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High temperature compression of Mo(Si,Al)$_2$-Al$_2$O$_3$ composites

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**A R T I C L E  I N F O**

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High temperature compression
Electron backscatter diffraction
Electroplasticity

**A B S T R A C T**

The aim of this study was to investigate the effect on high temperature of mechanical properties of adding Al$_2$O$_3$ particles to polycrystalline Mo(Si,Al)$_2$. Mo(Si,Al)$_2$-Al$_2$O$_3$ composites, containing 0–25 wt% Al$_2$O$_3$ particles have been compression tested at 1300 °C, and the microstructure after deformation was studied using electron backscatter diffraction. It was shown that even small amounts (5 wt%) of Al$_2$O$_3$ particles resulted in a grain-refined material through inhibition of grain growth during sintering, which lead to lower flow stresses compared to the coarse-grained Al$_2$O$_3$-free material. The inverse grain size effect and post-test microstructure investigations suggest that creep-like deformation mechanisms dominate in fine grained Mo(Si,Al)$_2$-Al$_2$O$_3$ composites at 1300 °C. In the materials containing 5–15 wt% Al$_2$O$_3$, the maximum stress decreased with increasing Al$_2$O$_3$ content. In materials with higher Al$_2$O$_3$ additions, the maximum stress increased with the Al$_2$O$_3$ addition, but did not reach the strength levels in the Al$_2$O$_3$-free reference material. It is suggested that the deformation behaviour is affected by electroplasticity effects as resistive heating was used. Electroplasticity contributes to the decrease in maximum stress observed in the lower Al$_2$O$_3$ containing materials, while this is outweighed by particle strengthening at higher Al$_2$O$_3$ contents.

1. Introduction

High temperature heating processes (above 1000 °C), for example within the steel industry, give rise to significant greenhouse gas emissions [1]. Most of the energy used for the processes is produced through combustion of fossil fuels [2], and their electrification therefore holds a great promise to reduce the greenhouse gas emission. Electric alternatives, for example indirect resistive heating using heating elements, are already used in small furnaces, operating in the MW rather than in the kW range. However, this requires an up-scaling of the size of the heating elements and probably a more complicated mounting geometry (horizontal rather than vertical) of the elements in the furnace. This will increase the demands on the mechanical properties of the heating elements since the gravitational force on them increases.

The choice of heating element material depends on the required temperature and operating environment. A material often used for high temperature heating elements in small furnaces is Mo(Si,Al)$_2$. It is a ceramic material with high melting temperature, high resistivity and excellent corrosion resistance in both oxidizing and reducing environments [3]. The latter makes it a suitable material choice for many industrial heating processes. While the oxidation properties of Mo(Si,Al)$_2$ have been thoroughly investigated [4–10], research on high temperature mechanical properties of polycrystalline Mo(Si,Al)$_2$ is scarce. Instead, most current knowledge is derived from tests of single crystals. It has been shown that Mo(Si,Al)$_2$, which possesses a hexagonal crystal structure (Strukturbericht Designation C40), only has one possible slip system, $\{21\bar{1}0\}$ $(0001)$, which is activated above 1100 °C [11]. However, due to the complex stress state and the influence of grain boundaries, the deformation mechanisms in polycrystalline materials are expected to be more complicated than in single crystals.

When it comes to polycrystalline C40-structured Mo(Si,Al)$_2$ very few studies have been reported. Hagihara et al. [12] and Umakoshi et al. [13] studied the temperature dependence on yield strength and fracture strength at elevated temperature (800–1600 °C). A ductile-brittle transition temperature (DBTT) of 1250 °C was reported. The strength levels were similar to C11$_{\parallel}$-structured MoSi$_2$ and fracture strains exceeding 20% were observed above the DBTT [12,13]. However, no complete stress–strain curves were presented. Nihara et al. [14] and Suzuki et al. [15] have studied the flexural strength of nanostructured Mo(Si,Al)$_2$ at temperatures up to 1300 °C. Common for the studies is that the microstructures after deformation were not investigated.

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In our previous study of polycrystalline Mo(Si,Al)₂ tested in compression at 1300 °C, we showed that a rapid softening took place after yielding [16]. We observed large variations in crystal orientations within the grains, which suggests that intragranular deformation through dislocation slip was an active deformation mechanism. The formation and movement of dislocations also gave rise to formation of low angle grain boundaries (LAGBs) within the grains. Furthermore, dynamic recrystallization (DRX), driven by the stored energy (dislocation density), took place during deformation.

It is suggested that particle strengthening could be used to improve the mechanical properties of Mo(Si,Al)₂. The particles are expected to pin the grain boundaries during sintering, which gives rise to a more fine-grained material [17–20]. A smaller grain size would, according to the Hall–Petch relationship, improve the strength of the material. On the other hand, a reduction in grain size has been shown to promote grain boundary sliding in other disilicides, but it has also been reported that addition of hard particles could suppress such sliding [18,21]. While C11₃-structured MoSi₂ has been the most extensively studied silicide-based material with respect to particle strengthening [14,15,17–19,22–24], Niihara et al. [14] and Suzuki et al. [15] have tested the flexural strength (three point bending test, from room temperature up to 1300 °C) of a Mo(Si,Al)₂–SiC nanocomposite. The chemical composition of the matrix phase was Mo₅₃.₄Si₇₅.₄Al₅.₅ (lower Al content compared to the material investigated in our present, and previous study [16]) and consisted of both C11₃ (84.9 vol.%) and C40 (15.1 vol.%). It was shown that the flexural strength was somewhat higher than that of a SiC-free Mo₅₃.₄Si₇₅.₄Al₅.₅ nanomaterial. The microstructure of the deformed material was not presented. However, no studies have been conducted in the case of a non-nanostructured C40 Mo(Si,Al)₂, even though particle strengthening has been used with success in other disilicides.

In this study we investigate the effect of Al₂O₃ additions on the high temperature mechanical properties of C40 Mo(Si,Al)₂. Al₂O₃ was selected because Al is already present in the material and will not affect the oxidation behaviour. Compression tests were performed at 1300 °C using a Gleeble thermomechanical simulator. The test specimens were heated by resistive heating. We show that Al₂O₃ additions lead to more fine-grained materials, in which no DRX or formation of LAGBs occur during compression. The strength of the fine-grained Al₂O₃-containing materials was lower than that of a coarse-grained Al₂O₃-free reference, suggesting that creep-like deformation mechanisms, such as grain boundary sliding, dominate. For materials containing 5–15 wt% added Al₂O₃, the maximum stress decreases with increased Al₂O₃ content. We suggest this could be related to electroplasticity. At higher volume fractions, the particle strengthening outweighs the strength reduction due to electroplasticity.

2. Materials and methods

2.1. Materials

Polycrystalline Mo(Si,Al)₂ was prepared by dry mixing of elemental Mo (99.9% purity, Cerac Inc.), Si (99.99% purity, Wacker) and Al (99.5% purity, GoodFellow) powders in a Mo-lined ball mill with Mo balls. The powder mixture was heated in inert gas to initiate a self-propagating reaction in which Mo(Si,Al)₂ (as well as small amounts of Mo₅(Si,Al)₁) and Al₂O₃ was formed. Mo₅(Si,Al)₁ is formed as a result of local variations in the unreacted Mo-Si-Al powder. According to [25], an Al-Si eutectic forms when the powder is heated. This leads to regions with lower Si and Al content, which promotes the formation of Mo₅(Si,Al)₁ rather than Mo₅(Si,Al)₂. Furthermore, Al is consumed by the formation of Al₂O₃ during the reaction, which further lowers the Al content of the unreacted powder. The reaction product was milled in ball mill for 228 h to obtain a fine-grained powder with an average particle size of around 5 μm. Five Mo(Si,Al)₂-Al₂O₃ composites, containing 5, 10, 15, 20 and 25 wt% Al₂O₃ respectively, were prepared by adding Al₂O₃ particles (AKP-53, Sumitomo Chemical, purity: 99.99%, mean particle size: 0.17 μm), to the milled Mo(Si,Al)₂ powder, see Table 1. The powders were milled for 1 min in a vibratory disc mill to prevent agglomeration of Al₂O₃ particles. A reference material, without added Al₂O₃, was also prepared. The powders were compacted to rods using cold isostatic pressing (CIP, EPSI) operating at 2000 bar, prior sintering for 1 h at 1650 °C in H₂ gas in a tube furnace (ramp time from room temperature was 5 h). The as-sintered materials had densities of 97%–98%, of the theoretical values, see Table 1.

2.2. Materials characterization

An FEI Quanta 200 FEG ESEM scanning electron microscope (SEM) was used for microstructure imaging and chemical composition analysis using energy-dispersive X-ray spectroscopy (EDS). An acceleration voltage of 20 kV was used. Electron backscatter diffraction (EBSD) was used to obtain crystal orientation and grain size data. A Tescan Gaia 3 SEM, operating at 15 kV, was used for this purpose. The EBSD data were analysed in MTEX [26], and the grain size was calculated as the equivalent sphere diameter, \( d = 2 \sqrt{A/\pi} \), where \( A \) is the grain area. Grain reference orientation (GROD) maps were used to quantify the internal misorientations within grains, which is related to the density of geometrically necessary dislocations (GNDs). X-ray diffraction (XRD), using a Cu-anode equipped Bruker D8 Advance, was, along with EBSD, used for phase identification.

2.3. Compression testing

The as-sintered Mo(Si,Al)₂-Al₂O₃ composite rods were ground to a diameter of 8 mm and cut into 12 mm long cylindrical specimens for compression testing. The specimens were tested with a strain rate of \( 10^{-4} \) s⁻¹ at 1300 °C using a Gleeble 3800 thermomechanical simulator. The temperature was measured using a pyrometer. Graphite foils and Ni-paste were used between the specimen and the anvils to minimize friction and ensure good electrical contact. More complete details and calibration of the experimental set-up are published elsewhere [16]. At least three specimens of each composite were tested.

3. Results

The mechanical properties and microstructure of the Al₂O₃-free material, A0, has been investigated in a previous paper [16], therefore, the results from this material will only be described in brief in this present paper.

3.1. Microstructure of as-sintered material

Backscattered electron (BSE) micrographs and EBSD phase maps of the as-sintered materials are shown in Fig. 1. All as-sintered materials consisted of three phases: Mo(Si,Al)₂, Mo₅(Si,Al)₁ and \( α\)-Al₂O₃. The chemical composition of the phases in A0 can be seen in Table 2. The composition of phases in the other materials are similar to those in A0. Since small amounts of Al₂O₃ was formed in the self-propagating reaction, also A0 contains Al₂O₃ (1.7 vol.%). The EBSD maps in Figs. 1
Fig. 1. BSE micrographs and EBSD phase maps of as-sintered materials, (a) A0, (b) A5, (c) A10, (d) A15, (e) A20, (f) A25, (g) cut of rod for microscopy study, (h) XRD diffractogram of A0-A25.

Table 2
EDS results on the chemical composition of the phases in A0.

<table>
<thead>
<tr>
<th>Phase</th>
<th>Chemical composition [at.%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mo(Si,Al)₂</td>
<td>32.4 36.6 28.8 2.3</td>
</tr>
<tr>
<td>Mo₅(Si,Al)₃</td>
<td>57.2 32.6 7.3 2.8</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>0.8 1.7 39.6 58.0</td>
</tr>
</tbody>
</table>

and 2 shows that the grain size is much finer in A5-A25 (around 5 μm) compared with A0 (around 21 μm), see Table 3. This indicates that the addition of Al₂O₃ particles have a grain boundary pinning effect during sintering, which is in line with previous research on other disilicide composites [18,27]. The grain sizes are similar to the as-milled powder, and there is only a very slight decrease in grain size with increasing Al₂O₃ content (Table 3), suggesting that already the lowest levels of additions are sufficient to effectively pin the boundaries.

The Al₂O₃ particles in A5-A25 are larger (but typically less than 5 μm) than those formed from the self-propagating reaction (from nm up to 2–3 μm in size), compare Fig. 1(a) and (b)-(f), and located at Mo(Si,Al)₂ grain boundaries. The grey regions in the EBSD maps are unindexed Mo(Si,Al)₂ (some orientations are difficult to index even though the EBSD patterns were clear and belonged to a hexagonal unit cell) and Mo₅(Si,Al)₃ grains. The chemical composition of the unindexed grains agreed with Mo(Si,Al)₂ and Mo₅(Si,Al)₃, respectively. The low volume fraction of Mo₅(Si,Al)₃ in A0-A25 is not expected to affect the mechanical properties of the composites to a great extent. Furthermore, if there is an affect, it should be similar in all materials presented in this paper because of the similar volume fractions. Therefore, the EBSD analysis presented in this paper is limited to the Mo(Si,Al)₂ phase only. The amount of porosity is low and similar in all materials and should not have a major effect on the strength. Furthermore, in particular as the samples are tested in compression.

The chemical composition of the phases is shown in Table 2. The O content in Mo(Si,Al)₂ and Mo₅(Si,Al)₃ is likely due to the presence of nano-sized Al₂O₃ particles dispersed in the two phases. Such particles have been found in our previous study of the A0 material [16] using SEM and transmission electron microscopy (TEM), and have been reported in studies on a similar material by Ingemarsson et al. [4,6,7]. The unstoichiometric composition of Al₂O₃ and the detection of Mo and
Table 3
Volume fractions, grain size and density (as percent of theoretical density) for the different materials.

<table>
<thead>
<tr>
<th>Volume fraction [%]</th>
<th>Mo(Si,Al)</th>
<th>Mo(Si,Al)₂</th>
<th>Al₂O₃</th>
<th>Pores</th>
<th>Mo₃(Si,Al)</th>
<th>Grain size μm (std)</th>
<th>% of theoretical density</th>
</tr>
</thead>
<tbody>
<tr>
<td>A0</td>
<td>94.2</td>
<td>3</td>
<td>1.7</td>
<td>1.1</td>
<td>21.41 (13.41)</td>
<td>95.3</td>
<td></td>
</tr>
<tr>
<td>A5</td>
<td>86.8</td>
<td>5.3</td>
<td>7.8</td>
<td>0.1</td>
<td>7.05 (3.30)</td>
<td>96.9</td>
<td></td>
</tr>
<tr>
<td>A10</td>
<td>81.1</td>
<td>4.7</td>
<td>14.1</td>
<td>0.1</td>
<td>5.12 (3.19)</td>
<td>97.9</td>
<td></td>
</tr>
<tr>
<td>A15</td>
<td>76.2</td>
<td>3.9</td>
<td>19.7</td>
<td>0.2</td>
<td>4.81 (2.41)</td>
<td>97.9</td>
<td></td>
</tr>
<tr>
<td>A20</td>
<td>67.8</td>
<td>3.7</td>
<td>26.9</td>
<td>1.5</td>
<td>3.80 (1.74)</td>
<td>97.7</td>
<td></td>
</tr>
<tr>
<td>A25</td>
<td>64.8</td>
<td>2.4</td>
<td>31.2</td>
<td>1.7</td>
<td>5.45 (2.21)</td>
<td>97.0</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 2. Grain size distribution of as-sintered and deformed materials. Average grain size in legends.

Si in this phase is probably due to large interaction volume in SEM, and/or Mo and Si traces remaining from the synthesis.

3.2. Deformation behaviour

All specimens were plastically deformed when tested in compression at 1300 °C. Stress–strain curves of all material are shown in Fig. 3. The final strain differ between the specimens as a consequence of the experiment design, which was designed to include only the peak stress and the subsequent softening. The compression was manually stopped when the stress reached low values, typically between 20 and 40 MPa. The stress–strain curves from all specimens are found in supplementary materials. Some of the curves in this figure showed abrupt stress drops and lower maximum stress (for example the curves labelled A5.2, A10.3 and A15.4 in Fig. S1) than other specimens of the same material. The drops are probably related to defects, such as pores or flaws, present in the materials. The specimens giving rise to such curves were not selected for further investigations.

In the initial part of the deformation, all tested specimen showed linear behaviour before plastic deformation begins. After reaching the maximum stress, the materials soften rapidly. The details of the stress–strain curve obtained from A0 has been reported in [16], and here we focus on samples A5-A25. Compared with A0, the stress–strain curves of the Al₂O₃-added materials deviate from linearity at lower
stress levels and the maximum stresses of the Al$_2$O$_3$-added materials are lower. This decrease in flow stress deviates from the grain size-strength relationship described by the Hall–Petch relationship. This indicates that a large grain boundary area is deleterious for the strength of polycrystalline Mo(Si,Al)$_2$. This in turn, indicates that the materials deform by creep-like mechanisms at 1300 °C.

The maximum stresses obtained from the compression tested specimens as a function of Al$_2$O$_3$ content are shown in Fig. 4. While the scatter in the A0 data was relatively small, all Al$_2$O$_3$-added materials showed larger scatter. Despite the scatter, a “U”-shaped trend of the maximum stress can be observed. In the case of A5-A15, the maximum stress decreases as the Al$_2$O$_3$ content increases, while it increases as the Al$_2$O$_3$ content is further increased.

3.3. Microstructure of deformed specimens

The specimen corresponding to the stress–strain curves in Fig. 3 were selected for microstructure investigations. The specimens were cut parallel to the compression axis, and prepared for EBSD as described in Section 2.2.

The EBSD investigation of the deformed specimens indicated that the deformation mechanisms differed between A0 and the Al$_2$O$_3$-added materials. However, all Al$_2$O$_3$-added materials had very similar microstructures. As presented in [16], the microstructure of the deformed A0 specimens showed clear signs of DRX, induced by a shift in grain size distribution towards smaller grain sizes after deformation, as can be seen in Fig. 2(a). No such decrease was observed in any of the Al$_2$O$_3$-added materials, see Fig. 2(b)-(f). Hence, the softening of polycrystalline Mo(Si,Al)$_2$ cannot be explained by DRX solely.

In [16] it was also shown that low angle grain boundaries (LAGBs) formed during compression testing of Mo(Si,Al)$_2$, which probably contributed to the observed softening through recovery. In Fig. 5(a) a grain reference orientation deviation (GROD) map of this material is shown, and a number of LAGBs are indicated by arrows. In the Al$_2$O$_3$-added materials tested in this present study, very few LAGBs were found. This is exemplified by the maps from the A15 and A25 samples in Fig. 5(b) and (c), respectively. All Al$_2$O$_3$-added materials showed very similar structures as A15 and A25, with very few LAGBs. From Fig. 5(a), (b) and (c), which share the same GROD colour scale, it can be seen that the overall intragranular (geometrically necessary) dislocation density, based on GROD values, is higher in A0 than in the Al$_2$O$_3$-added materials. As the samples were deformed to similar strains, and the intragranular dislocation density in the Al$_2$O$_3$-added materials was low, it is suggested that dislocation slip is not the dominating deformation mechanism in fine-grained Al$_2$O$_3$-added materials. However, the deformation was inhomogeneous, with some grains (typically grains with high Schmid factor for the [1120][0001] slip system), were more deformed than other, often with GROD values increasing in specific crystallographic directions of the grain (perpendicular to [0001] direction), as previously observed in the coarse-grained Al$_2$O$_3$ material [16]. This suggests that even though dislocation slip is not the main deformation mechanism, it does occur in favourably oriented grains.

4. Discussion

Al$_2$O$_3$ was added to polycrystalline Mo(Si,Al)$_2$ with the intention to improve the high temperature mechanical properties. As expected, the Al$_2$O$_3$ particles had a grain boundary pinning effect during sintering, which rendered materials with finer microstructure than a Al$_2$O$_3$-free material. However, all fine-grained materials showed lower maximum stresses compared to the coarse-grained reference material when tested in compression at 1300 °C. This indicates that the deformation of Mo(Si,Al)$_2$-Al$_2$O$_3$ composites is very sensitive to grain size and that the deformation fine-grained materials is primarily controlled by creep mechanisms, such as grain boundary sliding or diffusion creep, rather than dislocation glide. The transition from dislocation glide to grain boundary sliding results in a decreasing strength with decreasing grain size, i.e. an inverse Hall–Petch relationship, for small grain sizes. This is supported by the results from the microstructural investigation, which showed that the fine-grained materials showed lower levels of internal misorientations (dislocation density) and had not, in contrast to coarse-grained Al$_2$O$_3$-free material, undergone (dislocation density-driven) DRX or formed LAGBs or highly deformed grains during deformation.

Even though the microstructures of the Al$_2$O$_3$-added materials were similar after deformation (apart from the different Al$_2$O$_3$ fractions), there was a large variation in maximum stress. This indicates that the Al$_2$O$_3$ fraction significantly affects the deformation response. Furthermore, the “U”-shaped trend of the maximum stress versus Al$_2$O$_3$ content (Fig. 4) suggests that competing mechanisms are active during compression. On the one hand, there is a softening mechanism which dominates at low Al$_2$O$_3$ levels (5–15 vol.%). On the other hand, at higher levels (20–25 vol.%), there is a gradual recovery of the strength. A similar trend, where two different mechanisms affect the strength of MoSi$_2$-based material, has been reported by Sadananda and Feng [18]. In Al-free MoSi$_2$, an amorphous SiO$_2$ film is present at the grain boundaries which makes the material very prone to creep and grain boundary sliding. It was shown that when adding C to the material, SiC was formed in an in situ reaction between C and the SiO$_2$ film. The formation of SiC particles had a grain size refining affect, which was detrimental for the creep resistance at elevated temperature (1200 °C). However, this was balanced by a decrease in creep tendency due to consumption of SiO$_2$ film. When compensated for the grain size, Sadananda and Feng did show that the particle strengthening had a positive effect on creep properties when the final volume fraction of SiC was higher than 20 vol.% [18]. This resembles the behaviour in our present study, even though the mechanisms are probably not the same as no amorphous film is present at the grain boundaries in Mo(Si,Al)$_2$.

As discussed above, the lower stresses of the Al$_2$O$_3$ containing materials suggest that there is a strong effect of the grain size, and it is therefore probable that the strength recovery at higher fractions is related to the increased grain boundary strengthening effect from the added particles. However, the decreasing strength with increasing particle content at lower volume fractions (5–15 wt%) is harder to rationalize, as the grain size is approximately constant.
Calculations showing an influence of charging on the optical phonon promote grain boundary sliding. This was supported by first principle the main contributor to electroplasticity. It was argued that such imbalance over grain boundaries is to rapid heat transfer from grain boundaries to grain interior. Instead, et al. [28] have shown that such localized heating is hard to explain due to the decrease in flow stress [31], and localized melting of grain boundaries has also been observed [32]. On the other hand, using simulations, Kim [30] has shown that the decreasing strength with increasing particle content in fine-grained Al2O3 contents (20 wt% or more) the particles act as obstacles which prevent grain boundary sliding as discussed above. This is in line with previous research on MoSi2-SiC composites [18]. Therefore, we suggest that two competing mechanisms are active during deformation: (1) electropoly- lation, which leads to a decrease in maximum stress, and (2) Al2O3 particle strengthening, which prevents grain boundary sliding and increases the maximum stress. At lower volume fractions the particle strengthening is not sufficient and the strength therefore decrease with increasing fraction. At a critical Al2O3 volume fraction, which in this study was around 15 wt% Al2O3, the particle strengthening become efficient enough to outweigh the electropoly- lation-induced softening, and the strength increases with particle content. This is schematically shown in Fig. 7. The figure shows that the change in maximum stress decreases in flow stress of polycrystalline Al2O3 in the presence of an electric field. This was attributed to a change in diffusion mechanism, from relatively slow lattice diffusion to faster diffusion at grain boundaries.

In the case of Al2O3-added Mo(Si,Al)2, it is expected that the presence of particles with high electrical resistivity at the grain boundaries will increase the local current density by reducing the effective cross-section through which current is flowing. We qualitatively demonstrate this through simplified coupled thermo-electrical simulations using COMSOL Multiphysics 6.0 software. Three cases were investigated. Case 1 contained a single grain boundary, whereas cases 2 and 3 contained an increasing number of particles with high resistivity (simulating Al2O3 particles) at the boundary. The grain boundary was modelled as a 2 nm wide region with a resistivity \( \rho_{gb} = 100 \rho_0 \) where \( \rho_0 \) is the bulk resistivity, while the resistivity of the particles was set to \( \rho_p = 1000 \rho_0 \). Fig. 6 shows the resulting current density across the grain boundary for the different cases. We note that the presence of particles leads to a significant increase in the current density within some 250 nm from the boundary, which increases drastically as the spacing between particles is decreased (Fig. 6(b)). The increase is directly proportional to the reduction in area, as expected. We also note a very local increase in the space charge density in the vicinity of the boundary, which increases with decreasing particle spacing, that is, increasing the total particle volume. While this could support the charge imbalance mechanism proposed by Kim et al. [28], further detailed simulations are needed to draw any firm conclusions. Note that only time-independent (equilibrium) simulations were performed, so local Joule heating could not be observed.

Hence, independent of the actual mechanism(s) operating (local Joule heating, bond weakening or enhanced diffusion) an increased particle density is expected to lead to more pronounced electropoly- lation through an increase in the local current density. This would explain the decreasing strength with increasing particle content in fine-grained Mo(Si,Al)2 containing 5 to 15 wt% Al2O3. At higher Al2O3 contents (20 wt% or more) the particles act as obstacles which prevent grain boundary sliding as discussed above. This is in line with previous research on MoSi2-SiC composites [18]. Therefore, we suggest that two competing mechanisms are active during deformation: (1) electropoly- lation, which leads to a decrease in maximum stress, and (2) Al2O3 particle strengthening, which prevents grain boundary sliding and increases the maximum stress. At lower volume fractions the particle strengthening is not sufficient and the strength therefore decrease with increasing fraction. At a critical Al2O3 volume fraction, which in this study was around 15 wt% Al2O3, the particle strengthening become efficient enough to outweigh the electropoly- lation-induced softening, and the strength increases with particle content. This is schematically shown in Fig. 7. The figure shows that the change in maximum stress

It is possible that the “U”-shaped trend observed in Fig. 4 is related to the use of resistive heating. It is known that metals and ceramic materials often deform at lower stress levels when subjected to an electro current. This has been shown to be true even without a significant increase in temperature due to Joule heating, or when the increase in temperature has been corrected for in the analysis [28,29]. The phenomenon is referred to as electroplasticity and was first reported by Troitskii and Likhtman in 1963 [30], but is not yet fully understood. It has been argued that localized heating of grain boundaries contributes to the decrease in flow stress [31], and localized melting of grain boundaries has also been observed [32]. On the other hand, using simulations, Kim et al. [28] have shown that such localized heating is hard to explain due to rapid heat transfer from grain boundaries to grain interior. Instead, Kim et al. suggested that charge imbalance over grain boundaries is the main contributor to electropoly- lation. It was argued that such imbalance should lead to weakening of atomic bonds, which could in turn promote grain boundary sliding. This was supported by first principle calculations showing an influence of charging on the optical phonon frequencies at grain boundaries [28]. Measurements of temperature compensated elastic modulus showed that the effect increases with decreasing grain size and increasing current density. Kim et al. further suggested that also the atomic diffusion rate might increase due to the charge imbalance. Campbell et al. [33] measured decreasing flow stress of Al2O3, and was first reported by Troitskii [29].
5. Summary

- The maximum stress of polycrystalline Mo(Si,Al)12 is sensitive to grain size. Fine-grained materials deform plastically at lower stresses than a coarse-grained material, suggesting that creep-like mechanisms dominate under the current test conditions (1300 °C and a nominal strain rate of 10^-4 s^-1).

- Contrary to the coarse-grained Al2O3-free reference, no grain refinement through DRX occurred in the fine-grained materials, and very few LAGBs could be observed. Furthermore, the internal misorientation gradients were small in the deformed state. This supports the suggestion that diffusion dependent creep mechanisms dominate, rather than intragranular dislocation motion.

- We observe a complex Al2O3 particle fraction dependence of the strength, where an increased fraction initially leads to a decrease in the maximum stress. A minimum is reached around 15 wt%.

after which the strength gradually recovers with increasing fractions. We suggest that this is an effect of the resistive heating mode, leading to the occurrence of electroplasticity, which reduces the strength with increasing fraction. This is counteracted by grain boundary strengthening, which increases with increasing fraction. Electroplasticity dominates at lower fractions, whereas at higher fractions the particle strengthening takes over. This also suggests that in the absence of current-induced electroplasticity Al2O3 additions would lead to a continuous strength increase with volume fraction at constant grain size.

CRediT authorship contribution statement

Aina Edgren: Conceptualization, Methodology, Investigation, Writing – original draft, Visualization. Erik Ström: Supervision, Conceptualization, Resources, Writing – review & editing. Lars Frisk: Investigation, Resources. Farid Akhtar: Investigation, Resources, Writing – review & editing. Magnus Hörnqvist Collander: Supervision, Conceptualization, Writing – review & editing, Funding acquisition, Project administration.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Aina Edgren and Erik Ström are employees of Kanthal AB, a manufacturer of commercial heating elements.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.msea.2023.144583.

References


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