsilon}) \), was used as initial yield stress and second term as isotropic hardening function. \( \sigma_y (T, \dot{\varepsilon}) \) and \( K \) values derived by Eqs. (8) and (9) are given in Table 4.

The geometry of the notches with FE mesh is shown in Fig. 3. The entire geometry was meshed with

### Table 3

<table>
<thead>
<tr>
<th>Parameter description</th>
<th>Parameter Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coefficient for self-diffusion</td>
<td>( D_n )</td>
<td>( 1.3 \times 10^{-5} \text{ m}^2/\text{s} )</td>
</tr>
<tr>
<td>Activation energy for self-diffusion</td>
<td>( Q )</td>
<td>( 198,000 \text{ J/mol} )</td>
</tr>
<tr>
<td>Burgers’ vector</td>
<td>( b )</td>
<td>( 2.56 \times 10^{-10} \text{ m} )</td>
</tr>
<tr>
<td>Taylor factor</td>
<td>( m )</td>
<td>( 3.06 \times 10^{17} )</td>
</tr>
<tr>
<td>Dynamic recovery constant</td>
<td>( \sigma )</td>
<td>( 4.75 \times 10^{19} )</td>
</tr>
<tr>
<td>Poisson’s ratio</td>
<td>( \nu )</td>
<td>( 0.308 )</td>
</tr>
<tr>
<td>Constant</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Max back stress</td>
<td>( \sigma_{\text{max}} )</td>
<td>( 257 \text{ MPa} )</td>
</tr>
<tr>
<td>Dislocation line tension</td>
<td>( \tau_L )</td>
<td>( 7.94 \times 10^{-16} \text{ Mn} )</td>
</tr>
<tr>
<td>Boltzmann’s constant</td>
<td>( k_B )</td>
<td>( 1.381 \times 10^{-23} \text{ J/grad} )</td>
</tr>
<tr>
<td>Shear modulus</td>
<td>( G )</td>
<td></td>
</tr>
<tr>
<td>Maximum interaction energy between a P solute and a dislocation</td>
<td>( U_{p}^{\text{max}} )</td>
<td>( 8220 \text{ J/mol} )</td>
</tr>
</tbody>
</table>

### Table 4

<table>
<thead>
<tr>
<th>Temperature (°C)</th>
<th>Strain rate (s(^{-1}))</th>
<th>( \sigma_y (T, \dot{\varepsilon}) ) (MPa)</th>
<th>K (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>75</td>
<td>( 10^{-6} )</td>
<td>59.7</td>
<td>187.9</td>
</tr>
<tr>
<td>75</td>
<td>( 10^{-7} )</td>
<td>57.7</td>
<td>182.1</td>
</tr>
<tr>
<td>125</td>
<td>( 10^{-6} )</td>
<td>56.3</td>
<td>170.2</td>
</tr>
<tr>
<td>125</td>
<td>( 10^{-7} )</td>
<td>53.5</td>
<td>162.9</td>
</tr>
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</table>
4. Results

4.1. Experimental results

16 tests were conducted in total and all the specimens went to rupture. Fig. 4 shows the stress strain flow curves. As can be seen in Fig. 4, the ultimate tensile stress under uniaxial stress state (notch acuity 0) is much higher than those under multiaxial stress state (notch acuity 0.5, 2 and 5). Under multiaxial stress state, the tensile strength gradually increases with notch acuity. Moreover, the temperature dependence of the flow strength is evident. The flow strength decreases with increasing temperatures at a given strain rate. The influence of the strain rate on the flow strength is not as pronounced as the temperature dependence.

In addition, the rupture elongation of the uniaxial tests is much higher (about 3 times) than that in the multiaxial tests. However, different notch acuities had little effect on the rupture elongation.

The reduction in area is a valuable measure of ductility in notched specimens [29–31]. The reduction in area as a function of the notch acuity is plotted in Fig. 5. In general, all the specimens under both uniaxial and multiaxial stress states gave high ductility. The reduction in area always exceeded 70%, even with the presence of sharp notches. It can be seen that the reduction in area of the notched specimens is lower than the plain specimens. With the increase of notch acuity, the reduction in area is lowered, i.e. the sharper the notch, the lower the ductility. Similar trends have been observed by Charde et al. on 316 L (N) austenitic stainless steel [29]. Since both austenitic stainless steel and copper are fcc materials, their properties are related. At the same strain rate, the ductility of copper at 75 °C is higher than that at 125 °C, Fig. 5.

Metallographic studies were carried out on the fracture...
surfaces of the specimens to examine the failure mode. Typical ductile failure with cup and cone type appearance was obtained (Fig. 6). Shear lip and dimple part are present in Fig. 6b. Predominantly dimple fracture can be observed at the center (Fig. 6d), where the dimples are much bigger in size and deeper in depth compared with the outer shear lip part (Fig. 6c). The fracture mode was found to be independent of notch acuity and test conditions. All the failed specimens showed similar ductile failure appearance as in Fig. 6.

After tensile tests, cavitations were examined at the longitudinal sections near the ruptured notches. Only few randomly distributed creep cavities were observed at 125 °C, as shown in Fig. 7. However, no coalescence of cavities along grain boundaries was found. The observed cavities are considered to be too few to influence the properties significantly. Intergranular creep cavitation has been found by Wu et al. on the same material in creep tests [11,14]. Comparing the present SSRT test with the previous creep tests, the duration is shorter. The longest SSRT test in the present study lasted for 1500 hours while the creep tests were conducted for up to 10,000 h. With shorter duration, there is less possibility to form creep cavities.

4.2. Model results and discussion

In the FEM computation, the second term in Eq. (7) was used as the hardening equation. Although the equation has the similar expression as the Voce empirical model, it is the same basic model as given in refs [5,6]. It is derived from basic dislocation models. The parameters in the equation are all based on physical mechanisms and precise values are derived. None of the parameters are fitted to the experimental data. It has been used and compared with the experimental results from SSRT tests under both uniaxial and multiaxial stress states. The results are shown as engineering stress strain curves. The reason is that it is easy to transfer engineering results to true results in uniaxial tests, while there is no unique way to transfer the multiaxial tests results to true results. Also, using engineering curves is convenient to assess the uniform elongation value, which can be found directly from the curves.

The results for the four notch acuities are illustrated in Fig. 8. The general behaviors of the experimental flow curves and FEM results are the same, indicating that the model can reproduce the
stress strain curves in a satisfactory way.

Since the model does not take necking into account, comparison of the flow curve with experimental data after necking is meaningless. However, the maximum stress point, and thereby the tensile strength and the uniform elongation could be predicted. The measured and modeled tensile strength values are presented in Fig. 9. The notch dependence of the tensile strength is quite apparent. There is a dramatic decrease from notch 0 to notch 0.5; after that the tensile strength increases slightly with increase of notch acuity. It is evident that the FE computation can predict the notch acuity dependence. The tensile strength is higher at 75 °C than at 125 °C. The experimental and modeled tensile strength are larger at the higher strain rate $10^{-6} \text{s}^{-1}$ than at $10^{-7} \text{s}^{-1}$ except at acuity 0. The agreement of test data and model values indicates that the model could predict temperature and strain rate dependence.

The maximum stress point in the stress strain curve also gives the uniform elongation. The comparison of the measured uniform elongation with the FE modeled values is shown in Fig. 10. Notch acuity dependence seems to be the dominant effect on this value and the trend is the same as for the tensile strength. The uniform elongation decreases sharply from uniaxial to multiaxial state and

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**Fig. 7.** Light optical microscope images showing few cavities in: (a) notch acuity 0.5 specimen tested at 125 °C, $10^{-6} \text{s}^{-1}$ and (b) plain specimen tested at 125 °C, $10^{-7} \text{s}^{-1}$.

**Fig. 8.** Comparison of the flow curves between the model and experiments of specimens with: (a) notch acuity 0, 75 °C, $10^{-6} \text{s}^{-1}$; (b) notch acuity 0.5, 125 °C, $10^{-6} \text{s}^{-1}$; (c) notch acuity 2, 125 °C, $10^{-6} \text{s}^{-1}$ and (d) notch acuity 5, 125 °C, $10^{-7} \text{s}^{-1}$.
Fig. 9. Measured and modeled tensile strength at 75 °C, 10^{-6} \text{s}^{-1}; 125 °C, 10^{-6} \text{s}^{-1}; 75 °C, 10^{-7} \text{s}^{-1}; 125 °C, 10^{-7} \text{s}^{-1}.

Fig. 10. Comparison of uniform elongation with FE modeled values at 75 °C, 10^{-6} \text{s}^{-1}; 125 °C, 10^{-6} \text{s}^{-1}; 75 °C, 10^{-7} \text{s}^{-1}; 125 °C, 10^{-7} \text{s}^{-1}.

then increases slightly with the increasing notch acuity. This confirms that the model can take into account the influence of the notch acuity.

The temperature and strain rate dependence is weak in the model results. At a given notch acuity, the model gives almost the same uniform elongation value despite of varying temperatures and strain rates. Although the uniform elongations measured at different temperatures and strain rates are not identical, the differences are modest. The predicted values for the uniaxial tests (notch acuity 0) are slightly lower than the experimental data, while for the multiaxial tests they are in the range of the experimental data. It should be noticed that the stress strain curves under uniaxial stress state are quite flat when they reach the maximum stress (Fig. 4), so there is some uncertainty in the uniform elongation value. Therefore, the model is thought to give a satisfactory description of the uniform elongation value.

According to Eq. (8), the $K$ values represent the maximum stress in the flow curves apart from the yield strength. $K$ is plotted as a function of strain rate for Cu-OFP at 75 °C in Fig. 11. The $K$ values are compared to creep data. Experimental secondary creep rates from ref [6] are shown at different applied stresses at 75 °C. It can be seen that the modeled $K$ values at a given strain rate agree well with the creep stress that gives the same secondary creep rate as the strain rate in the tensile tests. As pointed out above, the assumption is made that the maximum dislocation density is the same when only static respectively only dynamic recovery is taken into account. A consequence is that the flow stress curves are directly related to the creep data. The $K$ values at a given strain rate are identical to the creep stress at the same secondary creep rate. In addition, the $K$ values can be compared with experimental creep data directly. Fig. 11 demonstrates that the assumption made above is reasonable. The good agreement between modeled $K$ values at a given strain rate and creep stress at the same secondary creep rate indicates that the basic model (without any fitting parameters) can describe both creep and stress strain data.

The model can represent the stress strain flow curves and also predict the necking point. This model is based on dislocation mechanisms. All the involved parameters are available or can be derived and none of them is fitted to the experimental data. It has been demonstrated that the basic models can be used to describe both strain controlled and stress controlled deformations under uniaxial and multiaxial stress states. Examples of stress controlled deformation can be found in Refs [5,12]. The models are also applicable to other materials. Results on stainless steels are for example given in ref [32].

5. Conclusions

SSRT tests of Cu-OFP have been performed under both uniaxial and multiaxial stress state under different test conditions (75 and 125 °C with strain rates of 10^{-6} and 10^{-7} \text{s}^{-1}). FE analysis was conducted to analyze the data. A physically based model for plastic deformation without any fitting parameters has been used to compare with the test results. The following conclusions can be drawn:

- The presence of a notch decreased the tensile strength and the elongation and the reduction in area. The sharper the notch, the
lower the ductility.

- Few creep cavities coalescence were observed. Ductility exhaustion and reduction of load bearing capacity were likely to be the dominant rupture mechanism.
- A previously developed basic model for stress strain flow curves could describe the experimental data under uniaxial and multiaxial stress states at 75 and 125 °C with strain rates of $10^{-6}$ and $10^{-7}$ s$^{-1}$.
- Together with previously published results, it has been demonstrated that the basic model can be used to describe both strain controlled and stress controlled deformation under uniaxial and multiaxial stress states without the use of adjustable parameters.

Acknowledgements

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References

Creep Tests on Notched Specimens of Copper

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Abstract

In Sweden, spent nuclear fuel is planned to be disposed off by placing it in canisters which are made of oxygen free copper alloyed with 50 ppm phosphorus. The canisters are expected to stay intact for thousands of years. During the long term disposal, the canisters will be exposed to mechanical pressure from the surroundings at temperatures up to 100 °C and this will result in creep. To investigate the role of the complex stress conditions on the canisters, creep tests under multiaxial stress state are needed. In the present work, creep tests under multiaxial stress state with three different notch profiles (acuity 0.5, 2, and 5, respectively) at 75 °C with net section stresses ranging from 170 MPa to 245 MPa have been performed.

To interpret the experimental results, finite element computations have been conducted. With the help of the reference stress, the rupture lifetime in the multiaxial tests was estimated. The prediction was more precise for the higher acuities than for the lower one. In order to predict the creep deformation of the canisters for the long service period, fundamental creep models are considered. Previously developed basic models are used to compute the creep deformation in the multiaxial tests. Although the scatter is large, the agreement with the experiments is considered as acceptable, indicating that the basic models which have been successfully developed for uniaxial creep tests can also be used to describe multiaxial creep tests. Notch strengthening was observed for copper.

Keywords

Multiaxial stress state; Creep; Notched specimen; Finite element modelling; Copper

1. Introduction

Deep geological disposal is generally considered as a suitable solution for long term management of spent nuclear fuel. It is based on adopting multi barrier methods; different barriers can serve to reduce an overall risk of leakage of radionuclides. A designed concept in Sweden, denoted as the KBS-3 concept, means that the spent nuclear fuel assemblies are placed in iron inserts inside sealed copper canisters which are in turn placed in deposition holes, embedded in a bentonite clay, and buried 500 meters down in the bedrock as the final repository [1]. The repository is planned to be functional for thousands of years until the radioactive waste is no longer hazardous. After disposal, the bentonite will gradually be saturated by
ground water and swell. The copper canister will be subjected to hydrostatic pressure from the ground water and swelling pressure from bentonite. The heat from radioactive waste will raise the temperature in the repository to up to 100 ºC. The pressure at elevated temperature will give rise to creep deformation of the copper canisters.

The canister is five meters long with a diameter of one meter. It is composed of three parts, the lid, tube and base. The lid and base are sealed to the tube by friction stir welding. At the connections between lid/base and tube there exists slits about 75 mm in length and 0.15-0.33 mm in width. Considering the existence of slits, large stress concentration will appear. The top section of the canister is exposed to horizontal and vertical loads that give rise to complex multiaxial stresses [2, 3].

To simulate the service condition of the canisters, creep tests under multiaxial stress state have been performed. Practically, creep tests of components are difficult and expensive to perform. A well-established way to introduce multiaxial stress state is to subject circumferentially notched bars to axial load [4, 5]. Extensive studies have been carried out with notched bars to examine the notch sensitivity of materials with respect to creep failure [6-9].

Finite element modelling (FEM) has been widely used for estimating stress distribution and damage evolution around the notches under creep conditions [5, 10-13]. To interpret the experimental results, finite element computations have been conducted. For safe extrapolation, the controlling mechanisms of creep deformation should be fully understood. Considering the extensive periods that the copper canisters have to stay intact in the repository, the use of fundamental models is of major importance [14]. Fundamental models based only on deformation mechanisms have been developed [15, 16]. It has been demonstrated that the models can successfully reproduce uniaxial creep tests and stress strain curves under both uniaxial and multiaxial stress state for different kinds of materials [14, 17-19].

The purpose of the present work is to perform creep tests under multiaxial stress state and use the data to verify that the basic models can be used to interpret creep tests under multiaxial stress state.

2. Material and testing

The test materials used in this study were extracted from a forged lid provided by Svensk Kärnbränslehantering AB (Swedish Nuclear Fuel and Waste Management Company, SKB). The lid was made of oxygen free copper doped with about 50 ppm phosphorus (Cu-OFP). The test material is the same as used in Ref [20], where its chemical composition is specified. The microstructure of the material before testing is shown in Figure 1. Grains with different sizes and twin boundaries were observed. The grain size was ranging from 85 µm to 156 µm.
In order to simulate multiaxial stress states, double notched cylindrical specimens were used in this study. These specimens were made with a gauge length of 47 mm and a total length of 129 mm. Three kinds of notch profile were introduced with notch acuity 0.5, 2 and 5, representing different stress states. The notch acuity was defined as the notch root radius (a) divided by the notch throat radius (R). The geometry of the notched specimens and detailed notch sizes are shown in Figure 2.

![Figure 1](image1.png) Figure 1 Microstructure of Cu-OFP material.

![Figure 2](image2.png) Figure 2 Specimen geometry used in creep tests.
The specimens were tested at 75 °C under constant dead load. The initial applied net section stress was calculated by the load divided by the minimum notch cross section area. The stresses are listed in Table 1. The tests were planned to run to rupture, but some tests were interrupted due to unexpected long test duration. A few uniaxial creep tests were also conducted for the same batch of material.

<table>
<thead>
<tr>
<th>Table 1 Specimen test conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Notch acuity</td>
</tr>
<tr>
<td>0</td>
</tr>
<tr>
<td>0.5</td>
</tr>
<tr>
<td>2</td>
</tr>
<tr>
<td>5</td>
</tr>
</tbody>
</table>

The specimens were double notched. After creep testing, only one notch was ruptured. Creep cavitation investigation was carried out using light optical microscope on both ruptured and unruptured notches, representing rupture state and near rupture state, respectively. Samples were taken from longitudinally sectioned creep specimens. The samples were mounted, grinded, polished to 0.25 µm and etched in a solution containing 4 g CrO3, 0.75 g HN4Cl, 5 ml H2SO4, 5 ml HNO3 and 190 ml H2O.

3. Computation

3.1 Model for creep curve

The uniaxial model for the creep curve that is used in the FEM computations has been derived and analysed in several papers [16, 21, 22]. Only a brief summary will be given here. A typical creep curve shows three stages. The creep rate decreases during the primary stage, reaches a steady state value in secondary creep, accelerates during tertiary creep and terminates at rupture. For many materials the high initial creep rate is due to a low starting dislocations density and the accompanying low back stress. The work hardening gives rise to a decreasing creep rate. At the same time the recovery due to the annihilation of dislocations starts to become of importance. When recovery and work hardening balance, the stationary creep stage is reached.

From the description above, during creep two main processes take place, work hardening and recovery. To describe how the dislocation density develops during creep, the contributions from these two processes are added.

\[
\frac{d\rho}{d\varepsilon} = \frac{m}{bc_L} \rho^{1/2} - 2\tau_L M \rho^2 / \dot{\varepsilon} 
\]

(1)

The two terms in the right hand side of the equation represent work hardening and static recovery, respectively. In eq. (1), \(\rho\) is the dislocation density, \(\varepsilon\) the strain, \(m\) the Taylor factor, \(b\) Burger's vector and \(c_L\) is a constant. \(\tau_L\) is the dislocation line tension, \(\dot{\varepsilon}\) the stationary strain rate. \(M\) is the creep mobility, which takes both glide and climb into account. The derivation of eq. (1) can be found in several papers [16, 21,
In principle, there are two kinds of recovery, static and dynamic, which are time dependent and strain dependent, respectively. In a number of cases, it is important to take both dynamic and static recovery into account. For example, this is the case when describing tertiary creep or the influence of cold work [23-25]. The dynamic recovery will be ignored in the present analysis, and this is the case in most modelling work on creep.

The dislocation mobility is given by

\[ M_{OFP}(T, \sigma) = f_Q \frac{D_s b}{k_B T} e^{\frac{(\sigma - \sigma_{\text{break}}) b^3}{k_B T}} e^{-\frac{Q}{RT}} \left[ 1 - \left( \frac{\sigma - \sigma_{\text{break}}}{\sigma_{\text{max}}} \right)^2 \right] \]  

where \( T \) is the absolute temperature, \( \sigma \) the applied stress, \( D_s \) the pre-exponential coefficient for self-diffusion, and \( k_B \) Boltzmann’s constant, \( Q \) the activation energy for self-diffusion, \( R \) the gas constant and \( \sigma_{\text{max}} \) the max back stress which is taken as the tensile strength at room temperature. \( \sigma_{\text{break}} \) represents the stress that is needed for the dislocations to break away from the Cottrell atmospheres of phosphorus atoms. \( f_Q \) describes the influence of phosphorus on the activation energy of creep.

\[ f_Q = e^{-U_p^{\text{max}} / RT} \]  

\( U_p^{\text{max}} \) is the maximum interaction energy between a phosphorus solute and a dislocation [26]. The work hardening and static recovery balance during stationary creep, i.e. the two terms on the right hand side of eq. (1) are equal. Then we can obtain the stationary creep rate \( \dot{\varepsilon}_{\text{stat}} \)

\[ \dot{\varepsilon}_{\text{stat}} = \frac{2b \gamma L}{n m} M_{OFP}(T, \sigma) \sigma^{3/2} \]  

Taylor’s equation can be used to transfer the dislocation density to stress,

\[ \sigma = \sigma_y + m \alpha G b \rho \]  

where \( \alpha \) is a constant, \( G \) the shear modulus, and \( \sigma_y \) the yield strength. Combining eqs. (4) and (5), the stationary creep rate takes the following form

\[ \dot{\varepsilon}_{\text{stat}} = \frac{2b \gamma L}{n m} M_{OFP}(T, \sigma) \left( \frac{\sigma}{\alpha n G b} \right)^3 e^{-U_p^{\text{max}} / RT} = h(\sigma, T) \]  

A model representing the tensile and compressive flow curves was derived in [15]

\[ \sigma = \sigma_{\gamma}(T, \dot{\varepsilon}) + K(1 - e^{-\alpha \dot{\varepsilon}^2}) \]  

where \( K = \sigma_{\text{max}} - \sigma_{\gamma}(T, \dot{\varepsilon}) \). \( \sigma_{\text{max}} \) is the maximum stress in the flow curves. Eqs. (6) and (7) represent tests at constant load and at constant strain rate, respectively. As marked in the formula, the yield strength is temperature and strain rate dependent. The maximum stress in the flow curve is related to the stationary creep stress [15]. Eqs. (6) and (7) can be combined to derive an expression for strain versus time curves during primary creep. By extracting \( \sigma_{\text{max}} \) from eq. (7) and insert the expression into eq. (6), we find an
equation that covers both primary and secondary creep. Full details of this derivation can be found in Ref [15].

\[
\frac{d\epsilon_c}{dt} = h(\sigma_y(T, \dot{\epsilon}) + \frac{\sigma - \sigma_y(T, \dot{\epsilon})}{1 - e^{-\omega (\epsilon/2)}}, T)
\]

Eq. (6) for the secondary creep rate is recovered when the strain is large, and the denominator becomes unity. During primary creep the strain is small, and consequently the strain rate calculated from eq. (8) becomes large. In this way, the large strain rates in primary creep can be described. Using Odqvist's equation [27], eq. (8) can be transferred to multiaxial stress states

\[
\frac{d\epsilon_c^{\text{eff}}}{dt} = \frac{3}{2} \frac{d\epsilon_c^{\text{eff}}}{dt} \frac{\sigma'}{\sigma_{\text{eff}}}
\]

where \(\epsilon_c\) is the creep strain tensor, \(\sigma_{\text{eff}}\) the effective (von Mises) stress and \(\sigma'\) the stress deviator tensor. The effective creep strain rate is given by

\[
\frac{d\epsilon_c^{\text{eff}}}{dt} = h(\sigma_y(T, \dot{\epsilon}^{\text{eff}}) + \frac{\sigma_{\text{eff}} - \sigma_y(T, \dot{\epsilon}^{\text{eff}})}{1 - e^{-\omega (\epsilon^{\text{eff}}/2)}}, T)
\]

It has been demonstrated that the creep model in eq. (8) can reproduce the creep curves for annealed Cu-OFP quite well. For annealed Cu-OFP, the yield strength is about 35 MPa. However, for material that is delivered as forged and has not been annealed the yield strength is about 70 MPa. The difference in yield strength between annealed and as delivered (not annealed) material reduces the strain on loading. In this way an initial strain compensation should be added when describing creep for material that is not annealed.

\[
\frac{d\epsilon_c}{dt} = h(\sigma_y(T, \dot{\epsilon}) + \frac{\sigma - \sigma_y(T, \dot{\epsilon})}{1 - e^{-\omega (\epsilon + \Delta\epsilon_{\text{init}})/2}}, T)
\]

where

\[
\Delta\epsilon_{\text{init}} = \epsilon_{\text{init,anneal}} - \epsilon_{\text{init,del}}
\]

\(\Delta\epsilon_{\text{init}}\) is the contribution due to the slight cold work (= 3%) in the forged condition. The difference in loading strain between the annealed and the as delivered state can then be found by inserting the appropriate values in eq. (13).

\[
\epsilon_{\text{init}} = -\frac{2}{\omega} \log(1 - \frac{\sigma - \sigma_y(\dot{\epsilon}, T)}{K(\dot{\epsilon}, T)})
\]

Eq. (11) is used as constitutive equation in the finite element computation. For multiaxial cases the applied stress \(\sigma\) in eq. (13) is replaced by the reference stress. The reference stress was obtained by calculating the collapse load using a perfect plastic model. Details are given in section 3.2.

Constants used in the model are listed in Table 2.
Table 2 Constants used in computations

<table>
<thead>
<tr>
<th>Parameter description</th>
<th>Parameter</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Work hardening constant</td>
<td>$c_L$</td>
<td>30</td>
<td>[16]</td>
</tr>
<tr>
<td>Coefficient for self-diffusion</td>
<td>$D_0$</td>
<td>$1.31 \times 10^{-5}$ m²/s</td>
<td>[28]</td>
</tr>
<tr>
<td>Activation energy for self-diffusion</td>
<td>$Q$</td>
<td>198000 J/mol</td>
<td>[28]</td>
</tr>
<tr>
<td>Burgers’ vector</td>
<td>$b$</td>
<td>$2.56 \times 10^{-10}$ m</td>
<td></td>
</tr>
<tr>
<td>Taylor factor</td>
<td>$m$</td>
<td>3.06</td>
<td></td>
</tr>
<tr>
<td>Dynamic recovery constant</td>
<td>$\rho$</td>
<td>14.7</td>
<td>[15]</td>
</tr>
<tr>
<td>Poisson’s ratio</td>
<td>$\nu$</td>
<td>0.308</td>
<td>[29]</td>
</tr>
<tr>
<td>Constant</td>
<td>$\alpha$</td>
<td>$(1-\nu/2)/2\pi(1-\nu)=0.19$</td>
<td>[30, 31]</td>
</tr>
<tr>
<td>Max back stress</td>
<td>$\sigma_{\text{max}}$</td>
<td>257 MPa</td>
<td>[32]</td>
</tr>
<tr>
<td>Dislocation line tension</td>
<td>$\tau_L$</td>
<td>$Gb^2/2=7.94 \times 10^{-16}$ MN</td>
<td>[33]</td>
</tr>
<tr>
<td>Boltzmann’s constant</td>
<td>$k_B$</td>
<td>$1.381 \times 10^{-23}$ J/grad</td>
<td></td>
</tr>
<tr>
<td>Shear modulus</td>
<td>$G$</td>
<td>$G = 45400(1-7.1 \times 10^{-4} (T - 20))$ MPa, $T$ in K</td>
<td>[34]</td>
</tr>
<tr>
<td>Maximum interaction energy between a P solute and a dislocation</td>
<td>$U_P^{\text{max}}$</td>
<td>8220 J/mol</td>
<td>[35]</td>
</tr>
</tbody>
</table>

3.2 Finite element implementation

The FEM computations were performed using the commercial software Comsol Multiphysics 5.2a. A two dimensional time dependent creep model was set up. The constitutive equation eq. (11) was implemented in the analysis to describe the creep rate.

As mentioned above, the multiaxial tests were performed subjecting the notched specimens to an axial load. In this way the test specimen exhibits 2D axial symmetry as well as a symmetry plane in the middle of the specimen. It is therefore possible to reduce the model geometry to a rectangle with width equal to the specimen radius and height equal to half of the specimen length with only one notch. The geometry of the notches with the FEM mesh is shown in Figure 3. The mesh size was selected to be “physics-controlled mesh” with “extra fine size”. The minimum mesh size was $2 \times 10^{-6}$ m. The number of degrees of freedom in the computations ranged from 40,000 to 60,000 for different cases.
To determine the reference stress, FEM analysis was performed with perfect plasticity conditions [36]. The load was first raised until a homogeneous band with the von Mises stress was established. Then, the stress was further raised until the specimen could not take any more load. The maximum von Mises stress in the band that the specimen could hold was taken as the reference stress.

4. Results and discussion

4.1 Experimental results

4.1.1 Creep curves

In total, 17 creep tests under multiaxial stress state were performed. Experimental creep curves (creep strain versus time) are given in Figure 4, 5 and 6. Uniaxial creep curves are shown in Figure 7 for the same batch of material. Creep strain was measured from the deformation within gauge length, including elastic strain. Most tests went to rupture, while others were interrupted due to unforeseen long test duration. The interrupted tests are marked with arrow in the figures. For acuity 0.5, the tests with net section stress of 170MPa and 180MPa were interrupted after 17000 hours and 12000 hours, respectively. The creep strain was still as low as 2%, indicating that a much longer test duration would have been needed to reach rupture. Although the tests were performed at 75 ºC (only 0.25 of the melting point), the creep curves for the ruptured tests showed distinguished primary, secondary and tertiary stages as is observed at higher temperatures. For all interrupted tests, primary and well developed secondary stages were found.

For all acuities, increased net section stress resulted in shorter rupture life. The notch acuity also affects the rupture strain. Under a given stress level, the rupture strain decreased significantly with increasing notch acuity.
Figure 4 Creep strain versus time curves for notch acuity 0.5. Interrupted tests were marked with arrow.

Figure 5 Creep strain versus time curves for notch acuity 2. Interrupted tests were marked with arrow.
Figure 6 Creep strain versus time curves for notch acuity 5. Interrupted tests were marked with arrow.

Figure 7 Comparison of experimental creep strain versus time curves with the model for the uniaxial tests, eq. (8).

The creep lifetime as a function of the net section stress is given in a semi-logarithmic plot in Figure 8. The rupture time for the uniaxial specimens is included for reference. The interrupted tests are marked with rectangular boxes. Two of these points actually indicate the result for three specimens. The creep lifetime under multiaxial stress state was longer than under uniaxial stress state at the same net section stress. For a given net section stress, the rupture time also increased with notch acuity. This phenomenon indicates notch strengthening of Cu-OFP. The strengthening effect increases with increasing acuity (increasing notch sharpness).
Figure 8 Creep rupture time as a function of net section stress. Interrupted tests are indicated by rectangular boxes. The curves are fitted to data for uniaxial tests and for tests with acuity 0.5 and 2.

4.1.2 Post-test metallography

The microstructure of both ruptured and unruptured notches of the creep specimens were examined. Very few well separated cavities were found (Figure 9). This is consistent with experimental and computational results on creep ductility and creep crack growth [37, 38]. In the latter case, the stress state is clearly multiaxial. At the investigated temperature, the modelled cavitated area fraction is lower than 0.5% [25].
4.2 Model results and discussion

4.2.1 Rupture time

The reference stress can be used to estimate the rupture life of specimens under multiaxial stress state [13, 39]. The modelled reference stresses of ruptured tests as a function of the observed rupture lifetime are shown in Figure 10. A line was fitted for the linear relationship between uniaxial stress and logarithmic rupture time. The reference stresses for specimens with acuity 2 and 5 were found to fully agree with the uniaxial data. For the acuity 0.5, the reference stresses suggest a longer life time than the one actually observed.
Reference stress as a function of rupture time for ruptured creep tests. The curve is fitted to uniaxial test data.

4.2.2 Creep curves

A comparison between experimental creep strain vs time curves for the specimens and FEM results is illustrated in Figure 11. The strains for the FEM curves are the sum of elastic and creep strain in the same way as for the experimental curves. The elastic strain contribution is quite small (lower than 0.001). The plasticity effect that is pronounced in the primary stage is included in the model. It should be noticed that the model used in FEM computation was based only on physical mechanisms and no fitting parameters are involved. In the model, tertiary creep is not taken into account. A fundamental model for tertiary creep has been developed that can describe uniaxial creep curves [25]. However, this model requires that the dislocation densities in the substructure are represented. This is complicated in FEM analysis and for that reason it has not been considered in the present paper. The use of the model in eq. (8) in the uniaxial case is illustrated in Figure 7. A satisfactory representation of primary and secondary creep is obtained.

The model curves for the notched specimens in Figure 11 show a distinct primary stage where the plastic part is an important component. There is a gradual transition from the primary to the secondary stage. That this can be expected is evident from Eq. (11).

A main difficulty in predicting creep curves is the size of the initial plastic deformation. This can be seen in Figure 11d. The reason for this discrepancy is not fully understood, but it is clear that the loading conditions during the start of a creep test are less well defined than in a tensile test. It has been observed for other creep tests that the scatter of the strain for copper at identical test conditions can be as large as a factor of 1.5 [15]. Although discrepancies exist, the comparisons of FEM results and experimental results demonstrate that the main features can be reproduced.
Figure 11 Comparison between experimental results and FEM computation results a) Acuity 0.5, 195 MPa b) Acuity 0.5, 200 MPa c) Acuity 2, 215 MPa d) Acuity 5, 215 MPa.

4.2.3 Stress state

FEM analysis of the stress distribution along the specimen axis was carried out. During the initial loading stage, the specimens underwent elastic-plastic deformation and then it was continuously deformed by creep. Figure 12 shows the 3D von Mises stress distributions at the initial loading and at interruption time for the specimen with an acuity 5 and net section stress 170 MPa. From Figure 12a, a notable stress concentration with a von Mises stress of 173 MPa can be seen at the notch root. During creep deformation, the stress redistributes and approaches a steady state. The stress increases at the centre of the circular notch during creep and decreases at the notch root simultaneously. Figure 12b shows the stress distribution at the time when the test was interrupted. The maximum von Mises stress is much lower than the applied net section stress and in close agreement with the reference stress, which is 128 MPa for this case. The stress redistribution is a quite fast process. Figure 13 shows the stress redistribution at the notch root as a function of normalized creep time for the case in Figure 12. The stress redistributes quickly to a steady value below the applied net section stress. In ductile materials, like copper and ferritic steels, the stress redistribution can be much larger than in brittle materials since the creep strain at failure is larger. [9, 19]. In brittle
materials, the creep damage development is faster, and failure can occur before extensive stress redistribution has taken place [9, 40, 41].

![Figure 12](image1.png)

Figure 12 3D von Mises stress distribution for specimen with notch Acuity 5, 170 MPa at a) initial loading and b) time of interruption.

![Figure 13](image2.png)

Figure 13 Redistribution of von Mises stress at notch root as a function of normalized creep time (time/time at interruption) for the case in Figure 12.

Figure 14 illustrates the final stress distribution for specimens with the three notch acuities (0.5, 2, 5) under the same net section stress (170 MPa). For the specimen with acuity 0.5, the maximum von Mises stress lies in the centre of the specimen, see Figure 14a. On the other hand for the specimens with acuity of 2 and 5, the maximum von Mises stresses are approximately located at a sphere around the centre of the specimen, see Figure 14c and e. The maximum von Mises stress values are close to the reference stresses, which are 143, 130, and 128 MPa for these three cases. The stresses in the notch centres for acuity 2 and 5 are lower than the reference stresses. This can be seen more clearly in Figure 14b, d and f which show the
stress distribution along the specimen axis. For acuity 0.5, the stress at notch centre has a fairly wide peak. For the acuity 2 specimen, the notch centre was subjected to lower stress. For acuity 5, the notch centre stress was even lower, and was almost equal to the stress outside the notch. Further, the volume of specimen subjected to maximum load was smaller for higher notch acuity specimens, which can be found from the 3D von Mises stress distribution in Figure 14 a, c and e. The combination of lower stress and smaller volume result in notch strengthening. The notch strengthening phenomenon has been observed for several other kinds of materials, such as nickel-based alloys [12], 9Cr-1Mo steel [13] and 2.25Cr-1Mo steel [10].

The notch with acuity 0.5 is often called a “C” shape notch. Acuity 2 and 5 are “U” shape notches. From the notch stress distribution shown in Figure 14, it can be seen that the locations of stress concentration are different in the specimens with these two shapes of notches. The location of stress concentration is the most likely place where the creep damage initiates. For “C” shape notch, the maximum stress lies in the notch centre. For “U” shape notch, the maximum stress is seen at the notch root. This agrees well with the observations in Ref [42], in which the creep damage initiated from the notch centre for “C” shape notches whilst from notch root for “U” shape notches.
Figure 14 3D and axial stress distribution a, b) Acuity 0.5, 170MPa c, d) Acuity 2, 170MPa e, f) Acuity 5, 170 MPa.

Figure 15 shows the variation of the von Mises stress across the notch plane for different notch acuities at the same net section stress 200MPa. The von Mises stress increases with decreasing of notch
acuity. For a blunt notch, acuity 0.5, the stress is the highest and is constant across the notch. The von Mises stresses at the sharper notches are much lower and increase gradually towards notch root surfaces. The effective strain along notch plane shows similar trends (Figure 16). The effective strain is more than 40% at the notch plane for notch acuity 0.5, which is much higher than the observed total strain (16%), indicating that most deformation has occurred across the notch. For acuity 2 and 5, the effective strain at the notch root is also higher than at the notch centre.

Figure 15 Von Mises stress distribution across the notch with a net section stress of 200 MPa for the notch acuities 0.5, 2 and 5.

Figure 16 Effective strain distribution across notch for a net section stress of 200 MPa for the notch acuities 0.5, 2 and 5.
4.2.4 Triaxiality

The triaxiality of the stress state is known to greatly influence the plastic strain. Stress triaxiality can be defined as the ratio between the hydrostatic stress, $\sigma_m$ (average of principal stresses) and von Mises effective stress, $\sigma_{eff}$. The stress triaxiality distribution for different notch geometries is given in Figure 17. The maximum triaxiality lies at the specimen centre and is strongly dependent on the notch geometry. With the increase of notch acuity, the maximum triaxiality increases. Similar trends have been identified by other investigators [43, 44]. It is independent of the applied stress and depends only on the notch geometry. The triaxiality ratio at the notch centre for acuity 2 and 5 are around 0.6 and 0.82, respectively. The values are very close to the calculation in Refs [4, 45] for the same notch geometry.

Figure 17 2D stress triaxiality distribution for a) Acuity 0.5, b) Acuity 2, c) Acuity 5.

The tests results showed that creep rupture strain of sharper notches were much lower than blunt notch specimen. It is known that the triaxiality of the stress state greatly affects the plastic strain. Increased stress triaxiality resulted in a reduction in ductility [46, 47]. The predicted stress triaxiality is increasing with notch acuity (Figure 17). The high stress triaxiality lowered the deformation rate of sharp notch specimen, resulting in lower strain. Predicted stress triaxiality from FEM computation has been compared with
Bridgeman’s formula [48], which gave much higher values. The lack of precision in Bridgeman’s formula has been previously mentioned in literature [46].

5. Conclusions

In the present work, creep tests under multiaxial stress state were conducted on phosphorus alloyed oxygen free copper (Cu-OFP) at 75°C. Double notched specimens with three different notch geometries were used. The deformation was continuously recorded. Most of the tests were run until rupture with rupture times up to 15000 h.

- Cu-OFP was found to show notch strengthening. At a given net section stress, the multiaxial tests showed longer lifetime than the uniaxial tests. The creep life increased with notch acuity (sharper notch) at a given net section stress.
- All the rupture tests demonstrated a ductile creep rupture. The unbroken notch on each specimen was investigated metallographically. Only a few cavities were observed, which is consistent with the ductile rupture.
- Stress analysis was performed with the help of FEM. Using reference stress values determined from perfect plastic models, the rupture life could be satisfactory predicted for the higher acuities.
- The creep deformation was computed using a basic creep model. A reasonable agreement with observations was obtained although with considerable scatter.
- Stress redistribution was found during creep deformation. At the initial loading, the von Mises stress at the notch root was about the same value as the applied net section stress. After stress redistribution, the maximum stress was lower than the net section stress and in close agreement with the reference stress.
- For the shallow notch, the maximum von Mises stress was at the notch centre. For the sharp notches, the maximum von Mises stress was found at the notch root.
- The stress triaxiality increases with notch acuity, attributing to the lower ductility of sharper notch specimens.
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References

Paper III
Basic modelling of tertiary creep of copper

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ABSTRACT
Mechanisms that are associated with acceleration of the creep rate in the tertiary stage such as microstructure degradation, cavitation, necking instability and recovery have been known for a long time. Numerous empirical models for tertiary creep exist in the literature, not least to describe the development of creep damage, which is vital for understanding creep rupture. Unfortunately, these models almost invariably involve parameters that are not accurately known and have to be fitted to experimental data. Basic models that take all the relevant mechanisms into account which makes them predictive have been missing. Only recently, quantitative basic models have been developed for the recovery of the dislocation structure during tertiary creep and for the formation and growth of creep cavities. These models are employed in the present paper to compute the creep strain versus time curves for copper including tertiary creep without the use of any adjustable parameters. A satisfactory representation of observed tertiary creep has been achieved. In addition, the role of necking is analysed with both uniaxial and multiaxial methods.

Introduction
Creep deformation is usually measured by exposing tensile specimens to a constant load or sometimes to a constant stress and recording the elongation of the specimens as a function of time. The result is given as the creep strain versus time, referred to as a creep curve. A creep curve is in most cases characterised by three stages: primary, secondary and tertiary. Creep deformation is induced by the generation, motion and annihilation of dislocations. The creep rate decreases during primary stage, reaches a steady value in secondary creep, accelerates during tertiary creep and terminates at rupture. For many materials, the high initial creep rate is due to a low starting dislocation density. Due to work hardening, new dislocations are generated and the dislocation density increases, leading to a decrease in the creep rate. At the same time, the recovery due to the annihilation of dislocations starts to become of importance. When achieving a balance between recovery and work hardening, the strain rate is approximately constant and the secondary creep stage is reached. It is also referred to as stationary creep. During tertiary creep, a modification of the microstructure occurs, leading to acceleration of the creep rate.

The scientific literature has to a large extent been focusing on the secondary stage due to its direct...
relation to the operating deformation mechanisms. Much less attention has been paid to primary creep and tertiary creep. Technically, both primary creep and tertiary creep are of utmost importance. For fcc alloys that cover a large fraction of the technically used creep-resistant materials, the creep strain in the primary stage is often of the same order as that in the secondary stage. Tertiary creep is also of major technical significance since it controls creep rupture. The increase in the creep rate in the tertiary stage due to changes in the microstructure is referred to as the formation of creep damage [1–3]. There are a number of creep damage mechanisms including particle coarsening, subgrain growth, cavitation and recovery of the dislocation structure, which can all accelerate the creep rate during tertiary creep. In addition, the creep rate is influenced by the necking instability. Failure induced by microstructure degradation has been commonly observed in creep-resistant martensitic steels, which have a complex microstructure. During long-term creep, fine carbonitrides (e.g. M_2X and MX) coarsen and dissolve and new brittle phases (e.g. Z-phase, Laves phase, M_4X carbides) are formed. The absence of fine particles reduces the creep strength. In addition, the new coarser phases can serve as sites for crack nucleation that lowers the creep strength further [4–6].

As a consequence, modelling of creep damage has almost invariably disregarded some important mechanisms and compensated this by using adjustable parameters. It was demonstrated in [14] that by just involving two adjustable parameters, a wide range of creep curves in the tertiary stage could be represented. There are good empirical models for describing tertiary creep. In particular, what is now usually referred to as the omega model can be mentioned [15–17]. However, with two adjustable parameters, a good empirical model is not essential. Many mathematical expressions can be used [14]. This implies that creep damage models with two or more adjustable parameters are not predictive and cannot be used to identify the operating mechanisms. It is therefore of vital importance that basic equations without adjustable parameters are employed.

Oxygen-free copper alloyed with 50 ppm phosphorus (Cu-OFP) has been selected as canister material for storing spent nuclear fuel in Sweden due to its excellent corrosion resistance and high ductility [18]. During storage, the spent nuclear fuel will release heat while decaying, increasing the temperature up to 100 °C in the canister. Both hydrostatic pressure and swelling pressure from clay will impact the copper canisters, which will be exposed to creep as a consequence. The copper canisters are expected to stay intact for 100000 years. In order to predict the creep damage under such long times, it is critical to use fundamental models based only on physical phenomena [19]. Cu-OFP is the material that will be investigated in the present paper.

The creep mechanisms at low temperatures (below 0.3 Tm, melting point) can be quite different from those at high temperatures. For many materials, logarithmic creep form is more appropriate than power law creep to describe the deformation behaviour. This applies, for example, to austenitic stainless steels, where creep never leaves the primary stage [20, 21]. However, for copper, this is not the case. A large number of creep tests have been performed for Cu-OFP at 75 °C (0.25 Tm), and the creep curves are quite similar to those achieved at high temperatures, where a well-developed and long-duration steady state is observed [14]. The mechanisms for low-temperature creep are not yet fully established. It has been suggested that low-temperature creep is controlled by glide and cross slip [22]. Dislocation climb was not considered to be active due to the low estimated climb rate at low temperatures. However, if the increase in the climb rate from the increase in vacancy concentration due to plastic deformation is taken into account, the observed creep rates at ambient temperatures for aluminium and copper can be accurately accounted for [23]. If climb is the operating mechanism, the observed extended secondary stage can be explained directly.

In Cu-OFP, changes in the dislocation structure could provide microstructure degradation. Accelerated recovery and an associated decrease in dislocation density as main creep damage mechanism were reported from experimental results [5, 24] and computation [25]. The nucleation of cavities followed by growth and interlinkage is believed to play an important role in creep failure of metals [9, 10].
Necking is known as a macroscale deformation inhomogeneity. When the material is plastically unstable, even small defects can promote localised deformation [26]. At some point during creep testing, strain localised in a small region takes place and necking appears. Studies on the effect of an initial defect on creep deformation have been carried out [27–30].

Dislocation recovery mechanism has been used to describe the three stages of creep deformation. Fundamental dislocation models based on this mechanism for primary creep and secondary creep were formulated [14]. It has been demonstrated that it can be used to describe the primary creep and secondary creep of Cu-OFP and also slow strain rate tensile tests under both uniaxial and multiaxial stress states [31, 32]. The purpose of the present paper is to model tertiary creep of Cu-OFP taking the relevant microstructure processes into account without involving adjustable parameters. The basis is a model that takes substructure development during creep into account. It was derived originally for cold-worked materials and will be employed to simulate accelerated recovery [13]. In order to evaluate the effect of necking on tertiary creep of Cu-OFP, a small imperfection is artificially introduced to the specimen in computation according to the method proposed by Burke and Nix [33]. Influence of cavitation is also considered when describing tertiary creep. The modelled results will be compared with experimental data for Cu-OFP.

**Model**

**Accelerated recovery model**

A dislocation model was developed in [14, 32] that could describe primary creep and secondary creep of copper. Some parts in the model were taken from the literature for granted but have been precisely derived recently [34]. It is believed that the dislocation model is general. Its validity has been demonstrated also for austenitic stainless steels [35] and for aluminium alloys [23]. By applying the model, it has been shown that the recovery during tertiary creep can be analysed by taking the role of the substructure into account [12, 13].

In copper and many other materials, a cell structure is formed during deformation. Already after 10% strain, the majority of the dislocations can be found in the cell boundaries [36], and after 20% strain, virtually all dislocations are in the cell boundaries [37]. In the model, only the dislocations in the cell walls are taken into account to avoid an excessive number of parameters. This assumption is also consistent with X-ray measurements done by Straub et al. [38], where the strength contribution from the cell interior is less than 10 MPa for pure copper.

The dislocations in the cell walls are divided into two sets, balanced and unbalanced. This assumption is natural based on the experimental evidence that dislocations in cell walls can be statistically distributed and polarised [39]. In [13], it was proposed that an important content the cell walls is also dislocation locks. This is in agreement with the modelling of strain hardening by Argon [40]. The dislocation locks are primarily Lomer–Cottrell locks, which are pairwise sessile dislocation segments of extended dislocations [41]. When the two sets of dislocation partials slip and meet each other, they form the dislocation locks. The Lomer–Cottrell locks have been frequently observed and are believed to play an important role during strain hardening of fcc alloys [40]. The distinction between balanced and unbalanced dislocations is vital for several properties as will be explained below. In the former set, for any dislocation on a given slip system, a dislocation with the opposite Burgers vector can always be found. The dislocations appear in pairs with opposite Burgers vectors, not necessarily close to each other. In simplified terminology, the numbers of dislocations of opposite signs are balanced. In the latter set, the dislocations are polarised and dislocations of opposite sign are missing. The unbalanced dislocations cannot annihilate each other since dislocations with the opposite Burgers vector are missing. Figure 1 shows a sketch of the formation mechanism for unbalanced dislocations. In a stress-free condition, the dislocations are randomly distributed in the cell interior (Fig. 1a). In the presence of a stress, the dislocations with opposite signs tend to move in different directions. Many dislocations end up on different sides of the cell and form a polarised set around the cell walls (Fig. 1b). These are the unbalanced dislocations. The remainder of the dislocations are considered as balanced. Some of dislocations form cell walls. A number of the dislocations move through the walls [13]. The polarised dislocations with marked sign in Fig. 1b are unbalanced...
dislocations. The ones that form dislocation locks in cell walls are balanced dislocations.

The balanced and unbalanced dislocation densities satisfy the following equations [12, 13]

\[
\frac{\mathrm{d} \rho_{\mathrm{bnd}}}{\mathrm{d} \varepsilon} = k_{\mathrm{bnd}} \frac{m \rho_{\mathrm{bnd}}^{1/2}}{b \gamma L} - \omega \rho_{\mathrm{bnd}} - 2 \tau L M \rho_{\mathrm{bnd}}^2 / \dot{\varepsilon},
\]

(1)

\[
\frac{\mathrm{d} \rho_{\mathrm{bnde}}}{\mathrm{d} \varepsilon} = k_{\mathrm{bnde}} \frac{m \rho_{\mathrm{bnde}}^{1/2}}{b \gamma L} - \omega \rho_{\mathrm{bnde}},
\]

(2)

\( \rho_{\mathrm{bnd}} \) and \( \rho_{\mathrm{bnde}} \) are the balanced and unbalanced dislocation densities in the cell walls, which are defined as the total length of the dislocations divided by the cell volume. \( \dot{\varepsilon} \) is the strain, \( m \) the Taylor factor, \( b \) Burger’s vector, \( \gamma L, k_{\mathrm{bnd}} \) and \( k_{\mathrm{bnde}} \) are work hardening constants, \( \omega \) a dynamic recovery constant, \( \tau L \) the dislocation line tension, \( \dot{\varepsilon} \) the strain rate and \( M \) the creep mobility. All the parameter derivations can be found in Refs. [13, 34, 42]. Parameter values are in the “Appendix”. The three terms on the right-hand side of Eq. (1) represent work hardening, dynamic recovery and static recovery. It is essential to take both dynamic recovery and static recovery into account when describing tertiary creep as will be evident below. Dynamic recovery is strain dependent while static recovery is time dependent [43]. Dynamic recovery will occur as long as straining takes place [44]. Static recovery describes how dislocations of opposite sign attracted each other and eventually annihilate. Dynamic recovery takes place by the rearrangement of dislocation into lower energy configurations [45]. In the spurt events during plastic straining, the dislocations typically pass through two or more cell boundaries [40, 46]. Argon suggests that when dislocations through the cell boundaries, they remove a certain fraction of the dislocation locks, which gives rise to the dynamic recovery effect [40].

Equation (1) has the same format as the basic model for homogeneous dislocations [32]. Since unbalanced dislocations cannot combine with dislocations of opposite Burgers vector, they are not exposed to static recovery. This is the reason for the absence of the static recovery term in Eq. (2). Both unbalanced and balanced dislocations are subjected to dynamic recovery.

A back stress is introduced to model the creep curves. In a number of publications in the past, a back stress has been considered as an intrinsic property that could be measured, for example, in a stress change experiment. With the help of dislocation dynamics simulations, the back stress from dislocations can be computed directly. It turns out that computed back stress is almost identical to the applied stress. This is also the case after a stress drop test. The change in the back stress takes place in less than 1 ms. The difference between the applied stress and the back stress is less than 1/500 of the applied stress [47]. Thus, an intrinsic back stress is not very meaningful to use in modelling, which has been realised by a number of authors, see, for example [48]. Although the back stress cannot be measured, it can be introduced if it is properly defined.

A back stress is introduced in our model as the extra hardening from the unbalanced dislocations in the cell walls [12]. This stress compensates for the sharp increase in true applied stress \( \sigma_{\text{app}} = \sigma_{\text{appl0}} \varepsilon^{\gamma} \) during secondary creep, where \( \sigma_{\text{appl0}} \) is the applied
nominal stress. Only creep testing under constant load is considered here, since all our creep data are for that case. It has been suggested in the literature that testing under constant stress is necessary to achieve a pronounced secondary stage, but that is clearly at variance with our experimental data, see below. It should be recalled that the stress exponent is quite high for creep at lower temperatures, often above 50 [14]. The magnitude of the back stress equals the dislocation stress minus the nominal stress. Only creep testing under constant stress is necessary to achieve a pronounced secondary stage, but that is not that case. It has been suggested in the literature that testing under constant stress is necessary to achieve a pronounced secondary stage, but that is clearly at variance with our experimental data, see below. It should be recalled that the stress exponent

\[ \sigma_{\text{back}} = \sigma_{\text{disl}} - \sigma_{\text{appl0}}, \]

where \( \sigma \) is a constant in Taylor’s equation, and \( G \) is the shear modulus. The effective creep stress is the true applied stress minus the accompanying back stress

\[ \sigma_{\text{creep}} = \sigma_{\text{appl}} - \sigma_{\text{back}} \quad (4) \]

From Eq. (1), an expression for the stationary creep rate can be obtained. In this expression, the creep stress in Eq. (4) should be applied

\[ \dot{\varepsilon}_{\text{sec}} = 2\tau_L M(T, \sigma_{\text{appl}} - \sigma_{\text{back}}) \rho_{\text{bnd}}^2 \left( k_{\text{bnd}} m \rho_{\text{bnd}}^{1/2} \rho_{\text{bnd}} - \omega \rho_{\text{bnd}} \right) \quad (5) \]

If we insert the expression for the back stress into Eq. (4), we find that

\[ \sigma_{\text{creep}} = \sigma_{\text{appl}} + \sigma_{\text{appl0}} - \sigma_{\text{disl}} \quad (6) \]

We now generalise Eq. (5) by assuming that it is not just valid for secondary creep but for the influence of the changes in the dislocation density on the whole creep, provided Eq. (6) is applied [13]

\[ \dot{\varepsilon} = 2\tau_L M(T, \sigma_{\text{appl}} + \sigma_{\text{appl0}} - \sigma_{\text{disl}}) \rho_{\text{bnd}}^{3/2} \left( k_{\text{bnd}} m \rho_{\text{bnd}}^{1/2} \rho_{\text{bnd}} - \omega \rho_{\text{bnd}}^{1/2} \right) \quad (7) \]

This approach suggests that if you know the stress dependence of the secondary creep rate, the strain dependence of the creep rate for the creep curve can be derived, provided the variation of the dislocation density is known.

How the different types of stresses change as a function of time is illustrated in Fig. 2 at the applied stress of 175 MPa. The three curves in Fig. 2 are calculated from the models described above. Firstly, by integrating the set of Eqs. (1), (2) and (7), creep strain \( \varepsilon \), balanced dislocation density \( \rho_{\text{bnd}} \) and unbalanced dislocation density \( \rho_{\text{bnd}} \), as a function of time can be computed. Then, the applied true stress is obtained by \( \sigma_{\text{appl}} = \sigma_{\text{appl0}} e^\dot{\varepsilon} \), where \( \sigma_{\text{appl0}} \) is 175 MPa in this case. The dislocation stress and creep stress are calculated according to Eqs. (3) and (6), respectively. Initially, the dislocation stress is quite low and the creep stress is high. During secondary creep, the applied true stress is balanced by the dislocation stress (their curves overlap each other during secondary creep as shown in Fig. 2). In the secondary stage, the creep stress is close to the nominal applied stress. In tertiary stage, the increase in the dislocation stress is lower than that of the true applied stress. So the creep stress increases, leading to an acceleration of the creep rate. Dynamic recovery also contributes to the acceleration of creep rate. This is evident from Eq. (7). Since the dislocation stress is increasing, so is the dislocation density. Thus, the dislocation density in the numerator in Eq. (7) increases and the denominator decreases, giving a further increase in the creep rate.

**Cavitation model**

Since creep cavitation gives rise to a loss of the load carrying cross section, it can give a contribution to tertiary creep. There are numerous models for nucleation and growth of creep cavities in the literature, but practically all of them involve parameters that are not accurately known and have to be fitted to the observations. Only recently, basic models for nucleation and growth of creep cavities have been formulated by He and Sandström [9, 10, 49]. For a
review, see [11]. The equations that are used to compute the amount of cavitation are listed and discussed here.

Grain boundary sliding (GBS) is believed to be essential for cavity nucleation. The GBS distance is approximately proportional to the creep strain. The ratio of the GBS displacement rate to the strain rate is a constant, denoted as $C_s$. The value of $C_s$ is approximately 50 μm for copper [50]. It is very well established that the number of nucleated cavities is proportional to the creep strain. Already Dyson [51] documented the validity of this relation for a range of materials. It is very difficult to explain the observed stress and strain dependences of cavity nucleation unless GBS is the dominating mechanism [9]. Experiments for GBS demonstrate that about the same value is obtained for Cu with different experimental techniques from 125 to 600 °C at different strain rates [50]. It demonstrates that $C_s$ is approximately constant over a wide range of conditions. Nucleation of cavities could take place at particles or at sub-boundaries. Sulphides and oxides can be found in the material. However, the total measured area fraction of particles is less than $1 \times 10^{-5}$, and consequently particles do not play any significant role in cavity nucleation. However, nucleation can take place at the sub-boundary junctions. This has been verified to be thermodynamically feasible [50]. According to the double ledge model, the nucleation rate of cavities is proportional to the creep strain rate, which is in full agreement with experimental data, for example, for austenitic stainless steels [9]. This gives strong support for cavity nucleation being controlled by GBS. The nucleation sites are the intersections of sub-boundaries with sub-boundary corners on the opposite side of a sliding grain boundary. Low angle and twin boundaries are cavity resistant. The cavity nucleation rate at intersections of sub-boundary/sub-boundary corner is [9]

$$ \frac{dn_{cav}}{dt} = \frac{0.9C_s}{d_{sub}^3} \dot{\varepsilon}, $$

where $n_{cav}$ is the number of cavities, $dn_{cav}/dt$ the cavity nucleation rate, $d_{sub}$ the subgrain size, and $\dot{\varepsilon}$ the creep strain rate.

When the nucleated cavity exceeds a critical size, it will start to grow. The growth of cavities is diffusion controlled by stress. Strain-controlled growth has also been considered in the literature, see, for example [52]. In the investigated cases, they give a lower growth rate than diffusion control, and they will not be discussed further. To fix the unphysical exaggerated growth rate predicted by traditional diffusion-controlled cavity growth models, Dyson [53] suggested that the cavity growth rate should not be larger than the creep rate, which is referred to as constrained growth. The constrained cavity growth rate is [54]

$$ \frac{dR_{cav}}{dt} = 2D_0K_f(\sigma_{red} - \sigma_0) \frac{1}{R_{cav}^2}, $$

where $R_{cav}$ is the cavity radius, $dR_{cav}/dt$ the cavity growth rate, $\sigma_0$ the sinter stress, $D_0$ a grain boundary diffusion parameter, $K_f$ a factor related to the cavitated area fraction $A_{cav}$. The parameters values can be found in the “Appendix”. $\sigma_{red}$ is the reduced stress, which can be determined by solving a differential equation [10]

$$ 2\pi D_0K_f(\sigma_{red} - \sigma_0)/L^2R_{cav} + \dot{\varepsilon}(\sigma_{red}) = \dot{\varepsilon}(\sigma_{appl}), $$

where $L$ is the cavity spacing. The cavitated area fraction on the grain boundaries can be expressed as [55]

$$ A_{cav} = \int_0^t \frac{dn_{cav}(t')}{dt'} \pi R_{cav}^2(t, t') dt', $$

where $R_{cav}(t, t')$ is the radius of the cavity at time $t$ that was formed at time $t'$. It is well established that when the cavitated area fraction exceeds 25%, rupture occurs [56]. In the investigated cases in the present paper, the modelled cavitated area fraction is as low as 0.3%. Its effect on tertiary creep can then be neglected.

**Necking model**

At sufficiently large strain during creep, a plastic instability takes place leading to the formation of a waist on the specimen that is usually referred to as necking. Necking is initiated by a geometrical imperfection or a material inhomogeneity in the specimen. Once the waist has been formed, its continuous growth does not depend on how it was initiated. In this analysis, the effect of a geometric defect on the creep deformation is studied.

A necking criterion was proposed by Hart for creep tests under constant load [57]. The onset point of instability can be expressed in terms of area fluctuation. When the variation in the area at a particular point is larger than zero, the deformation is stable. If
one applies the relation between area reduction and strain, the stability criterion can be expressed as
\[
\dot{\varepsilon} \leq \varepsilon^2,
\] (12)
where \( \dot{\varepsilon} \) is the strain rate and \( \varepsilon \) is strain acceleration, the second time derivative of the strain. Application of Eq. (12) to the experimental data was carried out for different test conditions to determine the onset of necking.

In a uniaxial model, Burke and Nix [33] analysed necking by considering the deformation response of an initially imperfect cylindrical bar. The initial cross-sectional area was assumed to vary along the \( x \)-axis in a smooth sinusoidal manner according to
\[
A(x,0) = A_0 - \frac{\Delta A}{2} \cos \frac{2\pi x}{L_i} ; \quad (0 \leq x \leq L_i),
\] (13)
where \( A_0 \) is the original cross-sectional area of specimen, and \( L_i \) the length of the specimen with the defect part. \( \Delta A \) represents the changes in initial cross-sectional area, which is chosen as \( \Delta A/A_0 = 0.01 \) in the current case. Accordingly, the maximum magnitude of imperfection is 1%. In terms of radius, thus a difference of 0.025 mm is introduced. By sectioning the specimen along its axis, the creep elongation in each section can be computed directly, since the load on each section is constant and equal to the applied load on the specimen. By adding the elongations of the sections, the creep strain of the specimen can be obtained directly. This average strain for the entire specimen can be used to compare with the experimental result.

Figure 3 shows how the initial strain and initial radius varied along \( x \)-axis. A symmetric nonuniformity is centred at position \( x = 0 \), varying along \( x \)-axis to \( x = 15 \) mm. At positions \( x > 15 \) mm, the initial radius is 5 mm and initial strain is zero. The gauge length of a tested specimen is 100 mm. So the initial nonuniformity only exists for 30% axially of the specimen.

When a waist is introduced, the stress state changes from uniaxial to multiaxial. Finite element (FEM) computations were performed to analyse the influence of multiaxiality. Due to limitation in the FEM software, only the secondary stage could be simulated. Severe necking was obtained. Experimental necking profiles were taken from ruptured creep specimens.

Results

Accelerated recovery results

By integrating the set of Eqs. (1), (2) and (7), creep strain versus time curves can be computed. All the parameters used in the model can be derived; none is used as adjustable parameter. The derivations of these parameters can be found in previous work Refs. [12–14, 32], and the values are summarised in “Appendix” with references given. The model has been applied to creep curves for Cu-OFP at 75°C. Experimental data are taken from Ref. [14]. A comparison of the modelled results with experimental curves is shown in Fig. 4. The three stages of creep curves (primary, secondary and tertiary) are present in the experiments. An extended secondary stage is present in spite of the high creep exponent. There are steps in the experimental curves due to the necessity of reloading the specimens when a certain strain was exceeded. Due to the large creep rupture elongation of Cu-OFP, during creep tests the lever arm had to be reset when a critical strain was reached. Otherwise, the lever arm would no longer be sufficiently

![Figure 3](image-url)
horizontal, and the dead weight might hit the floor. It is believed that during these temporary unloadings, the substructure is relaxed and that is the origin of the small new primary stage that is formed. No attempts have been made to reproduce these steps. The model can describe the three stages in a quite acceptable way. It is interesting to note that the model can describe the logarithmic decrease in the strain rate in the primary stage, which has been observed for a number of materials. This is analysed in [12].

There are some differences at the end of creep life, where the experimental strain increased sharper than the modelled strain. The modelled tertiary stage lasted for longer time and increased more smoothly. This difference will be analysed when the effect of necking is discussed below.

**Necking results**

Severe necking was observed on the Cu-OFP specimens after the creep tests, implying that the necking effect should be taken into account when modelling tertiary creep. An initial nonuniform cross-sectional area was introduced to investigate the necking effect on creep deformation of Cu-OFP.

Both uniaxial and multiaxial analyses have been performed. Figure 5 illustrates the results of the uniaxial analysis. In Fig. 5, modelled creep strain versus time curves are given at different distances from the necking centre. At necking centre, pronounced strain localisation was found and the final strain reached 1.8. The strain can be transformed into the reduction in cross-sectional area according to $\varepsilon = -\ln\left(\frac{A_0}{A}\right)$. When the strain is 1.8, the corresponding reduction in cross-sectional area is around 85%, which is consistent with the experimental measurements for Cu-OFP [14]. Since the curves overlap, a zoom-in view for the time interval from 980 h to the end of tests is shown in Fig. 5b. The final strain is inversely proportional to the distance from necking centre. At the position of 5 mm from necking centre, the final strain dropped to 0.31. This dramatic drop indicates that the strain is localised to a small area. At the positions of 15 mm and further from the necking centre, the uniform strain was 0.26.

The calculated average strain for the whole sample is used to compare with the experimental curves. The comparison is shown for different test conditions in Fig. 6. An improvement has been achieved for the
end of life in comparison with just the accelerated recovery results. The sharp increase in strain at the end of the tertiary stage can be modelled better by taking necking into account.

Figure 7 shows the strain distribution simulated by FEM at 75 °C with an applied stress of 175 MPa. The figure demonstrates that the deformation is uniform until the deformation becomes unstable and all further elongation takes place in the waist. A real specimen would fail very quickly under these conditions.

Hart’s criterion was applied to the experimental data to determine the onset of the unstable deformation. For all test conditions, the unstable deformation starts very close to the inflection point of the strain versus time curve. A plus marker is given in Fig. 6b indicating the necking starting point calculated by Hart’s criterion. The FEM modelling suggests that the
necking starts at a very late stage of creep, almost at the failure strain, resulting in a steep rise of the creep curve. The development of the neck is initially obviously quite a slow process.

Figure 9 shows a comparison between the experimental necking profile (15 mm along necking position) and the FEM results. The model reproduces the experimental radii within about 10%. The general behaviour is the same and the deviation between them is partially due to the necking elongation in the FEM modelling even after a well-developed neck has been formed. In the real test, the specimen was fractured. So adjacent to the necking position in Fig. 9, the deviation is larger.

Discussion

There is an extensive literature on the formation of creep damage and its influence on tertiary creep. As pointed out in the introduction, these models are almost invariably empirical. One model that has been used frequently is the one that Riedel presented in his book on creep fracture [58]. He derives the creep damage based on cavity formation. He assumes that the nucleation rate is proportional to the creep strain and that the volume growth rate is linear in time $t$. Also assuming that only secondary creep is of importance, he found that the area fraction of cavities is proportional to $t^{5/3}$. This covers the main development of the cavitation in a simple form. However, there are shortcomings. The nucleation rate constant is handled as an adjustable parameter. The effect of constrained growth and possible overlap between cavities is neglected, typically significantly overestimating the amount of cavitation. Today, there is no need to make these simplifications, since the additional effects can be taken into account without much computational effort [11].

The concept that dynamic recovery plays an important role during tertiary creep is relatively new but well established. Creep tests of 24% cold-worked Cu-OFP were performed at 75 °C [13]. For all the creep tests, the creep strain versus time curves were dominated by a continuously increasing strain rate, i.e. by tertiary creep. The creep curves could elegantly be reproduced by assuming that dynamic recovery according to a model similar to the one in the present paper was the dominating creep damage mechanisms. Very limited cavitation was observed. This was not surprising since the reduction in area at rupture was almost exactly 90% (89–91%).

For the specimens in the present investigation, the reduction in area at rupture was also very high (90–92%) indicating fully ductile rupture. This indicates that cavitation is of little importance for the failure. This was indeed confirmed by metallographic investigations and modelling. In both cases, the area fraction of cavities in the grain boundaries was less than 0.5%. This makes it natural to assume that dynamic recovery of the substructure is the dominating mechanism for tertiary creep until significant necking quickly develops at the very end of the creep life.

The effects of accelerated recovery, cavitation and necking on tertiary creep have been analysed in the present paper. Accelerated recovery gives the largest contribution to tertiary creep for the investigated alloys. In the model, two distinct sets of dislocations (balanced and unbalanced) are involved. The balanced dislocations are exposed to static recovery, and its density remains approximately constant during secondary creep. At the same time, the unbalanced dislocation density continuously increases and gives rise to a major back stress that matches the continuous increase in the true applied stress. At the end of the secondary stage, the true applied stress increases faster than the back stress. This means that the effective stress rises at the end of the secondary stage, resulting in the increase in the creep rate in the tertiary stage.

Basic models are now available for the nucleation and growth of creep cavities. With these models, the cavitated area fraction of grain boundaries and its influence on the creep curves can be predicted. For the investigated copper alloys, the predicted area
fraction at failure was less than 0.5%. Consequently, cavitation had no significant influence on tertiary creep.

Uniaxial and multiaxial models for necking have been considered. Hart’s criterion [57] indicates that an instability that would give rise to necking is formed directly at the end of secondary creep. This has also well known for other types of materials. For example, Lim et al. [59] found this for 9% Cr steels. The uniform strain calculated from uniaxial and multiaxial simulations is almost the same. The analyses suggest that significant necking only appears very close to the failure strain. This is also what is found in [59]. Necking simulations give a rapid increase in the strain near rupture in agreement with the observations. Figures 4a and 6b show a comparison of modelled and experimental creep curves for the same case. In both figures, primary creep and secondary creep can be well reproduced. Accelerated recovery exerts the influence on the entire tertiary stage while necking contributes to the very end of tertiary creep. The radii in the neck can be predicted quite well with the help of FEM computations. The model results lie within 10% of the observed values. In spite of the fact that Hart’s criterion gives an early start of the necking, the necking is not really developed until close to failure.

It is expected that the model for tertiary creep is also applicable to other fcc materials than copper. The basic dislocation model has been demonstrated to be valid also for austenitic stainless steels [35] and aluminium [60]. Due to lack of data, it has not been possible to verify that tertiary creep can be represented for these materials. However, the model cannot be used for martensitic 9 and 12% Cr steels. Tertiary creep is frequently studied in these materials, because of their extensive use in fossil-fired power plants. Tertiary creep in these materials typically show a linear increase in the creep rate with creep strain, see, for example [59]. This behaviour follows what is often referred to as the omega model [15, 17, 61]. Although this empirical model has been known for a long time. Empirical modelling of tertiary creep has been performed extensively for understanding the role of creep damage in creep rupture. In the present paper, basic models for tertiary creep are formulated for the relevant mechanisms for copper. Quantitative basic models for recovery during tertiary creep and for the nucleation and growth of creep cavities have only recently become available.

- The most important contribution to creep curves of copper comes from the dislocation structure. A dislocation model is presented that can be used to compute this contribution.
- To be able to describe the contribution from the dislocation structure to the both secondary creep and tertiary creep, there are three important requirements.
  - The dislocations in the cell walls must be taken into account.
  - Both balanced and unbalanced dislocations must be considered.
  - Both dynamic recovery and static recovery must be covered in the equations for the dislocation densities.
- Cavitation often plays an important role in tertiary creep. At the considered temperature 75°C, the cavitated area fraction is less than 0.5% and the contribution from cavitation can be ignored.
- The influence of necking on the creep curve has been analysed with uniaxial assumptions as well as with multiaxial methods. It turns that both approaches predict that pronounced necking does not take place until the failure strain has almost been reached. The uniaxial computations give a necking that is narrower than the observed ones. However, the multiaxial approach using FEM predicts a necking that is in good accordance with experiments with computed neck radii within about 10% of the observed values.

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**Compliance with ethical standards**

**Conflict of interest** The authors declare that they have no conflict of interest.

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**Appendix: Parameter values**

See Table 1.

**References**


<table>
<thead>
<tr>
<th>Table 1 Parameters used in this paper</th>
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<tbody>
<tr>
<td>Parameter description</td>
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<tr>
<td>--------------------------------------</td>
</tr>
<tr>
<td>Burgers’ vector</td>
</tr>
<tr>
<td>Taylor factor</td>
</tr>
<tr>
<td>Dynamic recovery constant</td>
</tr>
<tr>
<td>Mobility</td>
</tr>
<tr>
<td>Dislocation line tension</td>
</tr>
<tr>
<td>Boltzmann’s constant</td>
</tr>
<tr>
<td>Constant in Taylor’s equation</td>
</tr>
<tr>
<td>Shear modulus</td>
</tr>
<tr>
<td>Work hardening constant</td>
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<tr>
<td>Work hardening constant</td>
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</tr>
<tr>
<td>Grain size</td>
</tr>
<tr>
<td>Cavity radius</td>
</tr>
<tr>
<td>GBS parameter</td>
</tr>
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<tr>
<td>Parameter in the cavitation growth</td>
</tr>
<tr>
<td>equation</td>
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<tr>
<td>Grain boundary diffusion coefficient</td>
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<td>Cavity spacing</td>
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<tr>
<td>Atomic volume</td>
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<td>Grain boundary diffusion parameter</td>
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ABSTRACT
Extensive creep tests have been performed on oxygen free copper with 50 ppm phosphorus at both low and high temperatures. It is the candidate material for storage of spent nuclear fuel in Sweden. Basic models without fitting parameters have been formulated to reproduce primary and secondary creep. For a long time, only empirical models existed for fitting of tertiary creep. To understand the role of creep damage, including recovery, cavitation and necking, basic models that do not involve adjustable parameters are in urgent demand. Only recently, basic models taking the relevant mechanisms into account have been developed. These models were used to predict the tertiary creep for copper at 75°C. The modelled results were compared with experimental creep curves and good agreement has been found. In the present paper, the models are applied to creep tests at higher temperatures (215 and 250°C). A similar representation with good accuracy is obtained. This demonstrates that the fundamental model for back stress is applicable for the higher temperature tests as well.

INTRODUCTION
Oxygen free copper alloyed with 50 ppm phosphorus (Cu-OFP) has been selected as canister material for long term underground storage of spent nuclear fuel in Sweden [1]. The copper canisters are expected to stay intact for thousands of years. Creep damage might occur during storage since the canisters are exposed to creep deformation due to a pressure of around 15 MPa at a temperature up to 100 ºC. In order to predict the creep damage under such long time periods, it is critical to investigate the role of tertiary creep of copper with the use of fundamental models based only on physical phenomena.

The formation of creep damage is due to a number of mechanisms including particle coarsening, subgrain growth, cavitation and recovery of the dislocation structure, which can all accelerate the creep rate during tertiary creep. In addition, the creep rate is influenced by the necking instability. Empirical models describing creep damage accumulation have been widely applied for representing tertiary creep. However, basic models that take all important creep damage mechanisms into account have been absent. Very recently the authors have developed basic models to describe tertiary creep of copper for the relevant mechanisms [2]. In [2], a model for recovery of the dislocation structure was used to represent a major part of tertiary creep [3, 4]. It is suggested that the most important contribution of tertiary creep for copper comes from dynamic recovery. Models for the formation and growth of creep cavities were used to evaluate the cavitation evolution during tertiary creep [5-7]. At the investigated temperature 75 ºC, the contribution from cavitation was so small that it could be ignored. Necking instability was considered by introducing an initial imperfection on the specimen [8]. It gave a contribution at the end of the creep life.

In has been demonstrated that the models can represent the tertiary creep of copper at 75 ºC with good accuracy. The purpose of present study is to apply the models for the experimental data at higher temperatures to verify the approach to such conditions.

MATERIAL AND TESTING
The tested material was oxygen free copper alloyed with 50 ppm phosphorus. The chemical composition is given in Table1.
Table 1 Chemical composition of the Cu-OFP bars, ppm

<table>
<thead>
<tr>
<th></th>
<th>Cu</th>
<th>Ag</th>
<th>As</th>
<th>Bi</th>
<th>Cd</th>
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</tr>
<tr>
<td>O</td>
<td>Sb</td>
<td>S</td>
<td>Pb</td>
<td>Se</td>
<td>Sn</td>
<td>H</td>
<td>Te</td>
<td></td>
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<tr>
<td>0.9</td>
<td>4</td>
<td>6</td>
<td>&lt;1</td>
<td>2</td>
<td>&lt;3</td>
<td>&lt;0.1</td>
<td>&lt;3</td>
<td></td>
</tr>
<tr>
<td>Al</td>
<td>Co</td>
<td>Cr</td>
<td>Hg</td>
<td>Si</td>
<td>Zr</td>
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<td>&lt;3</td>
<td>&lt;1</td>
<td>50</td>
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</table>

Constant load creep tests were carried out in air in uniaxial tension on cylindrical specimens. The specimens were 5 mm in diameter and 50 mm in gauge length. The test temperatures considered in this paper are 215 and 250°C. The tests were performed with a wide range of applied stresses from 60 MPa to 150 MPa.

MODELS FOR TERTIARY CREEP

Accelerated recovery model

The accelerated recovery model is based on a basic model [9], which has successfully been used to model creep curves and stress strain curves of Cu-OFP. A brief summary of the basic model is given here.

\[
\frac{d \rho}{d \varepsilon} = \frac{m \rho^{1/2}}{b c_L} - \alpha \rho - 2 \tau_k \rho^2 / \dot{\varepsilon}
\]

Eq. (1) describes how the dislocation density, \( \rho \), varies with strain, \( \varepsilon \), during plastic deformation. The first term on the right hand side of the equation represents work hardening, where \( m \) is the Taylor factor, \( b \) Burger’s vector and \( c_L \) a constant. The second term describes dynamic recovery, where \( \alpha \) is a constant that controls the amount of dynamic recovery. The third term is static recovery, where \( \tau_k \) is the dislocation line tension, \( \dot{\varepsilon} \) the stationary strain rate and \( M \) the dislocation mobility. Dynamic recovery is strain dependent and takes place by the rearrangement of dislocation into lower energy configurations [10]. Static recovery is a time dependent process describing how dislocations of opposite sign attract each other and eventually annihilate. The basic model is used to describe dislocation density evolution during plastic deformation for annealed copper, which contains homogenous distributions of isolated dislocations. With as received materials, cell structures are commonly observed [11]. When the role of substructure is taken into account, an important assumption is made on the base of experimental evidence that dislocations in cell walls can be statistically distributed and polarised [12]. The dislocations in the cell walls can be classified as balanced and unbalanced dislocations. In the former set, the dislocations appear in pairs with opposite Burgers vectors, which makes the numbers of dislocations of opposite signs are balanced. Whereas for unbalanced dislocations, the dislocations are polarised and dislocations of opposite sign are missing. A more detailed definition can be found in [2]. Their density as a function of strain are given by [4]

\[
\frac{d \rho_{bnd}}{d \varepsilon} = k_{bnd} \frac{m \rho_{bnd}^{1/2}}{b c_L} - \alpha \rho_{bnd} - 2 \tau_k \rho_{bnd}^2 / \dot{\varepsilon}
\]

where \( \rho_{bnd} \) and \( \rho_{bnde} \) are the balanced and unbalanced dislocation density in the cell walls. Eq. (2) has the same form as Eq.(1), where both dynamic recovery and static recovery are considered. In Eq. (3), the static recovery term is missing. The reason is that unbalanced dislocations cannot be subjected to static recovery since they cannot find dislocations with opposite Burger’s vector to annihilate. Since the cell structure is a low energy configuration, the dislocations there give a lower contribution to the strength. Parameters \( k_{bnd} \) and \( k_{bnde} \) are introduced to take this into account and to control the amount of balanced and unbalanced dislocations.

A long-term internal stress is formed by the dislocations in cell walls. The stress in the cells can then be calculated with the help of Taylor’s equation, Eq.(4). Only dislocations in cell walls are taken into account.

\[
\sigma_{disl} = \sigma_y + \frac{m \alpha G b}{2} \sqrt{\rho_{bnd} + \rho_{bnde}}
\]

where \( \sigma_{disl} \) is the dislocation stress, \( \sigma_y \) the yield strength, \( \alpha \) a constant in Taylor’s equation, and \( G \) the shear modulus.

In the past, it has often been assumed that back stress can be measured by stress drop test or calculated by dislocation dynamics. However, both the measured and calculated values are almost identical to the applied stress [13], resulting in less meaning to use in modelling. Although A back stress formed by the extra hardening from dislocations in cell walls was introduced to compensate the sharp increase in true applied stress during secondary creep [4]. Its magnitude is equal to the dislocation stress minus the nominal applied stress

\[
\sigma_{back} = \sigma_{disl} - \sigma_{app} \cdot 0
\]

The back stress is introduced to simplify the interpretation of the model. However, it is not a quantity that can be measured directly. When the stress balancing requirement is fulfilled, the values of \( k_{bnd} \) and \( k_{bnde} \) can be determined. The effective creep stress is the true applied stress minus the accompanying back stress,

\[
\sigma_{creep} = \sigma_{app} - \sigma_{back}
\]

where \( \sigma_{app} \) is the true applied stress \( \sigma_{app} \cdot \exp (\varepsilon) \).

The secondary creep rate can be derived from Eq.(2). When a balanced state is reached during secondary creep, the work hardening (dislocation generation) rate and recovery (dislocation annihilation) rate should be the same. Therefore the strain derivative of the dislocation density is zero, indicating that the right side of Eq. (2) is zero. In addition, the effective creep stress should be applied and we get the expression for the whole creep curve instead of only primary and secondary creep.

\[
\varepsilon = 2 \tau_k (M, \sigma_{creep}) \rho_{bnd}^{1/2} / (k_{bnd} \frac{m}{b c_L} - \alpha \rho_{bnd}^{1/2})
\]

Dislocation densities for both sets of dislocations were computed for Cu-OFP tested at 215 °C with an applied stress of 120 MPa. The experimental creep curve is given in Figure 4a. To compute dislocation densities, the applied stress of 120MPa
was used to calculate creep stress using eq. (5). The creep stress
was then plugged in into eq. (6). The differential equations eq.
(2), (3) and (6) were integrated to the experimental rupture
time. Then the time vs density relation can be found, shown as
Figure 1. The balanced dislocation density reached a plateau
during secondary creep. On the other hand, the unbalanced
dislocation density kept increasing during the whole creep life.

It is evident from the expression of creep stress (Eq. (5)) that its
magnitude depends on the difference between the true applied
stress and the dislocation stress. The evolution of the
dislocation stress and true applied stress minus yield strength
with strain is plotted in

Figure 2. It can be clearly seen, during primary stage, the
difference is large and consequently the effective creep stress. A
high creep rate in primary creep is obtained. When the
stationary stage is reached, the dislocation stress is close to the
true applied stress, where the minimum creep rate is reached.
Afterwards, the difference increases again resulting in
increasing of creep stress. The accelerated creep rate of tertiary
creep can then be modelled. Another main contribution for the
acceleration of creep rate in tertiary stage is due to dynamic
recovery. In Figure 1, at the end of creep life, the balanced
dislocation density has increased. This means the numerator in
Eq.(6) increases and the denominator decreases leading to the
further increasing of creep rate during tertiary creep.

Figure 1 Balanced and unbalanced dislocation densities (Eq. (2)
and (3)) versus time for Cu-OFP at 215°C, 120MPa

Cavitation model

The nucleation of cavities, followed by growth and
interlinkage are believed to play an important role in creep
failure of metals [6, 7]. Quantitative basic models for
nucleation and growth of cavities have been developed recently
by He and Sandström [5-7]. For a review, see [14]. The models
will be introduced in the present study to investigate the
amount of cavitation in the grain boundaries. The equations
used in computation are listed here.

The cavity nucleation rate at intersections of sub-
boundary/sub-boundary corner is [6]
\[
\frac{dn_{cav}}{dt} = 0.9C_c \dot{\varepsilon}
\]  \hspace{1cm} (7)
where \(n_{cav}\) is the number of cavities, \(dn_{cav}/dt\) the cavity
nucleation rate, \(d_{sub}\) the subgrain size, and \(\dot{\varepsilon}\) the creep strain
rate. The constrained cavity growth rate is [15]
\[
\frac{dR_{cav}}{dt} = 2D_0K_f (\sigma_{red} - \sigma_0) - \frac{1}{R_{cav}^2}
\]  \hspace{1cm} (8)
where \(R_{cav}\) is the cavity radius, \(dR_{cav}/dt\) the cavity growth rate,
\(\sigma_0\) the sinter stress, \(D_0\) a grain boundary diffusion
coefficient, \(K_f\) a factor related to the cavitated area fraction at the grain
boundaries \(A_{cav}\). The parameters values can be found in
Appendix A. \(\sigma_{red}\) is the reduced stress, which can be determined
by solving a differential equation [7]
\[
2\pi D_0K_f (\sigma_{red} - \sigma_0)n_{cav}R_{cav} + \dot{\varepsilon}(\sigma_{red}) = \dot{\varepsilon}(\sigma_{appl})
\]  \hspace{1cm} (9)

The cavitated area fraction on the grain boundaries can be
expressed as [16]
\[
A_{cav} = \int_0^t \frac{dn_{cav}(t')}{dt} \pi R_{cav}^2(t,t') dt',
\]  \hspace{1cm} (10)
where \(R_{cav}(t, t')\) is the radius of the cavity at time \(t\) that was
formed at time \(t'\). It is well established that when the cavitated
area fraction exceeds 25\%, rupture occurs [17].

Figure 2 Comparison of dislocation stress minus yield strength,
Eq. (4), with true applied stress minus yield strength as a
function of strain for Cu-OFP at 250 °C, 90 MPa
Necking model

Studies on the effect of an initial defect on creep deformation have been carried out [18-21]. In the present study, a uniaxial necking model proposed by Burke and Nix [8] was employed. The basic idea is to artificially introduce a non-uniform cross section area to a cylinder bar from the beginning of test. The initial cross section area varied along the x-axis in a sinusoidal manner

\[ A(x,0) = A_0 - \frac{\Delta A}{2} \cos \frac{2\pi x}{L_i}, \quad (0 \leq x \leq L_i) \]

where \( A_0 \) is the original cross section area of the specimen, and \( L_i \) the length of the specimen with the defect part. \( \Delta A \) is the change in initial cross section area, which is chosen as \( \Delta A/A_0=0.01 \) in the current case. Accordingly the maximum strain in the beginning of the test is 1%. The original radius of the specimen was 5 mm; after introducing the imperfection, a minimum radius of the necking part was 4.975 mm. When a neck is introduced, the stress state changes from uniaxial to multiaxial. In order to avoid dealing with multiaxial stress state in computation, the specimen was sectioned into several slices. The specimen is consequently represented by a set of disc shaped elements. Within each element, the gradient of cross section area is small enough to be neglected. In this way a uniaxial stress state can be assumed, which means the deformation is homogenous for each element. The governing equation Eq. (6) will be integrated numerically in all these sets of elements. The elongation of each element (with different initial strain) can then be calculated for a given time step directly, since the load on each section is constant and equal to the applied load on the specimen. The average strain for the whole specimen is calculated by summing the deformation of each element divided by the gauge length of the specimen. This average strain for the entire specimen can be used to compare with the experimental data.

The influence of multiaxiality on necking is studied with finite element methods (FEM). Details are given below.

RESULTS

Cavitation results

Results corresponding to two creep tests will be shown, 215°C 120 MPa and 250°C 90 MPa. The computed number of cavities per unit grain boundary area and cavity radius as a function of rupture time are given in Figure 3. The number of cavities increases sharply during the initial stages of creep and more gradually afterwards. The cavity radius keeps increasing during the whole creep test.

![Figure 3](image-url)

The cavitated area fraction on grain boundaries at rupture can then be calculated using Eq. (10). Its magnitude is about 7%. When integrating the strain vs time curves, the strain difference is only 2% at rupture, a difference that is hardly possible to see on a plotted creep strain versus time curve. For the other case 250°C 90 MPa, the amount of cavitation is even smaller. The cavitated area was 3% and strain at rupture was increased by less than 1%. The computed area fractions of cavities are consistent with metallographic observations on rupture specimens. Detailed measurements of the cavitation were never performed so a detailed comparison cannot be made. In the same way as for the analysis at 75°C in [2], cavitation gives a very small contribution to tertiary creep, and it will be neglected in the present paper.

Accelerated recovery results

By integrating the set of Eqs. (2), (3) and (6), creep strain versus time curves can be computed. A comparison of experimental creep curves and modelled results is given in Figure 4. In both test conditions, the modelled creep curves can represent the experimental data in a good way. In addition, it
can be noted that the model can describe the logarithmic decrease in the strain rate in the primary stage, and the rapid increase in the tertiary can also be represented, shown in Figure 5.

![Image](image1.png)

Figure 4 Comparison of experimental creep curve with accelerated recovery model Eq.(6) for Cu-OFP at a) 215 °C, 120MPa; b) 250 °C, 120 MPa

Necking results

Comparison of experimental curve with the model taken necking into account is shown in Figure 6. It gives a better representation of the final part of the creep curve. The sharp increase of strain can be modelled, indicating that the necking gives a significant contribution at the end of creep life. In fact, the results show that necking is of importance at the end of the tests.

![Image](image2.png)

Figure 6 Comparison of experimental creep curve with necking model results for Cu-OFP at 250 °C, 120 MPa

The necking was also computed by finite element method for the case at 250 °C with an applied stress of 120MPa using a commercial software Comsol Multiphysics 5.3. The strain distribution after necking is shown in Figure 7. The maximum local strain at necking position is as high as 1.2, indicating a reduction in area around 70%.
DISCUSSION

Dislocation recovery mechanisms have been used to describe the three stages of creep deformation. Fundamental dislocation models based on this mechanism for primary and secondary creep were formulated [9, 22]. The models involve no fitting parameters. It has been demonstrated that they can be used to describe the primary and secondary creep of Cu-OFP and also slow strain rate tensile tests under both uniaxial and multiaxial stress states [23, 24]. It has also been successfully applied to austenitic stainless steels [25] and aluminum alloys [26]. By taking the recovery of substructure into account the effect of cold work on the deformation of copper can be modelled [3, 4]. To be able to describe this effect, a distinction of balanced and unbalanced dislocation in the cell walls has to be introduced. For balanced dislocations, dislocations with opposite Burgers vector are always present, while the unbalanced dislocations cannot find dislocations with the opposite Burgers vector. The unbalanced dislocations are not subject to static recovery but only to dynamic recovery.

The accelerated model formulated for cold worked material has been employed to predict the tertiary creep for Cu-OFP at low temperatures with a good accuracy [2]. In the present paper, the creep curves at temperatures around 225°C are modelled using the same sets of models. The creep curves at the higher temperatures look quite similar to those obtained at low temperatures. The low temperature creep mechanism has not been developed sufficiently. It has been suggested that the low temperature creep may be controlled by climb mediated by vacancies produced by plastic deformation [27], [28]. This supposed mechanism has been supported by the successful representation of creep rates at ambient temperatures for aluminum and copper [28]. If climb is the operating mechanism for low temperature creep of copper, it is natural that the creep curves at high and low temperature are similar.

The modelled results of high temperature tests showed that all three stages of creep curves are well reproduced. It gave a similar prediction as at lower temperature (75°C). Accelerated recovery predicted a longer tertiary time with smoothly increase. Necking modelled the steep increase of strain and contributed to the end of creep life, which is consistent with experimental necking observations for 9% Cr steels in [29].

CONCLUSIONS

There are several possible creep damage mechanisms including microstructure degradation, cavitation, necking instability and recovery which can accelerate the creep rate during tertiary creep. Fundamental models taking the relevant mechanisms into account have been used to predict tertiary creep for copper at low temperatures. In the present paper, the models were applied to higher temperatures at 215 and 250°C. According to the results, some main conclusions can be drawn.

- By taking dynamic recovery of dislocation substructure into account, the recovery rate during tertiary can be strongly accelerated. It has given the largest contribution to tertiary creep.

Figure 7 FEM modelling results of strain distribution after necking for Cu-OFP at 250 °C, 120 MPa

The total strain compared with uniform strain is shown in Figure 8. The total strain was calculated by the elongation of specimen divided by the gauge length. The uniform strain was computed by the area reduction of one point away from necking position, according to \( \varepsilon = \ln(A_0 / A) \). The total strain showed a linear relationship with uniform strain until necking occurred. Afterwards, the uniform strain was constant indicating that all the further deformation took place around the neck.

Figure 8 Total strain compared with uniform strain for Cu-OFP at 250 °C, 120 MPa
• The three stages of creep curves can be well reproduced.
• At a higher temperature, the cavitated area fraction on grain boundaries at rupture time is about 7% or less. This contributes to a 2% change to strain at rupture. Consequently, the cavitation does not influence the appearance of creep curves significantly.
• When necking is taken into account, it gives a contribution only at the very end of creep life.
• The fundamental models for tertiary creep of copper at low temperatures (75°C) can be used also for the higher temperatures (215 and 250°C).

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REFERENCES
# APPENDIX A

## PARAMETER VALUES

Table A1 Parameters used in this paper

<table>
<thead>
<tr>
<th>Parameter description</th>
<th>Parameter</th>
<th>Value</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Burgers’ vector</td>
<td>( b )</td>
<td>( 2.56 \times 10^{-10} \text{ m} )</td>
<td>[9]</td>
</tr>
<tr>
<td>Taylor factor</td>
<td>( m )</td>
<td>3.06</td>
<td>[30]</td>
</tr>
<tr>
<td>Dynamic recovery constant</td>
<td>( \omega )</td>
<td>14.7</td>
<td></td>
</tr>
<tr>
<td>Mobility</td>
<td>( M )</td>
<td></td>
<td>[31]</td>
</tr>
<tr>
<td>Dislocation line tension</td>
<td>( \tau_L )</td>
<td>( Gb^2/2 = 7.94 \times 10^{-16} \text{ MN} )</td>
<td>[31]</td>
</tr>
<tr>
<td>Boltzmann’s constant</td>
<td>( k_B )</td>
<td>( 1.381 \times 10^{-23} \text{ J/grad} )</td>
<td></td>
</tr>
<tr>
<td>Constant in Taylor’s equation</td>
<td>( \alpha )</td>
<td>((1-v/2)/2\pi(1-v)=0.19 \text{ with Poisson’s ratio } v=0.308)</td>
<td>[32, 33]</td>
</tr>
<tr>
<td>Shear modulus</td>
<td>( G )</td>
<td>( G = 45400(1 - 7.1 \times 10^{-4}(T - 20)) \text{ MPa, } T \text{ in } °C )</td>
<td>[34]</td>
</tr>
<tr>
<td>Work hardening constant</td>
<td>( k_{bnd}, k_{bnde} )</td>
<td>2.3 for 215°C and 3.1 for 250°C</td>
<td></td>
</tr>
<tr>
<td>Work hardening constant</td>
<td>( C_L )</td>
<td>30</td>
<td>[22]</td>
</tr>
<tr>
<td>Grain size</td>
<td>( d )</td>
<td>( 1 \times 10^{-4} \text{ m} )</td>
<td>[9]</td>
</tr>
<tr>
<td>Cavity radius</td>
<td>( r_h )</td>
<td>( 1 \times 10^{-6} \text{ m} )</td>
<td>[16]</td>
</tr>
<tr>
<td>GBS parameter</td>
<td>( C_s )</td>
<td>( 50 \mu \text{ m} )</td>
<td>[14]</td>
</tr>
<tr>
<td>Subgrain size</td>
<td>( d_{sub} )</td>
<td>( \frac{Kgb}{\sigma_{appl}} \text{ with K=11 for copper} )</td>
<td>[14]</td>
</tr>
<tr>
<td>Parameter in the cavitation growth equation</td>
<td>( K_f )</td>
<td>( -2 \log A_{cav} - (1 - A_{cav})(3 - A_{cav}) )</td>
<td>[7]</td>
</tr>
<tr>
<td>Grain boundary diffusion parameter</td>
<td>( \delta D_{gb} )</td>
<td>( \delta D_{gb0} \exp(\frac{Q_{gb}}{RT}) \text{ with } \delta=2b, D_{gb0}=2.4 \times 10^{-6} \text{ m}^2\text{s}^{-1} \text{ and } Q_{gb}=8.49 \times 10^{4} \text{ J/mol} )</td>
<td>[35]</td>
</tr>
<tr>
<td>Cavity spacing</td>
<td>( L )</td>
<td>( 1/\sqrt{n_{cav}} )</td>
<td>[14]</td>
</tr>
<tr>
<td>Atomic volume</td>
<td>( \Omega )</td>
<td>( 1.18 \times 10^{-29} \text{ m}^3 )</td>
<td></td>
</tr>
<tr>
<td>Grain boundary diffusion parameter</td>
<td>( D_0 )</td>
<td>( \delta D_{gb0} \Omega/k_B T )</td>
<td>[7]</td>
</tr>
</tbody>
</table>
Creep Strength Contribution due to Precipitation Hardening in Copper-Cobalt Alloys

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Abstract

In spite of its huge technical significance, there does not seem to be consensus about how to model the precipitation contribution to the creep strength. Most contributions in the literature are based on a constant internal stress (also called back stress or threshold stress) from the precipitation. It is well-known and it will also be demonstrated in the paper that this assumption is at variance with observations except for some ODS alloys. There is however one model developed by Eliasson et al. (2000) that seems to be able to represent experimental data without the use of any adjustable parameters. It has successfully been applied to describe the creep strength of austenitic stainless steels. Due to the fact that various mechanisms contribute to the creep strength in these steels, the model has not been fully verified. The purpose of this paper is to apply the model to published creep data for Cu-Co alloys, where the precipitation totally dominates the strength contribution to validate the model. In the paper, it is demonstrated that the model can indeed describe the influence of applied stress, alloy composition and heat treatment for the three analysed Cu-Co alloys.

Keywords

Creep strength; Precipitation hardening; Copper-cobalt

1. Introduction

General methods to increase the resistance to plastic deformation of metallic materials are cold working, precipitation hardening and solid solution strengthening [1]. Precipitation hardening is often the most efficient way to improve the creep strength of high temperature
alloys. A classical approach to analyse precipitation hardening during creep is to introduce a constant internal stress $\sigma_i$ in a power law creep rate expression [2].

$$\dot{\varepsilon} = A(\sigma - \sigma_i)^n$$  \hspace{1cm} (1)

where $A$ is a constant at a given temperature and $n$ stress exponent for the alloy corresponding to the matrix. The internal stress is often also called back stress or threshold stress. $\sigma_i$ was in general treated as an adjustable parameter for fitting a linear relationship of double logarithmic creep rate versus reduced stress. A direct way of determining $\sigma_i$ was obtained by using a Lagneborg-Bergman plot [3]. A constant $\sigma_i$, results in a threshold stress below which no creep takes place. For oxide dispersion strengthening (ODS) alloys such a threshold has been observed [4, 5] and it can be essentially independent of temperature. Eq. (1) gives a creep exponent that decreases with increasing stress. This has been found for some ODS alloys [5], but rarely for precipitation strengthened steels, where the stress exponent is raised with increasing stress. Consequently, a constant threshold stress is not applicable to precipitation strengthened alloys.

Orowan bowing has often been used to estimate the threshold stress but it usually results in an overestimate of the creep strength, since dislocations can climb across the particles at high temperatures. A threshold stress has also been determined by taking climb into account [6]. In the past, many attempts were made to compute this threshold stress and the predicted values were quite different, depending sensitively on the assumptions of the models [7]. Two kinds of models have been brought forward. One kind was based on local climb assumption proposed by Brown and Ham [8], which postulated that the climbing dislocation segment was attached to the particle and the dislocation between the particles remained in its glide plane. The predicted stress was about half of the Orowan stress, which was consistent with the experimental values of some precipitation strengthened materials. However, the assumption itself was unrealistic since the sharp bend in dislocation can be rapidly relaxed by diffusion. Under the assumption of general climb, the climbing dislocation is only in contact with the particle at a single point. Gradually researches found decreasing threshold stress [4, 5, 9]. Only threshold stresses as low as 0.02 to 0.06 of the Orowan stress were eventually predicted [4, 10]. The general climb assumption was more realistic than the local climb assumption and has been demonstrated to be the energetically favourable case. However, the stress or energy barrier for dislocation to climb over particles was quite small which cannot explain the contribution of particles to the creep strength in general [5,
One possibility is to assume an attractive dislocation-particle interaction. Such an interaction has been observed for oxide dispersion strengthened (ODE) alloys [11, 12], but rarely for precipitation hardened systems. Another important difference between dispersion and precipitation strengthened systems is that a true threshold stress can be present in ODE alloys [5]. In such a case, the creep rate is negligible below the threshold stress. However, for precipitation strengthened materials, the situation is different. For example, for common creep resistant CrMo steels that are precipitation strengthened, no threshold stress is found. There are also ODS alloys where no threshold stress is observed [6].

In general the climb threshold stresses are too low to explain the observed influence of particles on the creep strength. To handle this situation, a time controlled climb model was then proposed by Eliasson et al. to evaluate the actual amount of climb [13]. For particles with different sizes, it will take different time for a dislocation to climb across. A critical size is introduced to describe the maximum particle radius that the dislocations can climb within the lifetime. The assumption of this model is that the key controlling mechanism for a dislocation to climb across a particle or not is the time it takes. The actual amount of climb is calculated by comparing the time for dislocations to glide and to climb. Only particles that are large enough cannot be passed by climbing dislocations give a contribution to the creep strength. The model has been used to evaluate the particle contribution for 9 and 12Cr steels and austenitic stainless steels [14, 15]. A good agreement with the experimental values was obtained.

In fact, creep strength is gained not only from precipitation hardening but also from solid solution hardening and dislocation densities. In previous studies, the creep strength has been calculated taking the three contributions into account. In the present paper, the contribution of precipitation hardening to the creep strength is evaluated for previously published creep data for copper-cobalt alloys with different cobalt content and different particle size distribution [16, 17]. It is verified that the contribution from solid solution hardening is negligible. The aim of present paper is to study the precipitation hardening mechanism using copper cobalt system without having to take into account the influence of many kinds of particles or of elements in solid solution. In this way, the accuracy of the precipitation hardening model can be verified.
2. Material and testing

Creep data for three copper cobalt alloys (with 0.88 wt.-%, 2.48 wt.- % and wt.- 4.04% cobalt) published by Wilshire et al. are analysed in the present paper [16, 17]. Copper cobalt is a good system for the study of precipitation hardening since it is a simple alloy containing only coherent cobalt particles with little disturbance from elements in solid solution. The precipitates nucleate shortly after the aging process starts, so the volume fraction of precipitates remains constant during the aging process. The aging was performed at a higher temperature than the creep test, and in this way the creep induced precipitation could be avoided. In addition, the precipitation hardening is the dominate contribution to the creep strength.

The three copper cobalt alloys were solution treated followed by water quenching, which resulted in a uniform grain size of about 200 nm. In order to obtain a uniform distribution of particles, aging treatment was carried out for the alloys during various time. The detailed aging procedures are given in Ref. [16]. Cu4.04Co was aged at a higher temperature than the other alloys due to sluggish precipitation, but the alloy was stabilised at the lower heat treatment temperature to avoid precipitation during creep. After aging, the particle sizes were measured under microscope. The particle characteristics for the different alloys is summarised in Table 1.

<table>
<thead>
<tr>
<th>Co, wt.%</th>
<th>Heat treatment temperature, °C</th>
<th>Heat treatment type</th>
<th>Particle volume fraction</th>
<th>Co in solid solution, wt.%</th>
<th>Particle radius, nm</th>
<th>Particle spacing, nm</th>
<th>Orowan stress, MPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.88</td>
<td>600</td>
<td>underaged</td>
<td>0.00567</td>
<td>0.33</td>
<td>1.2</td>
<td>41</td>
<td>593</td>
</tr>
<tr>
<td>0.88</td>
<td>600</td>
<td>aged</td>
<td>0.00567</td>
<td>0.33</td>
<td>4.2</td>
<td>98</td>
<td>250</td>
</tr>
<tr>
<td>0.88</td>
<td>600</td>
<td>overaged</td>
<td>0.00567</td>
<td>0.33</td>
<td>17.2</td>
<td>405</td>
<td>60</td>
</tr>
<tr>
<td>2.48</td>
<td>600</td>
<td>aged</td>
<td>0.0222</td>
<td>0.33</td>
<td>7.6</td>
<td>90</td>
<td>272</td>
</tr>
<tr>
<td>4.04</td>
<td>700</td>
<td>aged</td>
<td>0.0344</td>
<td>0.33*</td>
<td>22.6</td>
<td>215</td>
<td>113</td>
</tr>
</tbody>
</table>

*Stabilised at 600°C after heat treatment at 700°C

Micro hardness for the Cu0.88Co alloys at different ageing time was measured. The heat treatment that gave maximum micro hardness is referred to as (fully) aged. When the radius is smaller than in the aged condition, it is called underaged and when it is larger overaged. The volume fraction of the precipitates and the amount of Co in solid solution were calculated with
the thermodynamic software Thermo-Calc. The interparticle spacing was determined with the expressions for planar square lattice particle spacing $\lambda_s$,

$$\lambda_s = \bar{r} \sqrt{\frac{2\pi}{3f}}$$  \hspace{1cm} (2)

where $\bar{r}$ is the mean radius of the particles and $f$ the volume fraction of precipitate. The constant stress tensile creep tests were conducted at 439°C. No particle growth was recorded during the creep tests [16].

3. Model for precipitation hardening

3.1 Dislocation creep

It will be assumed that the precipitation hardening during creep can be taken into account with the help of an internal stress. This assumption has practically always been made in the literature. The internal stress is also called back stress or threshold stress. When a threshold stress is introduced, it is in general assumed as a constant, i.e. is essentially independent of temperature and applied stress. This terminology will be followed in the present paper. But before the details about the precipitation hardening can be given, a model for secondary creep is needed for a particle free material. In the past, the influence of applied stress, $\sigma$, and temperature $T$ on the secondary creep rate $\dot{\varepsilon}_{sec}$ has been represented by the conventional power-law and Arrhenius equations in the form

$$\dot{\varepsilon}_{sec} = A D_{0sd} G b \left( \frac{\sigma}{G} \right)^n \exp\left( -\frac{Q_{sd}}{k_B T} \right)$$  \hspace{1cm} (3)

where $G$ is the shear modulus, $b$ the length of the Burgers vector, $k_B$ the Boltzmann’s constant and $R_g$ the gas constant. $D_{0sd}$ in eq. (3) is the pre-exponential factor in the Arrhenius equation for self-diffusion and $Q_{sd}$ is the activation energy for self-diffusion in the alloy. There are several limitations of eq. (3). There are at least two constants, $A$ and $n$, that are used as adjustable parameters and fitted to experimental data. This means that the equation cannot be used for predicting creep properties. This makes it also quite risky to extrapolate results to new conditions. Furthermore, its use is restricted to the power-law regime where $n$ is approximately constant, so it
cannot describe creep at lower temperatures where the creep exponent \( n \) rapidly increases with decreasing temperature.

To avoid all these limitations, a new model for secondary creep has been developed. It was originally formulated for copper [18] but has been shown to be valid also for aluminium alloys [19, 20] and stainless steels [21, 22]. The model is derived from fundamental dislocation formulations. The derivation can be found in several places [23, 24] and it will not be repeated here. The important aspect is that it is based on a balance between work hardening and recovery in the secondary stage. The resulting expression for the secondary creep rate is

\[
\dot{\varepsilon}_{\text{sec}} = \frac{2\tau L b}{m} M_{cl}(T, \sigma_{\text{disl}}) \left( \frac{\sigma_{\text{disl}}}{\alpha_T m G b} \right)^3
\]

(4)

\( \tau_L \) is the dislocation line tension, \( c_L \) a work hardening parameter, and \( m = 3.06 \) the Taylor factor. The constant \( \alpha_T \) appears in Taylor’s equation

\[
\sigma_{\text{disl}} = \alpha_T m G b \rho^{1/2} = \sigma - \sigma_i
\]

(5)

\( \sigma_{\text{disl}} \) is the dislocation stress. \( \sigma_i \) is an internal stress that can have contributions from the yield strength, solid solution hardening and precipitation hardening. The dislocation climb mobility is given by

\[
M_{cl}(T, \sigma) = \frac{D_{0\text{sd}} b \sigma b^3}{k_B T} e^{\frac{Q_{\text{sd}}}{k_B T}} e^{\frac{\sigma}{R_m} \left[ 1 - \left( \frac{\sigma}{R_m} \right)^2 \right]}
\]

(6)

where \( R_m \) is the tensile strength (at room temperature). All the parameters in eqs. (4) to (6) are given and none is used as adjustable parameter and fitted to the mechanical test data. Equation (4) has been generalized to describe primary and tertiary creep as well [24-26] but since only secondary creep will be discussed in the present paper, these extensions will not be considered here.

### 3.2 Precipitation hardening

Three main mechanisms for how dislocations bypass particles are in general considered: particle shearing, Orowan bowing and climb across particles. The stress required for Orowan bowing \( \sigma_O \) is

\[
\sigma_O = \frac{m C G b}{\lambda}
\]

(7)
where $\lambda$ is the interparticle spacing and $C = 0.8$ is a constant [27]. $\sigma_0$ is only weakly temperature dependent (through $G$). Eq. (7) has sometimes been used to estimate the particle contribution to the creep strength, but in general it will largely overestimate the contribution. This will be exemplified in section 5. The reason is that the stress required for climb is much less. Initially the threshold stress was estimated to be quite high, but later studies gave lower values [2, 4, 5, 28, 29]. Reviews are given in [5],[6]. The best estimate of the threshold stress, i.e. the minimum climb stress $\sigma_{\text{clmin}}$ is now considered to be [4, 5]

$$\frac{\sigma_{\text{clmin}}}{\sigma_0} = \frac{\alpha}{\alpha + 2C}$$

(8)

where

$$\alpha = \frac{2f}{3\lambda} = \sqrt{\frac{2f}{3\pi}}$$

(9)

The minimum climb stress is related to the Orowan stress through the parameter $\alpha$, which is referred to as the \textit{climb resistance}. The final step in eq. (9) is obtained by assuming that the planar square lattice particle spacing $\bar{r}\sqrt{2\pi f/3f}$ is used for $\lambda$. $\alpha$ is quite small. For volume fractions $f$ of 1 and 5%, $\alpha$ is 0.05 and 0.10, which gives threshold stresses of $0.03\sigma_0$ and $0.06\sigma_0$, respectively. Later analysis has confirmed these low values [7].

Although the threshold stress is quite small it is not negligible, but it obviously does not give the full picture. To handle this situation, a different track will be followed. The following assumptions will be made [13, 14].

- Only precipitation hardened alloys will be considered. Consequently, the attractive interaction between particles and dislocations will be neglected.
- It will be assumed that it is the time it takes for a dislocation to climb across a particle that is the controlling mechanism instead of the threshold stress. Thus, it will be the time it takes for the dislocation to climb across the particles that determines whether a particle will be climbed or not.
- The maximum particle size where there is enough time for dislocations to climb across particles is referred to as the critical radius $r_{\text{crit}}$.
- For smaller particles there is sufficient time for the dislocations to climb across them. These particles will not contribute to the creep strength.
Particle shearing is not considered since the dislocations are assumed to climb across small particles freely.

Larger particles have to be passed by Orowan bowing, which determines their contribution to the strength.

These principles have been applied to austenitic stainless steels. It has been possible to predict the total creep strength quite accurately [15, 21, 22, 30].

For climb to be of significance, the time for a dislocation to climb across a particle \( t_{\text{climb}} \) must be at least as long as the glide time between the particles \( t_{\text{glide}} \). This criterion can be used to find the critical radius.

\[
t_{\text{climb}} = t_{\text{glide}}
\]

The climb time is equal to the critical radius \( r_{\text{crit}} \) divided by the climb velocity \( v_{\text{climb}} \)

\[
t_{\text{climb}} = \frac{r_{\text{crit}}}{v_{\text{climb}}}
\]

The climb velocity is proportional to the climb mobility, eq. (6)

\[
v_{\text{climb}} = M_{\text{cl}}(T, \sigma) b \sigma
\]

It should be noticed that it is the full applied stress that appears in eq. (12) and not the dislocation stress, eq. (5). The reason is that the climb rate is used to compute the time to pass the particles, i.e. their effective resistance against climb. If the dislocation stress would be used, the influence of the particles would be taken into account twice. In previous versions of the model, the dislocation stress was applied in eq. (12). This is now considered to be a mistake. The reason that it did not affect the results much was that the internal stress from the particles was only a smaller part of the total creep strength. For the Cu-Co system this is not the case as will be seen below.

The glide time is given by the interparticle spacing \( \lambda \) divided by the glide velocity \( v_{\text{glide}} \)

\[
t_{\text{glide}} = \frac{\lambda}{v_{\text{glide}}}
\]

The glide velocity can be found from the Orowan equation for the creep rate

\[
\dot{\varepsilon} = v_{\text{glide}} \frac{b \rho}{m}
\]
where \( \rho \) is the dislocation density. Combining eqs. (10) to (14) gives an expression for the critical radius.

\[
    r_{\text{crit}} = M_{cl}(T, \sigma)b^2 \lambda_F \frac{\rho}{\dot{\varepsilon}_{\text{sec}} m} 
\]  

(15)

The secondary creep rate in eq. (4) is used for the creep rate in eq. (15) but with the dislocation stress, eq. (5) instead of the applied stress. In eq. (15), the Friedel spacing \( \lambda_F \) is introduced, which is a change in relation to previous versions of the model. It is believed to represent the actual spacing of particles along the dislocation line better than the planar square lattice particle spacing \( \lambda_s \) [1, 5]. \( \lambda_F \) depends on the force \( F \) acting on a climbing segment

\[
    \left( \frac{\lambda_s}{\lambda_F} \right)^2 = \frac{F}{2\tau_L} = \frac{\sigma_{\text{clmin}} b \lambda_F}{2m \tau_L} = \frac{\sigma_{\text{clmin}} \lambda_F}{\sigma_O \lambda_s} 
\]  

(16)

From eqs. (8) and (16) we find that

\[
    \left( \frac{\lambda_s}{\lambda_F} \right)^3 = \frac{\alpha}{\alpha + 2C} 
\]  

(17)

With this relation the Friedel spacing can be obtained directly.

**3.3 Size distributions**

The authors have found that precipitates in creep resistant steels often form exponential size distributions, see for example [22, 31]. The number of particles per unit area \( N_A \) can be described by

\[
    N_A = N_{A0} e^{-k(r-r_0)} 
\]  

(18)

where \( N_{A0} = 1/\lambda_s^2 \), and \( r \) is the particle radius. \( r_0 \) is a small quantity taking into account that there is often no reliable observations at very small particle sizes; \( r_0 \) is taken as 1 nm. \( k \) is related to the average particle size \( \bar{r} : k = 1/(\bar{r} - r_0) \). As emphasized above, only particles larger than \( r_{\text{crit}} \) are assumed to contribute to the creep strength. The average spacing between such particles is referred to as \( \lambda_{\text{crit}} \).

\[
    \lambda_{\text{crit}} = \sqrt{N_{A0} e^{-k(r_{\text{crit}}-r_0)/2}} 
\]  

(19)
Assuming that particles larger than $r_{\text{crit}}$ contribute to the creep strength $\sigma_{\text{part creep}}$ through the Orowan mechanism, we find that

$$\sigma_{\text{part creep}} = \frac{CGbm}{\lambda_{\text{crit}}} = \sigma_{\text{O}} e^{-k(r_{\text{crit}}-\eta_0)/2} \quad (20)$$

Since $r_{\text{crit}}$ depends on temperature and applied stress, so does $\sigma_{\text{part creep}}$. $\sigma_{\text{part creep}}$ is the internal stress that should be inserted in eq. (5).

### 3.4. Solid solution hardening

The investigated alloys contains 0.33 wt% Co in solid solution, see Table 1. For completeness, the contribution to the creep strength will be calculated in spite of the low Co content. The linear size misfit between Co and Cu atoms is 1.28% [32]. The principles for solid solution hardening during creep are presented in [33]. The maximum interaction energy between a solute and a dislocation is $U_{\text{max}}=2.88\times10^{-40}$ J. The value of $\beta$ is $U_{\text{max}}/b = 1.13\times10^{-30}$ J/m. The precise expression for the solid solution hardening depends on the radius $p$

$$p = \frac{\beta}{k_B T} \quad (21)$$

If $p$ is less than the core radius which is taken as $b$, the following expression applies for the amount of solid drag [34]

$$\sigma_{\text{drag}} = \frac{v_{\text{glide}} c_0 \beta^2}{D_{\text{Co}} k_B T b} \ln\left(\frac{D_{\text{Co}}}{v_{\text{glide}} b}\right) \quad (22)$$

where $v_{\text{glide}}$ is given by eq. (14), $c_0$ is the amount of Co in solid solution (atom fraction), and $D_{\text{Co}}$ is diffusion coefficient for Co in Cu. The resulting solid solution hardening lies between 0.15 and 0.25 MPa for the Cu-Co alloys. This low value is neglected in the analysis below.

### 4. Results

#### 4.1 Pure copper

Except for the creep tests for Cu-Co alloys at 439°C, creep tests for oxygen free pure copper (CuOF) were performed at the same temperature [16, 17]. In addition, creep data for CuOF have been taken from ref. [35] at different temperatures. A comparison between these experimental data and the model in eq. (4) is shown in Figure 1. It is obvious that the temperature dependence
of the creep rate is larger in the model than in the experiments. The reason is that the creep activation energy is smaller than that for self-diffusion in this temperature range, which has not been fully explained. However, the accuracy of the prediction around 439°C is sufficient for a meaningful comparison to Cu-Co alloys.

![Graph showing creep rate vs. stress for different temperatures](image)

Figure 1 Modelling of stationary creep rate (eq.(4)) for CuOF at different test temperatures compared with experimental data (Experimental data from [35] and [16])

### 4.2 Cu-Co alloys

As discussed in section 3, the maximum radius $r_{\text{crit}}$ for which the dislocations have time to climb across the particles, play an important role. Exponential size distributions are assumed, since in previous studies, such size distributions were found in creep resistant steels both for the austenitic stainless steels 310NbN and Sanicro 25 as well as for the Cr-Mo-steels P91 and P92 [14, 22]. The assumed exponential size distributions Cu0.88Co are illustrated in Figure 2. Critical radii of particles at 439°C for the experimental stress ranges were calculated by eq. (15). The critical radius decreases with increasing applied stress.
Figure 2 Size distributions (eq. (18)) for Cu0.88Co particles in aged, underaged and overaged conditions with critical radii marked (eq. (15))

The internal stress from the particles (critical Orowan stress) according to eq. (20) is illustrated in Figure 3. The internal stress increases with applied stress. This means that $r_{\text{crit}}$ decreases with increasing applied stress as pointed out above. The difference between the internal stress and the applied stress and the ratio between them increase with increasing applied stress.
Figure 3 Critical Orowan stress (eq. (20)) versus applied stress for Cu0.88Co. For comparison, a 1:1 line for the applied stress is included in the diagram.

The creep rates for the Cu-Co alloys can now be determined. Eq. (4) for the creep rate of pure copper is used but with the dislocation stress $\sigma - \sigma_i$, eq. (5). $\sigma_i$ is then given by eq. (20). The result is illustrated in Figure 4, where the modelled results are compared with the experimental data of Cu-Co alloys. For the considered Cu-Co alloys, the precipitation hardening has quite a dramatic effect. The creep rate of Cu-Co alloys is two orders of magnitude or more below that of pure copper. With increasing Co content, the creep rate decreases. This is a consequence of the denser particle distribution of the alloys with higher Co content.
Figure 4 Modelling of stationary creep rate (eq.(4)) for three Cu-Co alloys and pure copper compared with experimental data from [16]

In Figure 4, the slope of the curves for Cu-Co alloys increases with applied stress, i.e. the creep stress exponent is raised. There are two reasons for this behaviour. First, the dislocation stress $\sigma - \sigma_i$ increases faster than the applied stress. This is evident from Figure 3. Second, the expression for dislocation mobility in eq. (6) increases with applied stress. This is due to the strain induced increase in vacancy concentration at higher stresses [19, 20].

The creep data in [16] gives another possibility for comparison to the model. The Cu0.88Co alloy was investigated at different ageing times, corresponding to underaged, aged and overaged conditions, see Table 1. The comparison with the model is given in Figure 5. Again the model can represent the observations in a reasonable way.
5. Discussion

There is an extensive literature on the strengthening mechanism of particles during creep. The simplest model is to assume that the contribution from the particles is given by the Orowan stress. The values for the Orowan stress at 439°C are given in Table 1. Modelled values that at least approximately represent the experimental internal stresses can be found in Figure 3. Thus, a direct comparison can be made for the three Cu-Co alloys: 250 (10-25), 272 (20-65) and 113 (20-45) MPa. The first figure is the calculated Orowan stress and the values in brackets the modelled internal stress range in Figure 3. It is evident that the Orowan stress grossly overestimates the internal stress. In addition, the Orowan stress does not even rank the alloys in the right order.

Many attempts have been made in the literature to estimate a threshold stress for climb. The consensus now seems to be that this threshold stress is quite low and approximately given by eq. (8). For an alloy with a particle volume fraction of 1%, this gives 0.03 \( \sigma_0 \), where \( \sigma_0 \) is the
Orowan stress. This is a quite low value and cannot represent all of the precipitation hardening. For the Cu0.88Co alloy in the underaged condition, $\sigma_0$ is 597 MPa (Table 1). With a particle volume fraction of 0.57%, a threshold stress of the 0.02 $\sigma_0 = 12$ MPa would be expected. However, at low stresses, the computed internal stress in Figure 5 is 15 MPa (not shown). This value is thus very close to the predicted climb threshold. But there is no indication in the curve in Figure 5 that it is close to a threshold stress. If the low stress is close to a threshold stress, the curves would have bent down. Thus, even a threshold stress of 0.02 $\sigma_0$ is too large to be consistent with the experiments. Consequently, the meaning of the climb threshold stress is unclear.

There are other reasons why a constant threshold stress is in conflict with experiments except for some oxide dispersion strengthened (ODS) alloys. If eq. (1) is applied with a constant stress $\sigma_i$, the creep exponent would decrease with increasing applied stress and that is not observed in general. This was further discussed in the introduction. For example, in Figure 4 and Figure 5, the creep exponent increases with applied stress for all the curves. It is well established that for many particle strengthened steels, the creep strength and consequently the inverse creep rate using Monkman-Grant relation decrease approximately exponentially with increasing temperature and decreasing applied stress [36]. This is clearly inconsistent with a constant threshold stress.

In present paper, the amount of climb is used to explain the precipitation hardening mechanism. In the model, the climb rate is related to the fraction of particles and their size distribution. The actual amount of climb is based on a simple assumption by comparing the time for dislocation climb and glide. One key advantage of the present model is that it can be used to explain that the creep strength decreases exponentially with increasing temperature [15, 21, 22, 30]. Also the stress dependence of the creep rate is in agreement with observations, see Figure 4 and Figure 5.

Attempts have also been made for ODS alloys to explain the temperature and stress dependence. In [37], a model for creep in ODS alloys was formulated. Instead of a true threshold stress, the model introduced a parameter, a relaxation factor $k$, taking into account the temperature dependence of the attractive interaction between dislocation and dispersion particles. However, there were restrictions. The relaxation factor $k$ was handled as an adjustable parameter. For estimating the $k$ value, at least five creep tests under two temperatures and stresses were necessary. To increase the accuracy, even more tests were needed. Reppich [38] then improved
the attractive particle dislocation interaction theory. He treated the dislocation bypass as a serial process from detachment controlled to local climb controlled process. Instead of a constant threshold stress, it was calculated for detachment threshold and Orowan stress separately. The resulted overall threshold stress lied between 0.15 to 0.5 $\sigma_o$. However, the improvement was not dramatic and an adjustable $k$ was still used.

6. Conclusions

A previously developed model for the particle contribution to the creep strength is critically tested by comparison to published data for Cu-Co alloys. This alloy system is particularly useful for analysing precipitation hardening, because i) particles generate the main strengthening contribution, ii) only one type of particles is present homogeneously distributed, iii) the particles are stable during creep (after a suitable heat treatment), and iv) the amount of elements in solid solution is limited.

Precipitation hardening is in general the most potent way of increasing the creep strength of alloys. Consequently, an extensive literature on the topic is available. In spite of this, few predictive models are available that can describe the observations. For this reason, an attempt was made to critically test the model by Eliasson et al. Two changes in the model are made. The climb speed when particles are passed is now assumed to be controlled by the full applied stress, not just the dislocation stress. The Friedel particle spacing is used instead the planar lattice square spacing when computing the critical particle radius. The model is based on the following assumptions

- The controlling mechanism is the time it takes for a dislocation to climb across a particle.
- The key quantity of the model is the critical particle radius. Above this radius, the particles cannot be passed by climb.
- An exponential distribution of particle sizes is assumed following results for creep resistant steels. For small particles, since enough time exists for dislocations to climb, they do not contribute to the creep strength. Only particles big enough that cannot be climbed during the creep life contribute to the strength. This strength contribution is calculated with the Orowan mechanism.
- The model is fully predictive in the sense that no adjustable parameters are used.
The study gave the following results

- In some investigations, the Orowan stress is used to estimate the creep strength. It is demonstrated that this grossly overestimates the contribution to the creep strength for the Cu-Co alloys.
- In the past, quite a low value of about 0.03 $\sigma_O$ for a climb threshold stress has been derived, where $\sigma_O$ is the Orowan stress. However, even this low value is too large to be consistent with the data for one of the Cu-Co alloys.
- The model can describe the influence of applied stress, alloy composition and heat treatment for the Cu-Co alloys at least approximately. Previous studies for austenitic stainless steels demonstrate that the model can represent the temperature dependence.
- The model gives a stress and temperature dependent internal stress from the particles, which is a requirement to describe the experiments (see previous bullet). For example, a constant internal stress would give a stress exponent that decreases with increasing applied stress, which is inconsistent with observations except for some ODS alloys.

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