Impression creep behaviour of ultrasonically processed in-situ Al₃Ti reinforced aluminium composite

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ABSTRACT

The creep analysis of Al₃Ti reinforced Aluminium composites with different weight percent of Al₃Ti particles was carried out under stresses between 113 and 170 MPa and temperatures ranging from 543 to 603 K. The microstructure showed uniformly distributed micron scale Al₃Ti particles throughout the matrix due to ultrasonic stirring. The presence of Al₃Ti particles refined the microstructure of the composites as it promoted heterogeneous nucleation. The results obtained from creep analysis revealed that the composites had higher activation energy and stress exponent compared to the base alloy. The improved creep behaviour is attributed to the presence of homogeneously distributed Al₃Ti particles in the aluminium matrix. The obtained stress exponent and activation energy values suggest that the main creep mechanism in the base Al alloy and Al₃Ti reinforced composites was lattice diffusion-controlled dislocation climb.

1. Introduction

Particulate reinforced aluminium matrix composites are used in the aerospace and automobile industry due to their excellent properties, such as high temperature creep ability, low density, high strength, good thermal stability, etc. [1,2]. At room temperature, aluminium alloys show high strength but their mechanical properties drop to less than 50% at temperatures higher than 473 K, limiting their use at high temperature [3].

Conventional creep test is time consuming and inconvenient because it requires many samples and it is difficult to interpret the mechanisms due to microstructural modification during the test. Therefore, a localised creep in a limited duration as achieved in indentation testing is a better alternative. In indentation creep test, spherical, pyramidal or a conical indenter is used which do not show a steady state at constant load. However, at constant load, cylindrical indenter with a flat end not only reduce the difficulty to find out the strain rate for a spherical or pyramidal indenter but also provide constant strain rate under constant stress lending to steady state penetrating velocity. Such a creep test is called impression creep [4]. The term “impression creep” was first introduced by Chu and Li [5].

Impression creep has several advantages over conventional creep tests like (i) it demands very small amount of material for the tests, (ii) test duration is relatively short, (iii) one sample can give large amount of creep data which may result in reduction in effort for sample preparation and sample to sample variations in properties (iv) this test is nearly non-destructive because a very small indent is left on the specimen. This test has some limitations, such as (i) in this test compressive load is applied unlike conventional creep test where tensile load is applied, (ii) sample cannot be fractured, therefore, determination of rupture life of the sample is difficult by this technique, (iii) creep behaviour due to changes in microstructure in engineering alloys is difficult to evaluate because test duration is short, only about a few hundreds of hours [6].

Creep behaviour of various aluminium alloys reinforced with different types of reinforcements such as TiC [3], B₄C [7], SiC [8-11], Al₂O₃ [12] have been studied. Among these, silicon carbide (SiC) as the short fibres, particles or whiskers are mostly used as reinforcement in the Al alloys matrix. Tetragonally structured Al₃Ti can be a potential reinforcement for Al matrix composite for high temperature applications because it has high elastic modulus (217 GPa), low density (3.4 g cm⁻³), excellent corrosion resistance, excellent oxidation resistance and high melting point (1350 °C) [13].

In-situ process, due to good particle wetting, stable interface, low fabrication cost and improved mechanical properties, has been considered by many researchers [14-16]. Ultrasonic vibration are extensively used to purify, degas and refine the metallic melt [17,18]. When high intensity ultrasonic fields are injected into the melt, it
produces acoustic streaming and cavitation nonlinear effects in the molten metal which not only distribute particles uniformly throughout the matrix but also improves wettabillity between matrix and reinforced particles [19]. Many research works on the impression creep properties of the magnesium [20,21] and aluminium alloys matrix composites [22,23] are being carried out, but impression creep study of aluminide reinforced Al composites has not been reported so far. Therefore, the present study aims to investigate the creep properties of Al3Ti reinforced Al composite.

2. Experimental procedure

To fabricate composite, Al6061 alloy (Hindalco, India) and K2TiF6 salt powder (Madras Fluorine Private Ltd, Chennai, India) were used as the matrix and reinforcement, respectively. The processing of the composites are reported in an earlier publication [24]. The nomenclature of the developed composites is given in Table 1.

Table 1

<table>
<thead>
<tr>
<th>Casting Name</th>
<th>Amount of K2TiF6 addition (wt%)</th>
<th>Amount of reinforced Al3Ti (wt%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Base Al alloy</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>C1U</td>
<td>5</td>
<td>2.7</td>
</tr>
<tr>
<td>C2U</td>
<td>10</td>
<td>5.4</td>
</tr>
<tr>
<td>C3U</td>
<td>15</td>
<td>8.1</td>
</tr>
</tbody>
</table>

For impression creep experiments, samples of size 15 mm × 15 mm × 6 mm were cut from each casting by using a diamond cutter. At specific temperature and load, a tungsten carbide indenter of 1.5 mm in dia. was made to indent into the surface of sample and the creep rate was obtained from the curve between the index of indentation and time. These experiments were carried out on an indentation creep machine (Sprantronics, India) which consist of a linear variable displacement transducer (LVDT), cylindrical split type furnace with PID controller, a lever arm and a data acquisition system. The schematic diagram of impression creep is illustrated in Fig. 1. Creep is of engineering significance at a T > 0.5 Tm [25], where Tm is absolute melting temperature in Kelvin. For Al alloy, Tm is 933 K and therefore 0.5 Tm is ~467 K. So, the test temperature for creep analysis should be higher than 467 K. In the present study, the impression creep tests were carried out on base Al alloy, C1U, C2U and C3U samples, at three different temperatures (543 K, 573 K and 603 K) and three different stresses (113 MPa, 141 MPa and 170 MPa) for 10,000 s under a vacuum of 10^{-7} Pa. Optical microscope (Leica, DMI 5000 M) and scanning electron microscope (Carl Zeiss, EVO 18) in secondary electron imaging mode were used for the microstructural investigation. Phases present in the composites were confirmed by XRD analysis using Rigaku smart lab, X-ray diffractometer employing Cu Kα radiation. TEM (JEOL-JEM-3200FS) was used to critically analyse the interfacial characteristics. Thin foils for TEM were prepared by the ion-milling technique. For microstructural analysis, the samples were polished with SiC abrasive papers down to 2000 grit and cloth by using MgO powder as abrasive. After cloth polishing, samples were cleaned using ultrasonic cleaner to get rid of any abrasive particles on the surface and then surface of the samples were etched with Keller’s reagent for 20 s.

3. Results and discussions

3.1. Microstructure

Fig. 2 shows the optical micrograph of the base Al alloy, C1U, C2U and C3U. It is observed from the micrograph that Al3Ti particles, which appear as dark spots confirmed by EDX analysis (Fig. 3), are dispersed uniformly throughout the microstructure and increased in numbers from C1U to C3U when more amounts of K2TiF6 is added into the melt. The uniform distribution of in-situ formed Al3Ti particles is achieved by cavitation and streaming phenomena that occur in the melt when ultrasonic vibration is introduced into the melt. It is observed from micrograph that Al3Ti particles are present inside the grains and composites have more refined microstructure over base Al alloy, which is a result of combined effect of ultrasonication and presence of Al3Ti particles in the melt which act as nucleation sites during solidification [24]. The average size of Al3Ti particles in the composite was estimated to be 3.4 ± 1.2 µm. Fig. 4 shows the XRD patterns of base Al alloy and composites which confirm the XRD peaks corresponding to the in-situ formed Al3Ti particles in all the composites. Other intermetallics, such as Al2Ti, AlTi3 and elemental Ti reported in other studies were not detected suggesting that the reaction between dissolved Ti and Al went to completion. It is evident from the TEM analysis that the interface between the Al matrix and Al3Ti particles is clean as shown in Fig. 5 without the presence of porosity or any reaction product. The in-situ formation of Al3Ti particles within melt reduces the possibility of oxidation of the particles, thus improving the interfacial bonding between matrix and particles.

3.2. Post-creep test microstructure

To study the microstructural changes during creep, the tested samples were sectioned along the impression direction and the microstructure beneath the indentation was examined. Fig. 6(a) shows microstructure of a sectioned sample of C3U composite which was creep tested at 573 K and 141 MPa. In the figure, three different zones are observed which is differentiated by curved lines and labelled with different numbers. The line pattern is supposed to be associated with material flow during impression creep. These different deformation features can be attributed to the localised strain and stress field during impression creep [26,27]. The first zone, which is just beneath the indenter, has no microstructural changes due to hydrostatic stress distribution which is also reported by many researchers [28–30]. The second zone between the arc lines, where severe deformation occurred composed a semi-spherical zone. The Al3Ti particles are aligned along the flow direction which is indicated by arrows in Fig. 6(b). In this process, it is believed that under the applied stress an elastic-plastic zone is pushed into the material. Therefore, the velocity of this zone which proceeds into the material underneath the indenter governs the impression velocity. The diameter of the flow circle is comparable to the diameter of the indenter. The flow lines are more concentrated near the edges of the indenter than in front of the indenter which shows that the most severe plastic deformation takes place at the edges of the indenter. And, in the third zone, which is far away from the indenter is characterized by Al3Ti particles being randomly distributed indicative of a lack of flow pattern. Also, there is an absence of plastic deformation of the material.
3.3. Impression creep

The impression creep curves obtained after 10,000 s tests are shown in Figs. 7, 8, 9, 10 for the base Al alloy, C1U, C2U and C3U composites, respectively. It is observed from the curve that primary (Stage 1) creep followed by secondary (Stage 2) creep are observed. The tertiary stage is not observed because rigid (elastic) materials offer restriction by enveloping plastic zone which result in stable deformation under the punch. Therefore, steady state stage is maintained for long period of time and this is a unique advantage of the impression creep test over the conventional tensile creep test.

In the primary stage, creep rate decreases with time as work hardening is dominant in this stage, whereas, in the secondary stage, creep rate is constant because equilibrium is established between the work hardening and recovery. Similar impression creep curves are reported in the literature for other aluminium alloys [31,32]. It is observed from impression creep curves that on increasing temperature and stress, creep rate is also increased. The primary stage in creep curves of base Al alloy, C1U, C2U and C3U at temperature 603 K is small as compared to other temperatures as work hardening effect is low at high temperature. The minimum creep rate, \( \dot{\varepsilon} \), during the steady state creep is determined by

\[
\dot{\varepsilon} = \frac{\partial h}{\partial t}
\]

where \( h \) (mm) is the depth of indentation and \( t \) (s) is time.

In crystalline materials, steady state creep rate is given by

\[
\dot{\varepsilon} = \frac{A\sigma^n}{d^q}e^{-Q/RT}
\]

where \( \sigma \) is the applied stress, \( n \) is the stress exponent, \( d \) is the grain size, \( q \) is the grain size exponent, \( Q \) is the activation energy, \( T \) is temperature in Kelvin, \( R \) is the gas constant and \( A \) is a constant.

Power law relation is used for dominant creep mechanism which is as follows:
\[ \dot{\varepsilon} = \frac{A D_v G b}{kT} \left( \frac{\sigma}{G} \right)^n \quad (3) \]

where \( D_v \) is the diffusion coefficient, \( \dot{\varepsilon} \) is the minimum creep rate, \( b \) is the Burger vector, \( T \) is the temperature in Kelvin, \( A \) is the constant, \( k \) is the Boltzmann constant, \( G \) is the shear modulus and ‘\( n \)’ is the stress exponent. The equation of \( D_v \) is given by

\[ D_v = D_0 \exp \left( -\frac{Q}{RT} \right) \quad (4) \]

where ‘\( Q \)’ is the activation energy, ‘\( R \)’ is the gas constant and ‘\( T \)’ is the temperature in Kelvin. By combining Eqs. (3) and (4) we get

\[ \dot{\varepsilon} = A \left( \frac{G b D}{kT} \right) (\sigma/G)^n \exp \left( -\frac{Q}{RT} \right) \quad (5) \]

For stress exponent ‘\( n \)’, Norton’s equation is used

\[ \dot{\varepsilon} = A \sigma^n \exp \left( -\frac{Q}{RT} \right) \quad (6) \]

Which can be rewritten by taking logarithm on either side as

\[ \ln \dot{\varepsilon} = \ln A + n \ln \sigma - \frac{Q}{RT} \quad (7) \]

According to this model, at constant temperature, a plot between \( \ln \dot{\varepsilon} \) and \( \ln \sigma \) gives straight line having a slope equal to the stress exponent ‘\( n \)’. The values of ‘\( n \)’ at different temperatures for base Al alloy, C1U, C2U and C3U composites are different as shown in Fig. 11 and summarised in Table 2. These stress exponent values are helpful to identify the dominant creep mechanism in the material. Several operative creep mechanisms have been reported in the literature [33,34]. If the stress exponent ‘\( n \)’ is unity, the dominant creep mechanism is based on diffusional flow such as Nabarro-Herring creep and Coble creep. If ‘\( n \)’ is found to be 2, the creep controlling mechanism is grain boundary sliding. When ‘\( n \)’ is 3, it is a form of creep where the dislocations glide velocity is controlled by solute drag. When ‘\( n \)’ value is in the range of 4–7, the dominant mechanism is climb of edge dislocation. The value of ‘\( n \)’ > 7 can be observed by blocking the dislocation motion, lattice self-diffusion and grain boundary sliding which can be achieved by developing an intermediate phase within the matrix [35].

The average value of ‘\( n \)’ for base Al alloy, C1U, C2U and C3U are 3.6 ± 0.2, 4.2 ± 0.2, 5.2 ± 0.2, and 5.8 ± 0.3 respectively which indicates that the dominant creep mechanism in all the composites is the dislocation climb. As demonstrated, the stress exponent significantly decreased from 3.9 to 3.2, 4.6 to 4.2, 5.5 to 4.8 and 6.4 to 5.4 for base Al alloy, C1U, C2U and C3U composite, respectively with increasing temperature from 543 to 603 K. The drop of ‘\( n \)’ values with temperature reflects the instability of the microstructure at high deformation temperatures. The observed instability may be attributed to the softening of Al alloy matrix at high temperature. However, at 603 K, ‘\( n \)’ value of C1U, C2U and C3U composite is increased with the amount of Al3Ti which suggests that the presence of Al3Ti particles in the matrix resist the softening of the matrix at high temperature. This results in high stress exponent values as compared to that of the base Al alloy.

This mechanism is independent of grain size (\( q = 0 \)) [36,37] which is concluded from the \( \ln \dot{\varepsilon} \) vs. \( \ln d \) plot as shown in Fig. 12. It is observed from the plot that the creep rate decreases as grain size of the composites decrease which means that the grain size of the composites does not adversely affect the creep rate. Also, the increase in creep resistance due to Al3Ti particles reinforcement cancel out the adverse effect which arise due to grain refinement, if any.

In the deformation process, thermal activation is mainly responsible for creep and without thermal fluctuation creep cannot occur in the material. The activation energies for creep provide better understanding of the mechanism contributing to creep in the material. These
Fig. 7. Impression creep test of base Al alloy at 543 K, 573 K and 603 K temperature and stresses at 113 MPa, 141 MPa and 170 MPa.

Fig. 8. Impression creep test of C1U at 543 K, 573 K and 603 K temperature and stresses at 113 MPa, 141 MPa and 170 MPa.
Fig. 9. Impression creep test of C2U at 543 K, 573 K and 603 K temperature and stresses at 113 MPa, 141 MPa and 170 MPa.

Fig. 10. Impression creep test of C3U at 543 K, 573 K and 603 K temperature and stresses at 113 MPa, 141 MPa and 170 MPa.
mechanisms can be identified by their respective activation energy [38]. So, this activation energy is calculated using Arrhenius type rate equation:

\[ \dot{\varepsilon} = -A \varepsilon \dot{\sigma} e^{\frac{-Q}{R} \frac{1}{T}} \]  

(8)

For a constant stress, the plot between ln \( \dot{\varepsilon} \) vs. 1/T is a straight line which has a slope equal to \( \frac{Q}{R} \) where \( R \) is the gas constant (8.3 J/mol) and \( Q \) is the activation energy. The obtained values of \( Q \) are 111 ± 4 kJ/mol, 127 ± 3 kJ/mol, 143 ± 4 kJ/mol and 157 ± 5 kJ/mol for base Al alloy, C1U, C2U and C3U respectively as shown in Fig. 13. The standard deviation of the mean is reported as the error values. The activation energy of base Al alloy, C1U, C2U and C3U are close to lattice self-diffusion (142 kJ/mol) [39]. It indicates that the velocity of dislocation climb is controlled by diffusion of Al atoms through lattice. It is observed from Fig. 13 that the values of activation energy are decreased as stress is increased. This can be attributed to the formation of higher density of dislocation under an application of higher stresses. As a result, more rate of materials flow by diffusion through dislocation pipe than that through the lattice which lowers the activation energy [40]. Therefore, on the basis of stress exponent and activation energy values, it can be concluded that lattice diffusion-controlled dislocation climb is the dominant creep mechanism in the base Al alloy and the composites.

Fig. 14 shows minimum creep rate or steady state creep rate of base Al alloy and composites (C1U, C2U and C3U) at different temperature and different stresses. From the figure, it is observed that steady state creep rate or minimum creep rate of base Al alloy is higher than that of composites at all the test temperatures. This is because dislocation creep or power law creep is governed by the movement of dislocations assisted by dislocation climb aided by diffusion of vacancy [25]. This movement of dislocation is dependent on activation energy. The value of activation energies of the composites is larger than that of base Al alloy as shown in Fig. 13. For large activation energy, diffusion of vacancy is low which resist the movement of dislocation and thus, the

Table 2
Stress exponent \( 'n' \) values for base Al alloys and different composites.

<table>
<thead>
<tr>
<th>Composite</th>
<th>Creep test temperature, K</th>
<th>Stress exponent, n</th>
</tr>
</thead>
<tbody>
<tr>
<td>Base Al alloy</td>
<td>543</td>
<td>3.9</td>
</tr>
<tr>
<td></td>
<td>573</td>
<td>3.7</td>
</tr>
<tr>
<td></td>
<td>603</td>
<td>3.2</td>
</tr>
<tr>
<td>C1U</td>
<td>543</td>
<td>4.6</td>
</tr>
<tr>
<td></td>
<td>573</td>
<td>3.9</td>
</tr>
<tr>
<td></td>
<td>603</td>
<td>4.2</td>
</tr>
<tr>
<td>C2U</td>
<td>543</td>
<td>5.5</td>
</tr>
<tr>
<td></td>
<td>573</td>
<td>5.2</td>
</tr>
<tr>
<td></td>
<td>603</td>
<td>4.8</td>
</tr>
<tr>
<td>C3U</td>
<td>543</td>
<td>6.4</td>
</tr>
<tr>
<td></td>
<td>573</td>
<td>5.7</td>
</tr>
<tr>
<td></td>
<td>603</td>
<td>5.4</td>
</tr>
</tbody>
</table>

Fig. 11. Plot of ln \( \dot{\varepsilon} \) vs. ln \( \sigma \) for calculating the stress exponent for base Al alloy, C1U, C2U and C3U.

Fig. 12. Plot of ln \( \dot{\varepsilon} \) vs. ln \( d \) for calculating grain size exponent \( 'q' \) for all the three samples at 603 K temperature.
Fig. 13. Plot of \( \ln \dot{\varepsilon} \) vs. \( 1/T \) for calculating the activation energy for base Al alloy, C1U, C2U and C3U.

Fig. 14. Comparison of creep rate for base Al alloy, C1U, C2U and C3U under different stresses tested at 543 K, 573 K and 603 K.
creep rate is low. The C3U composite has the lowest creep rate among all the composites because it has the largest activation energy. It is also observed from the figure that the steady state creep rate is higher at high temperature than that at lower temperature for a given stress. At higher temperatures, the ability of dislocations to climb over particles is apparently fast enough to render the particles as ineffective dislocation barriers. At lower temperatures, the time needed for a dislocation to climb around a particle (for a given stress) is much longer, so the creep resistance is significantly improved [41].

Creep resistance of the composite also depends on coherency between matrix and reinforcement particles. A large mismatch in lattice coherency between matrix and reinforced particles give undesirable microstructure that have poor mechanical properties at elevated temperatures. Fig. 15(a) is a diagram illustrating a non-coherent particle having no crystal structural relationship with the aluminium atoms. When there is a lattice coherency, these dispersed particles are highly stable and provide good mechanical properties for the composite at elevated temperatures [42]. Fig. 15(b) is a diagram illustrating a coherent particle that has similar lattice parameters and crystal structure relationship with the surrounding aluminium matrix atoms. The crystal structure of \( \text{Al}_3\text{Ti} \) is a tetragonal with \( a = b = 0.385 \text{ nm} \) and \( c = 0.861 \text{ nm} \) [43]. On the other hand, \( \alpha\)-Al crystal has a fcc structure with \( a = b = c = 0.404 \text{ nm} \) [44]. So, the lattice misfit values between in-situ formed \( \text{Al}_3\text{Ti} \) and \( \alpha\)-Al in both \( a \) and \( c \) directions are 0.049 and 0.065, respectively. Based on this calculation, it can be concluded that in-situ formed \( \text{Al}_3\text{Ti} \) particle has fairly good lattice match with \( \alpha\)-Al and therefore, enhances creep property of the composite.

Intermetallic particles enhance creep properties by either resisting grain boundary sliding or suppressing dislocation annihilation (recovery) [40,45,46]. The high melting point of \( \text{Al}_3\text{Ti} \) particle confirms its stability at the creep test temperature. Therefore, it seems that the presence of the \( \text{Al}_3\text{Ti} \) particles inside the matrix phase enhances the creep properties of the composites by preventing dislocation easy gliding during the creep deformation. Further, the presence of \( \text{Al}_3\text{Ti} \) in the matrix improve its resistance to plastic deformation which also increases creep resistance of the composite. Therefore, it can be observed from Figs. 7, 8, 9, 10, that at a particular stress and temperature, the penetration depth of the indenter for any duration of time is decreased as the amount of reinforcement is increased.

4. Conclusions

The creep behaviour of \( \text{Al}_3\text{Ti} \) reinforced Al alloy matrix composites, fabricated by ultrasonic assisted casting method, is investigated at various temperatures ranging from 543 K to 603 K and under stresses between 113 MPa and 170 MPa. Creep resistance of C1U, C2U and C3U composites are higher than that of the base Al alloy suggesting that \( \text{Al}_3\text{Ti} \) particle addition is beneficial. C3U composite with the highest particle addition is the most creep resistant. The creep resistance is attributed to the lattice coherency between \( \text{Al}_3\text{Ti} \) particles and Al matrix reflected in higher activation energy values which resist the movement of dislocation. The values of calculated stress exponent \( n \) range from 3.2 to 3.9, 3.9 to 4.6, 4.8 to 5.5 and 5.4 to 6.4 for base Al alloy, C1U, C2U and C3U, respectively, suggesting that the dominant creep mechanism is the dislocation creep. The calculated activation energies for base Al alloy, C1U, C2U and C3U are approximately 111 ± 4 kJ/mol, 127 ± 3 kJ/mol, 143 ± 4 kJ/mol and 157 ± 5 kJ/mol respectively. The activation energy value for the creep of base Al alloy, C1U, C2U and C3U are close to that of the lattice self-diffusion (142 kJ/mol) of aluminium. This implies that the dominant creep mechanism in base Al alloy and composites (C1U, C2U and C3U) is the lattice self-diffusion-controlled dislocation creep. Since creep controling mechanism is dislocation creep mechanism which is independent of grain size, the refinement in microstructure of the composites, obtained from the incorporation of uniformly distributed \( \text{Al}_3\text{Ti} \) particles throughout the matrix, achieved by ultrasonication of the melt, do not affect the creep behaviour of the composites.

Data availability

The raw data required to reproduce these findings are available to download from http://dx.doi.org/10.17632/ddg8dxwst.1. The processed data required to reproduce these findings are available to download from http://dx.doi.org/10.17632/y2t3rzmm.1.

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