

# The role of cell structure during creep of cold worked copper



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## ABSTRACT

In previous work it was demonstrated that cold work could reduce the creep rate of phosphorus doped copper (Cu-OPF) by up to six orders of magnitude at 75 °C at a given applied stress. Cu-OPF will be used in canisters for final disposal of spent nuclear fuel. A dislocation model for the cell structure in the cold deformed material has been formulated. A distinction is made between the balanced dislocation content in the cell walls where the number of dislocations of opposite sign match and the unbalanced content where they do not. The recovery rate of the unbalanced content is much lower than that of the balanced content. Taking this into account, it has been possible to model the creep curves of both 12% and 24% cold worked Cu-OPF. The general appearance of the two sets of creep curves are distinctly different, which can be explained by the higher recovery rate in the 24% deformed state.

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## 1. Background

During plastic deformation of many FCC metals, cell structures are formed. The nature of the cell structure has often been characterized, but its quantitative influence on strength and other properties has been considered only to a limited extent [1,2]. In addition, there are phenomena that would be very difficult to explain if the cell structure is not taken into account. One such case is the influence of cold work on the creep rate in metals.

It is familiar that cold work can increase the creep strength, at least for strains up to 20%. This has been demonstrated in studies of austenitic stainless steels [3–5]. The deformation structure plays an important role. This is evident from the fact that the effect of cold work is reduced or removed if the material is partially or fully recrystallized during creep [6]. Also for oxygen free pure copper (Cu-OF) it has been found that cold work can increase the rupture time [7]. Wu et al. observed a dramatic increase in rupture time after cold work of Cu-OPF [8], Fig. 1. In fact, the rupture life at 75 °C is about three orders of magnitude higher after 12% cold work and about six orders after 24% cold work in tension if the results are extrapolated to the same applied stress. This corresponds to an increase in the creep strength by about 20 and 50 MPa, respectively. The increase in creep strength was, however very limited if the prestrain was performed in compression. The analysis will focus on the results in [8].

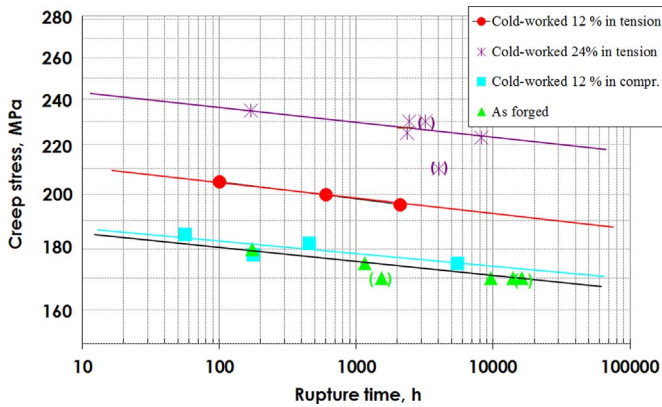
A creep test typically shows three familiar stages. Initially the deformation rate is very high. It slows down in the primary stage, reaches a stationary value in the secondary stage and accelerates

in the final tertiary stage, eventually leading to rupture. According to the established creep recovery theory [9–11], the dislocation density increases in the primary stage due to rapid work hardening for an annealed material. In the secondary stage a balance is reached between work hardening and recovery and a stationary dislocation density is obtained. In the tertiary stage, microstructure degradation for example due to the formation of cavities or reduction of the dislocation density takes place and this gives rise to an increased creep rate.

If the creep recovery theory is applied to a cold deformed material, the high initial dislocation density would be reduced in the primary stage and would in the secondary stage reach the same stationary value as for annealed material. This would give a creep rate that is the same as for the annealed material. To get a reduced creep rate in the cold deformed material, the recovery rate must be slower leaving a higher dislocation density.

In the traditional modelling of creep deformation, a homogeneous dislocation density is assumed. However, many materials form substructures during creep. After cold deformation a pronounced cell structure is often present. With an increasing amount of cold work, the cell structure gets more developed. The dislocation density in the cell walls is raised and it is reduced in the cell interiors at the same time as the cell walls become more narrow, see for example [8]. It has been suggested that the cell structure could explain the influence of cold work on creep [12]. It is known that the creep strength can be increased if the cell or subgrain size is reduced [13]. This fact is difficult to apply technically to raise the creep strength since the cell size is controlled by the applied stress. It is usually desirable to have high creep strength at low applied stress. However, when the creep stress is reduced, the cell size increases and the contribution from the

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**Fig. 1.** Creep rupture curves for Cu-OFP with and without cold work. Experimental data for 12% and 24% cold work in tension and 12% cold work in compression. For comparison data for as forged material without cold work is included. The lines are fitted to the data for easy interpretation. Data from [8].

substructure to the creep strength is lowered.

It has been found that dislocations in cell walls can be both statistically distributed and polarised [14]. Exposed to an applied stress, dislocations of different sign in the cell interiors will move in opposite directions and will end up at different places at the cell walls. This means that the sign of the dislocations can be different at opposite sides of the cell walls. It is thus quite natural that dislocations of opposite sign are located at different position after cold work. To recover these polarised dislocations they must travel through the cell walls to meet and recombine with the dislocations of opposite sign. This is a very slow process due to the back stress from the other dislocations in the cell wall. Consequently, these polarised dislocations can have a high stability during the creep process. These dislocations are generated during the cold work, but are not removed during the ordinary creep recovery process. Instead, they would represent an increased dislocation density that could explain why cold work material could have much higher creep strength. In the present paper a model for the influence of cold work on the creep rate will be developed. It will be based on the assumption that the polarised dislocations create the additional stability of the cell structure that is required to explain the increased creep strength.

The model will be applied to oxygen free copper that is alloyed with about 60 wt ppm of phosphorus (Cu-OFP). This material is planned to be used in canisters for disposal of spent nuclear fuel in Finland and Sweden. The waste package consists of a cast iron insert inside a copper canister. The sealed package will be deposited 500 m down in the bedrock in a repository [15]. The insert will take up the external load and the copper canister will give corrosion protection. Copper has a high thermodynamic stability in reducing ground water. Its stability is demonstrated by the frequent presence of native copper. The external hydrostatic pressure and the swelling pressure from the surrounding bentonite clay will also affect the copper canister and it will be exposed to creep [16,17]. It was observed already many years ago that oxygen free copper without phosphorus Cu-OF could have very low creep ductility, sometimes below 1% [18]. For this reason Cu-OF was replaced by Cu-OFP as canister material. Many creep tests have demonstrated that Cu-OFP has satisfactory creep ductility [19]. The purpose of the present paper is to model the influence of cold work on creep results of Cu-OFP by taking the distribution of dislocations in the substructure into account.

## 2. Model for stress strain and creep curves

### 2.1. Dislocation model

Stress strain curves and creep properties of Cu-OFP have been modelled successfully in several papers [20–22]. These models will serve as a basis when cell structure is explicitly taken into account in the next section. A brief summary of the models will be given here. The models are based on the development of the dislocation density  $\rho$  during plastic deformation

$$\frac{d\rho}{d\varepsilon} = \frac{m}{bL} - \omega\rho - 2\tau_l M\rho^2 / \dot{\varepsilon} \quad (1)$$

$\varepsilon$  is the strain,  $m$  the Taylor factor,  $b$  Burger's vector and  $L$  the "spurt" distance which the dislocation moves in each elementary release during deformation.  $L$  is related to the dislocation density as  $L = c_L / \sqrt{\rho}$ , where  $c_L$  is a work hardening constant.  $\omega$  is a constant that controls the amount of dynamic recovery.  $\tau_l$  is the dislocation line tension, and  $M$  the dislocation mobility, which is given by

$$M(T, \sigma) = M_{OF}(\sigma - \sigma_{break}, T) f_Q \quad (2)$$

where

$$M_{OF}(T, \sigma) = \frac{D_{s0} b}{k_B T} \frac{ab^3}{e^{k_B T}} e^{-\frac{Q}{RT}} \left[ 1 - \left( \frac{\sigma}{\sigma_{imax}} \right)^2 \right] \quad (3)$$

$$f_Q = e^{-U_p^{max}/RT} \quad (4)$$

$T$  is the absolute temperature,  $\sigma$  the applied stress,  $D_{s0}$  the preexponential coefficient for self-diffusion,  $Q$  the activation energy for self-diffusion,  $k_B$  Boltzmann's constant,  $R$  the gas constant, and  $\sigma_{imax}$  a stress that is taken as the tensile strength at room temperature.  $U_p^{max}$  is the maximum interaction energy between a P solute and a dislocation [23,24]. The dislocation mobility in Eq. (3) takes both climb and glide into account. Glide is included in the factor in the exponent involving  $\sigma_{imax}$ . If  $\sigma_{imax}$  is assumed to be infinite the climb mobility of dislocations originally derived by Hirth and Lothe is recovered [25]. All the parameter values in Eqs. (1)–(4) have been derived or are available from literature [20–22] and none is used for fitting to the mechanical test data.

Eq. (1) describes how the dislocation density is changed as a function of strain  $\varepsilon$ . The first term on the right hand side takes the increase due to work hardening into account. When dislocations of opposite sign get close enough they will combine into low energy configurations [1,26]. This is referred to as strain controlled or dynamic recovery. This is taken into account in the second term. Dislocations of opposite sign attract and annihilate each other in a slow time controlled process that is referred to as static recovery [11]. This is the reason for the third term.

The amount of phosphorus is quite low (about 60 wt ppm) in the canister Cu-OFP and it is entirely in solid solution. In spite of this, it generates a large increase in the creep strength. The origin of this effect is the formation of Cottrell atmospheres of phosphorus atoms around the dislocations. At the dislocations the concentration of phosphorus atoms can be a factor 100 higher than in the bulk. A stress  $\sigma_{break}$  is required for the dislocations to break away from these atmospheres [23]. In addition, there is an increase in the activation energy for the plastic deformation by  $U_p^{max}$ . This is taken into in the expression for the dislocation mobility in Eq. (2).

### 2.2. Stress strain curves

An expression for a stress strain curve can be derived from Eq. (1). Usually it is sufficient to take the first two terms on the right

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