Temperature Dependent Relaxation Behavior of Pressureless and Pressure Assisted Sintered Silver

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Abstract:
In this study, we investigate the creep properties of pressureless and pressure assisted sintered silver. Based on density analysis and tensile strength, the relaxation behavior of the series was observed at room temperature, at 130°C and at 200°C. A material model for mechanical properties of porous metals was derived from experimentally established time dependent material response under tensile load.

1. INTRODUCTION

Advancements in materials science allowed an increase of packaging density in power electronics applications to an incredible extent. The needs for lowering weight, size, and thermal management cost have raised the demand for packaging concepts allowing for higher operation temperature and improved predictability of lifetime and reliability of electronic assemblies. While glass fiber reinforced epoxy resin-based materials with a glass transition temperature of 200°C and more are available since recent years, interconnection techniques for such high temperatures are still a challenging hurdle. One promising industrially applicable alternative to soldering with high-lead containing solders is silver sintering. Much work has been spent in the past to explore the mechanical properties of bonding materials like SAC solder and high-lead containing solder including their temperature dependencies [1]. In particular, the conspicuous creep behavior of sintered silver layers resulting from its porous structure has been investigated [2-3] and proved to be stress relaxant. However, in order to take full advantage of silver sintering for industrially mass-produced electronic assemblies sufficiently accurate mechanical properties data need to be available during the design phase.

This work aims in formulating a material model ready for implementation in commercial finite element packages (e.g. Abaqus, ANSYS, GetFEM++) allowing to study time dependent loading cases of sintered silver layers in electronic assemblies. Starting point is a simple well-known creep model based on a Norton power law, where the tensile stress relaxation is described as the time derivative of the strain $\dot{\varepsilon}$ by

$$\dot{\varepsilon} = C \cdot e^{-Q/kT} \cdot q^n$$

(1)

where $C$ is a constant, $e^{-Q/kT}$ is called Arrhenius term (with the activation energy $Q$, the Boltzmann constant $k$ and the absolute temperature $T$), $q$ is the von Mises stress, and $n$ is the stress exponent. By applying this model to measurement results of the present study, features and limitations of this simple model are discussed. Furthermore, the quasi-static Gurson - Tvergaard - Needleman plasticity model [4,6,7] is extended to a creep model, whereby material parameters of the model are obtained from fits to tensile stress relaxation experiments results performed with sintered silver layers.

2. SAMPLE PREPARATION

A commercial silver paste (Heraeus mAgic 338) was used to prepare dumbbell-shaped Ag samples for tensile testing as seen in Figure 1 a-b. Specimens A-type and B-type were sintered pressureless at temperatures of 230°C and 300°C, respectively, C-type specimen were pressure sintered at 70 MPa and 230°C. For all series a sintering duration of 30 minutes was used. The average thicknesses of the Ag layers of type A, B, and C after sintering were 135 µm, 105 µm, and 75 µm. Their different structures are shown in the scanning electron microscope (SEM) images of fracture surfaces with the same magnification in Figure 2.
Sintering without pressure leads to rather rounded Ag-colloids of ca. 1 µm grain size (Fig. 2.a-b), while sintering under high pressure leads to a much denser microstructure with closed pores (Fig. 2.c).

In Table 1 the average densities $\rho$ of specimens of the three series are listed as ratio of mass $m$ and sample volume $V$

$$\rho = \frac{m}{V} \quad (2)$$

and as percentage of the pure Ag volume and the sample volume. The latter one was determined using a 3D-microscope of Keyence.

### 3. TENSILE STRENGTH AND STRESS RELAXATION

Due to unavoidable variations in preparing the samples their tensile strength values were slightly different. In order to define the stress peak levels for performing the relaxation tests, the average tensile strength of the specimen series was taken into account.

The tensile tests for several samples to each series (A, B, C) were measured at three temperatures: 25°C, 130°C and 200°C with a testing speed of 1 mm/min. In Table 2 the mean tensile values $\sigma_{\text{mean}}$ and corresponding stress peaks $\sigma_{\text{peak}}$ for relaxation in average are listed.

In Figure 3 representative stress-strain curves of single tests for the series are shown to outline the brittle fraction at room temperature (Fig.3.a) and the ductile behavior at elevated temperature (Fig.3.b).

To observe the creep behavior, the samples were stretched to the defined load according $\sigma_{\text{peak}}$ and then the decay of stress was measured as a function of time for 90 minutes, as shown in Figure 4.
Fig. 3. Representative stress-strain curves of tensile strength for each series A (blue), B (green), C (pink/red) at testing speed of 1 mm/min and temperatures of 25°C (a) and 200°C (b), whereby (a) shows a brittle and (b) a ductile fracture behaviour.

For the pressureless (A- and B-type) and under high pressure (C-type) sintered silver, the average end-percentages of initial $\sigma_{\text{peak}}$ (100%) are listed in Table 3. The relaxation of the experimental curves could be implemented according to Norton creep described in the introduction section (1). The derived stress exponents from single test curves are used to deduce mean values for $n$, which were between 30-35 for the A-series, and 15-17 for the B and C series at 25°C.

Table 2: Average tensile strength values $\sigma_{\text{mean}}$ and average stress peak level $\sigma_{\text{peak}}$ for relaxation

<table>
<thead>
<tr>
<th>A: 230°C, 0 MPa</th>
<th>25°C</th>
<th>130°C</th>
<th>200°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_{\text{mean}}$ Tensile Strength</td>
<td>17 MPa</td>
<td>11 MPa</td>
<td>11 MPa</td>
</tr>
<tr>
<td>$\sigma_{\text{peak}}$ Av. Relaxation Stress</td>
<td>10 MPa</td>
<td>6 MPa</td>
<td>8 MPa</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>B: 300°C, 0 MPa</th>
<th>25°C</th>
<th>130°C</th>
<th>200°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_{\text{mean}}$ Tensile Strength</td>
<td>17 MPa</td>
<td>17 MPa</td>
<td>15 MPa</td>
</tr>
<tr>
<td>$\sigma_{\text{peak}}$ Av. Relaxation Stress</td>
<td>11 MPa</td>
<td>12 MPa</td>
<td>9 MPa</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>C: 230°C, 70 MPa</th>
<th>25°C</th>
<th>130°C</th>
<th>200°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_{\text{mean}}$ Tensile Strength</td>
<td>57 MPa</td>
<td>42 MPa</td>
<td>42 MPa</td>
</tr>
<tr>
<td>$\sigma_{\text{peak}}$ Av. Relaxation Stress</td>
<td>43 MPa</td>
<td>32 MPa</td>
<td>32 MPa</td>
</tr>
</tbody>
</table>

In general, a higher merit indicates a lower relaxation creep. At testing temperature of 130°C $n$ was 5-8 for pressureless and 8-9 for pressure assisted sintered samples, and respectively 2-4 and 4-5 at 200°C. The more porous samples show a varying relaxation profile at elevated temperatures compared to room temperature. Unlike the already dense samples (C), the limber specimen (A, B) have more scope for displacement creep, especially with the additional thermal energy. This might lead to a posterior hardening effect with subsequent stress resistance at higher temperatures. Therefore, for porous structure more relaxation may occur, where the range of tight samples is more confined with increasing temperature.

A modified material model based on Norton creep but also inelastic behavior of the porous Ag based on Gurson plasticity should be implemented to interpret the experimental results.

Table 3: Average end-percentages $\sigma_{\%}$ after relaxation for 90 minutes from the initial stress peak of 100% for the three series A, B, C and at three test temperatures

<table>
<thead>
<tr>
<th>Test Temp.</th>
<th>25°C</th>
<th>130°C</th>
<th>200°C</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>78%</td>
<td>12%</td>
<td>2%</td>
</tr>
<tr>
<td>B</td>
<td>67%</td>
<td>12%</td>
<td>5%</td>
</tr>
<tr>
<td>C</td>
<td>65%</td>
<td>34%</td>
<td>18%</td>
</tr>
</tbody>
</table>
4. **Unified Model of Plasticity and Creep**

The density measurements of sintered silver summarized in Table 1 indicate considerable porosity of the materials. From an investigation of McClintock [5] it is known that porosity has strong influence on the plastic behavior of metals. Around pores stress concentrations are formed during ongoing deformation. Therefore, the strength of a porous metal is reduced compared to bulk material. In conclusion, Gurson [4] defined a quasi-static flow rule for porous metals, where von Mises stress and hydrostatic pressure are both influencing the plastic strain. In the present approach, this flow rule is extended to time dependent inelastic behavior. Thereby, the Norton creep equation is reformulated to include hydrostatic pressure in addition to von Mises stress. It is demonstrated that the material parameters of the model can be fitted to the stress relaxation experiments of the present study.

4.1. **Gurson plasticity and Norton creep**

We use the Gurson - Tvergaard - Needleman model [4, 6, 7] as starting point of our theoretical interpretation. The flow rule of this model was defined as

\[
\frac{q}{\sigma_y} \cdot cosh\left(\frac{-3q_p}{2\sigma_y}\right) = (1 + q_3f^2) = 0 \quad (3)
\]

where \( q \) is the von Mises stress, \( \sigma_y \) is the flow stress of the dense material, \( p \) is the hydrostatic pressure stress and \( f \) is the porosity, i.e. the volume fraction of the pores in the material. Further, \( q_1 \), \( q_2 \) and \( q_3 \) are material constants. In the limit of \( f = 0 \) for dense materials, the model reduces to von Mises plasticity. On the other hand, for materials of high porosity the hydrostatic pressure contributes significantly to the plastic deformation behavior.

However, the quasi-static flow rule (3) cannot explain the stress relaxation behavior of Figures 4 (a-c). Therefore, it is here intended to develop a creep model, which combines the principles of Norton creep and Gurson plasticity. This model shall be used for Finite Element computer simulations. Thereby, a simplification of the Gurson model is suggested in order to obtain a model which is computationally inexpensive. The hyperbolic cosine function of equation (3) can be expanded into a Taylor series, and for small strains one may truncate the Taylor series after the term of second order. This leads to an approximation, where the hyperbolic cosine is replaced...
by a term, which is quadratic in $p$. Consequently, the flow condition under hydrostatic stress may to a good approximation be treated by analogy to flow under deviatoric stress, as will be shown in the subsequent section.

4.2. Definition of the constitutive model

In the following the orthogonal decomposition of the Cauchy stress tensor $\sigma_{ij}$ into hydrostatic and deviatoric stress

$$p = \frac{1}{3} \text{tr}(\sigma_{ij})$$

and deviatoric stress

$$S_{ij} = \sigma_{ij} + p \cdot I$$

is utilized, where $I$ is the identity matrix and $\text{tr}$ denotes the trace of a matrix. By analogy, the strain tensor may additively be decomposed into volumetric and deviatoric parts according to

$$\varepsilon_{ij} = \frac{1}{3} \text{tr}(\varepsilon_{ij}) \cdot I + \varepsilon_{ij}^\text{dev}$$

In short, we hereafter write

$$\varepsilon_{\text{vol}} = \text{tr}(\varepsilon)$$

for the volumetric strain. Furthermore, the decomposition of the total strain tensor

$$\varepsilon_{\text{tot}} = \varepsilon_{\text{el}} + \varepsilon_{\text{ine}}$$

into elastic and inelastic strains will be used. Thus, the strain rates

$$\|\varepsilon_{\text{inel}}\| = c_1 \cdot f \cdot e^{-Q/kT} \cdot |p|^{n_1}$$

and

$$\|\varepsilon_{\text{dev}}\| = c_2 \cdot e^{-Q/kT} \cdot \|S\|^{n_2}$$

are defined as constitutive equations of this model. The direction of inelastic flow under pressure is compressive, while flow under deviatoric stress is parallel to the deviatoric stress tensor $S_{ij}$. In equations (9 a, b), $|p|$ denotes the absolute value of the scalar $p$, and $\|S\|$ is the Euclidean norm of the tensor $S$. $Q$ is the activation energy, $n_1$ and $n_2$ are the stress exponents related to hydrostatic and deviatoric parts of the stress, respectively. $c_1$ and $c_2$ are material constants. The material parameters show a dependency on the sintering conditions. It should also be noticed that a change of volumetric strain during inelastic deformation causes a change of porosity $f$. Consequently, conservation of mass implies the evolution equation

$$\dot{f} = (1 - f) \cdot \dot{\varepsilon}_{\text{vol}}$$

4.3. Numerical fitting

Stress relaxation experiments were performed under conditions of time dependent uniaxial tensile stress

$$\sigma_{ij}(t) = \begin{pmatrix} \sigma_{11}(t) & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

Hence, the time dependence of hydrostatic pressure writes as

$$p(t) = \frac{1}{3} \sigma_{11}(t)$$

and the time dependence of the tensor norm of the deviatoric stress evaluates to

$$\| S_{ij}(t) \| = \frac{2}{3} \sigma_{11}(t)$$

Further, both, volumetric and deviatoric parts of the strain rate contribute to the rate of the inelastic strain component $\varepsilon_{11}^{\text{ine}}$ according to

$$\dot{\varepsilon}_{11}^{\text{ine}} = \frac{1}{3} \dot{\varepsilon}_{\text{vol}} + \frac{2}{3} \| \dot{\varepsilon}_{\text{dev}} \|$$

During stress relaxation, the total strain $\varepsilon_{11}^{\text{tot}}$ of the sample was constrained to a constant value, while the inelastic strain $\varepsilon_{11}^{\text{ine}}$ increased on the expense of elastic strain $\varepsilon_{11}^{\text{el}}$. Consequently, it is concluded that

$$\dot{\varepsilon}_{11}^{\text{el}} = -\dot{\varepsilon}_{11}^{\text{ine}}$$

Finally, the rate of the stress $\sigma_{11}$ is related to the rate of elastic strain $\varepsilon_{11}^{\text{el}}$ by

$$\dot{\sigma}_{11} = E^{\text{eff}} \cdot \dot{\varepsilon}_{11}^{\text{el}},$$

where $E^{\text{eff}}$ is the effective Young’s modulus of the porous material. Thus, we are in the position to express the time dependence of stress $\sigma_{11}$ as ordinary differential equation of first order. The stress rate $\dot{\sigma}_{11}$ is a function of the strain rates (9 a, b), which depend on the value of $\sigma_{11}$ and on the fit parameters of the model.

The activation energy $Q$ of creep was found to be 133 kJ/(mol K). The remaining fit parameters of this material model were considered as temperature dependent: The best fits are shown in Figure 4 (a-c). The fit parameters used are summarized in Table 4. In spite of obvious agreement between experimental results and model fits, it is here suggested to perform additional experiments under pure deviatoric stress in order to obtain unique values for the material parameters.
Table 4: Material parameters used for numerical fits shown in Fig 4 a-c. Stresses are in units of MPa.

<table>
<thead>
<tr>
<th>Series A</th>
<th>c₁</th>
<th>c₂</th>
<th>n₁</th>
<th>n₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>RT</td>
<td>96</td>
<td>2.08 · 10⁻¹⁵</td>
<td>10</td>
<td>35</td>
</tr>
<tr>
<td>130°C</td>
<td>1.08 · 10¹²</td>
<td>8.74 · 10⁻³</td>
<td>6</td>
<td>10</td>
</tr>
<tr>
<td>200°C</td>
<td>2.26 · 10⁹</td>
<td>2.45</td>
<td>3</td>
<td>7.9</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Series B</th>
<th>c₁</th>
<th>c₂</th>
<th>n₁</th>
<th>n₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>RT</td>
<td>3.59 · 10⁻¹²</td>
<td>2.08 · 10⁻¹⁵</td>
<td>8</td>
<td>35</td>
</tr>
<tr>
<td>130°C</td>
<td>1.21 · 10⁻¹⁰</td>
<td>8.74 · 10⁻³</td>
<td>6</td>
<td>10</td>
</tr>
<tr>
<td>200°C</td>
<td>6.78 · 10⁹</td>
<td>2.45</td>
<td>4</td>
<td>7.9</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Series C</th>
<th>c₁</th>
<th>c₂</th>
<th>n₁</th>
<th>n₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>RT</td>
<td>3.29</td>
<td>4.16 · 10⁻⁸¹</td>
<td>16</td>
<td>62.2</td>
</tr>
<tr>
<td>130°C</td>
<td>2.3 · 10⁴</td>
<td>1.22 · 10⁻²</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>200°C</td>
<td>1.29 · 10⁷</td>
<td>3.92</td>
<td>3</td>
<td>7.9</td>
</tr>
</tbody>
</table>

5. SUMMARY-CONCLUSION

To analyze the creep behavior of porous silver as interconnection material in packaging technology, pressureless and pressure assisted sintered-Ag samples were produced. The mass/volume densities of the sample series were determined (35% - 62%) and the ultimate tensile strength was measured at room and elevated temperatures. In order to observe the creep behavior at various testing temperatures (25°C, 130°C and 200°C), samples were exposed to uniaxial stress and the stress relaxation over time at constant sample length was monitored.

The theoretical interpretation of the material behavior was based on a combination of Norton creep and Gurson plasticity. Owing to the high porosities of the samples, it was necessary to consider the influence of hydrostatic pressure on plastic deformation. A constitutive material model was formulated, which makes use of an orthogonal decomposition of stress and strain tensors into volumetric and deviatoric parts. Consequently, the time dependent inelastic deformation of the material was described by a system of two equations. It was demonstrated that the material parameters of the model can be fitted to stress relaxation experiments. Nevertheless, it was suggested to perform additional experiments under pure deviatoric stress to obtain unique solutions of the fit parameters. For this purpose, data of stress relaxation experiments under shear stress is already planned to be implemented in a subsequent publication.

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REFERENCES


