Supergravity-Induced Separation of Oxide and Nitride Inclusions from Inconel 718 Superalloy Melt

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Herein, a method of supergravity-enhanced separation was used to remove oxide and nitride inclusions from Inconel 718 superalloy melt, with elucidating the inclusion removal behavior by varying the gravity coefficients (G) and separation times (t) used for melt treatment. Under supergravity conditions, inclusions concentrated at the sample top and are almost absent at the sample bottom. Moreover, the inclusion number density and average size showed a gradient distribution along the supergravity direction, and the steepness of this gradient rapidly increased with increasing G and t. The experimentally determined inclusion movement velocities agreed well with those calculated using Stokes’s law at G ≤ 210 and t ≤ 10 min. At G = 210 and t = 10 min, the total oxygen and nitrogen contents of the sample decreased from 34.4 to 8.7 ppm and 133.4 to 34.1 ppm, respectively, corresponding to oxide and nitride removal efficiencies of 74.7% and 74.4%, respectively.

KEY WORDS: supergravity; oxide inclusion; nitride inclusion; separation; superalloy.

1. Introduction

Superalloy 718 is extensively used in the manufacturing of high-performance aircraft engine components because of its excellent corrosion resistance, high strength, and good oxidation resistance at elevated temperatures.1) However, as in the cases of other superalloys, the operation efficiency of superalloy 718 can be limited by the unavoidable presence of nonmetallic inclusions (mainly of the oxide and nitride type).2,3) During melt solidification, oxide inclusions typically act as nucleation sites for nitride inclusions, which are small in size but have sharp edges and corners.4) Inclusions of both types are harmful to the mechanical properties and solidification behavior of Ni-based superalloys, inducing the occurrence of cracking and decreasing fatigue lifetime.5,6) Moreover, matrix–inclusion lattice plane incoherence results in stress concentration, promotes cracking, and can ultimately lead to component failure.7)

The current industrial technologies employed to refine superalloys includes secondary vacuum induction melting, electro-slag refining, and vacuum arc re-melting. A combinations of these techniques are widely used in the processing of most superalloys and special steels.8) Although most of the inclusions in superalloys can be removed by these methods, a great number of inclusions still remain in the bulk of the alloys.9) This is mainly attributed to the mechanism of inclusion removal in the currently used methods. When the superalloy is melted, molten droplets go through molten slag or vacuum, and inclusion particles can be absorbed into slag if they happen to be located on the droplet surface. The inclusion particles that do not come to the surface of the droplet cannot be removed. Nevertheless, none of these techniques can ensure sufficient stirring and turbulence in molten droplets, which limits the removal efficiency of the inclusions.10) Compared with oxide inclusions, nitride inclusions are more difficult to be removed by the current methods, since nitride inclusions dissolve when the superalloy melt and precipitate again as the temperature decreases in the solidification process of the superalloy. It is found that the dissolution of these inclusions has hardly effect on the removal process.11–13) Therefore, the development of a novel, green, low-cost, and highly efficient method of superalloy purification is a task of high practical significance.14)

Recently, density differences between impurities and the melt have been used as the basis for supergravity-based large-scale purification of metals.15) For example, Matsubara et al.16) studied the removal of Fe from a liquid Al alloy under a supergravity field during Al solidification and attributed the observed separation to supergravity-induced macro-segregation during solidification. Song et al.17) studied the removal of inclusions from Al melt by supergravity filtration, demonstrating that 97.3% of oxide inclusions could be eliminated. Li et al.18) successfully extended the supergravity technology to the separation of fine Al2O3 inclusions from liquid steel, reducing its total oxygen content from 191.7 to 8.4 ppm.

In view of these successful applications of supergravity, this technique is introduced to separate non-metallic inclusions from superalloys. Since the behavior of nitride...
inclusion at high temperature is quite different with oxide inclusion, the feasibility of supergravity technology on the removal of oxide and nitride inclusions was examined respectively in this study. The effects of gravity coefficient \((G)\) and separation time \((t)\) on the removal rate, distribution, microstructure and size of the oxide and nitride inclusions in Inconel 718 superalloy were investigated.

2. Experimental

2.1. Raw Material

Inconel 718 superalloy rods (composition provided in Table 1) were produced in a vacuum induction furnace and then machined into 26-mm-diameter cylinders for supergravity separation experiments.

2.2. Supergravity-Promoted Separation

The supergravity field was generated by a centrifugal apparatus whose schematic is shown in Fig. 1. Two thermal insulators were installed symmetrically onto the centrifugal axis for sample placement and weight balancing, respectively. An alumina crucible (inner diameter = 26 mm, height = 30 mm) was filled with an Inconel 718 superalloy sample (about 170 g) and placed into a graphite crucible (inner diameter = 28 mm) with the outer wall coated by microwave-absorbing materials. The graphite crucible containing sample was then placed horizontally in one thermal insulator. The microwave-absorbing materials can be heated up to 1 873 K by absorbing energy from the microwave generator, and the sample was heated through heat transfer at the same time. On the opposite side, another graphite crucible without a microwave-absorbing coating was filled with the same amount of sample and then loaded into the second thermal insulator to keep balance of the centrifugal apparatus.

The temperature was controlled by a programmable temperature controller (PTC) with a type B thermocouple which was placed closed to the outer wall of the graphite crucible with the microwave-absorbing coating. The thermocouple cannot be affected by microwave since there is no any microwave-absorbing material on it. PTC converted the electrical signal obtained by thermocouple into temperature value and compared it with the set value. Based on the difference between these values, PTC controlled the connection and disconnection of the microwave generator to achieve the set temperature or heating/cooling rate. When the rotation was started, the transmission of electrical signals was realized by the slip ring fixed on the centrifugal axis.

To evaluate the supergravity field, the gravity coefficient \((G)\) is defined in Eq. (1) as a ratio of the centrifugal acceleration to the normal gravity acceleration.

\[
G = \frac{\sqrt{g^2 + (\omega^2 \cdot r)^2}}{g} = \frac{\sqrt{\frac{N^2 \cdot \pi^2 \cdot r^2}{900}}}{g} \tag{1}
\]

where \(N\) is the centrifuge rotation speed (rpm), \(\omega\) is the angular velocity (rad/s), \(r\) is the distance from the centrifugal axis to the sample (0.27 m in this study), and \(g\) is the normal gravitational acceleration (9.8 m/s\(^2\)).

To conduct the supergravity-promoted separation experiment, the superalloy sample was heated to 1 873 K at a rate of 26 K/min and kept at this temperature for 40 min to ensure its complete melting. Then the centrifuge was started and adjusted to the specified angular velocity of 500, 700, and 850 rpm (i.e. \(G = 70, 140,\) and \(210\), respectively) for \(t = 1, 5,\) and 10 min. It took about 30, 40 and 50 seconds for the centrifuge to arrive the specified angular velocity of 500, 700, and 850 rpm, respectively. After that, the sample was cooled at 15 K/min to 1 373 K (at which point the superalloy sample was fully solidified), and then the centrifuge was switched off and the graphite crucible with the sample was taken out. In parallel, the experiment was carried out for 1 min under the conditions of normal gravity \((G = 1)\) for comparison.

Table 1. Chemical composition of Inconel 718 (Mass%).

<table>
<thead>
<tr>
<th>C</th>
<th>Ni</th>
<th>Cr</th>
<th>Fe</th>
<th>Nb</th>
<th>Mo</th>
<th>S</th>
<th>Al</th>
<th>Ti</th>
<th>Cs</th>
<th>Ca</th>
<th>O</th>
<th>N</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.179 bal.</td>
<td>18.85</td>
<td>18.58</td>
<td>4.093</td>
<td>2.386</td>
<td>0.018</td>
<td>0.913</td>
<td>0.844</td>
<td>0.309</td>
<td>0.235</td>
<td>0.003</td>
<td>0.013</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 1. Schematic diagram of the experimental apparatus. 1: centrifugal axis; 2: microwave generator; 3: thermal insulator; 4: counterbalance sample; 5: alumina crucible; 6: graphite crucible; 7: slip ring; 8: programmable temperature controller; 9: thermocouple; 10: Inconel 718 superalloy sample. (Online version in color.)
2.3. Characterization of Oxide and Nitride Inclusions

After supergravity treatment, the sample was longitudinally divided along the center axis into two halves. One part was polished and analyzed by scanning electron microscopy coupled with energy-dispersive spectroscopy (SEM-EDS; MLA250, FEI Quanta, USA) and X-ray diffraction (XRD; Smartlab, Rigaku, Japan) to elucidate the composition and distribution of inclusions. An automatic inclusion analysis system (EVO18-INCAsteel, USA) was used to gain further knowledge on the distribution, number, size, and type of inclusions. The inclusion size obtained by the automatic inclusion analysis system is the equivalent circular diameter. The equivalent circular diameter of a non-circular object is equal to the diameter of a circular with the same area. To determine the efficiencies of oxide and nitride inclusions removal, the other half was used to measure the total oxygen and nitrogen contents at different positions along the center axis using an O, N, and H analyzer (HORIBA, EMGR-830, Japan).

3. Results

3.1. Characterization of Oxide and Nitride Inclusions in Original Inconel 718

Figure 2 shows the SEM-EDS–determined composition and distribution of inclusions in the pristine superalloy, demonstrating that inclusions comprised $\text{Al}_2\text{O}_3$, TiN, and $\text{Al}_2\text{O}_3$ surrounded by TiN and were uniformly dispersed. Figure 2(d) shows that oxide inclusions acted as nucleation sites of nitride inclusions, which, in turn, could also act as nucleation site for outermost nitride inclusion precipitation. The above results were in good agreement with those of XRD analysis (Fig. 3).

3.2. Macro- and Micro-Imaging of Samples Obtained under Supergravity

Figure 4 shows the cross-sections of samples obtained by supergravity-promoted separation at $G = 210$ ($N = 850$ rpm) and $t = 1$ min and compares them to those obtained in the case of $G = 1$, $t = 1$ min. To investigate the effect of $G$ on inclusion distribution, areas A–F in Fig. 4 were observed by SEM, with the results shown in Fig. 5. Figure 5(a) reveals that inclusions in the normal-gravity sample were uniformly distributed, while those in the sample obtained at $G = 210$ presented a distribution with a gradient along the central axis, as indicated in Figs. 5(b)–5(f). In the latter sample, a large number of oxide and nitride inclusions aggregated at the upper sample part (Fig. 5(b)), while nearly no inclusions were found in the lower part (Figs. 5(e) and 5(f)). Figure 6 shows a high-magnification SEM image of the area marked by a rectangle in Fig. 5(b) and the corresponding elemental mappings, demonstrating that the types of inclusions in the upper part of the $G = 210$ sample were identical to those in the original sample (Fig. 2).
3.3. Inclusion Number Density and Size Distribution

To further examine the distribution of inclusions, their number density (Fig. 7) and average size (Fig. 8) at different distances from the sample bottom were determined for various G and t. Figure 7 shows that inclusion number density in samples obtained under supergravity conditions exhibited a clear gradient distribution along the supergravity direction. At $G \geq 70$ and $t \geq 1$ min, the inclusion number density was rather low and close to zero (much lower than those obtained under normal gravity), but increased sharply at the sample top. Moreover, the steepness of the above gradient increased with increasing $G$ and $t$.

Inclusion average size featured a distribution similar to that of number density, as shown in Fig. 8. At $G = 1$, the size distribution was relatively uniform and was within 2 to 4 $\mu$m; while at $G = 70 – 210$, the average size of inclusions at the sample top ranged from 7 to 9 $\mu$m and was much larger than that of inclusions in middle/lower parts of samples.

3.4. Efficiency of Oxide and Nitride Inclusion Removal

Figure 9 shows the total oxygen and nitrogen contents at different positions of samples obtained under various conditions, demonstrating that the oxygen and nitrogen distribu-
tions were consistent with those of inclusion number density (Fig. 7). In samples obtained at normal gravity, the oxygen and nitrogen were evenly distributed and featured average contents of 34.4 and 133.4 ppm, respectively. Conversely, after supergravity-promoted separation, the total oxygen and nitrogen contents presented an obvious gradient distribution along the direction of supergravity, with maximal values observed at the sample top. Moreover, the gradient became steeper with increasing $G$ and $t$.

Table 2 lists the total oxygen and nitrogen contents as well as removal efficiencies obtained at different $G$ and $t$ for the position of 15 mm from the sample bottom, showing that at $G = 210$, $t = 10$ min, the total oxygen and nitrogen

Table 2. Total oxygen/nitrogen contents and O/N removal efficiencies at the position of 15 mm from the sample bottom.

<table>
<thead>
<tr>
<th>Conditions</th>
<th>Total oxygen content (ppm)</th>
<th>Total nitrogen content (ppm)</th>
<th>ηO (%)</th>
<th>ηN (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$G = 1$, $t = 1$ min</td>
<td>34.4</td>
<td>133.4</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>$G = 70$, $t = 1$ min</td>
<td>34.3</td>
<td>74.8</td>
<td>0.08</td>
<td>43.9</td>
</tr>
<tr>
<td>$G = 140$, $t = 1$ min</td>
<td>14.8</td>
<td>48.5</td>
<td>56.8</td>
<td>63.6</td>
</tr>
<tr>
<td>$G = 210$, $t = 1$ min</td>
<td>13.4</td>
<td>43.2</td>
<td>60.9</td>
<td>67.6</td>
</tr>
<tr>
<td>$G = 210$, $t = 5$ min</td>
<td>11.1</td>
<td>40.8</td>
<td>67.9</td>
<td>69.4</td>
</tr>
<tr>
<td>$G = 210$, $t = 10$ min</td>
<td>8.7</td>
<td>34.1</td>
<td>74.7</td>
<td>74.4</td>
</tr>
</tbody>
</table>

Fig. 8. Average size of inclusions at different distances from the bottom of samples obtained at (a) $t = 1$ min, $G = 1$, 70, 140, and 210; (b) $G = 210$, $t = 0$, 1, 5, and 10 min. (Online version in color.)

Fig. 9. Total oxygen and nitrogen contents observed at different distances from the sample bottom under various conditions. (a) $t = 1$ min, $G = 1$, 70, 140, and 210; (b) $G = 210$, $t = 0$, 1, 5, and 10 min; (c) $t = 1$ min, $G = 1$, 70, 140, and 210; (d) $G = 210$, $t = 0$, 1, 5, and 10 min. (Online version in color.)
contents decreased to 8.7 and 34.1 ppm, respectively, which corresponded to oxygen and nitrogen removal efficiencies (calculated using Eq. (2)) of 74.7% and 74.4%, respectively.

\[ \eta = \frac{w_n - w_i}{w_n} \times 100\% \]

where \( \eta \) is the oxygen or nitrogen removal efficiency, \( w_n \) and \( w_i \) are the total oxygen or nitrogen content in the original and supergravity-treated samples, respectively.

4. Discussion

The obtained results clearly demonstrate that supergravity has a significant effect on the removal of different types of inclusions from the superalloy. Under normal gravity, inclusions are evenly distributed in the sample because of the high melting point of Al\(_2\)O\(_3\). \( \Delta \)O\(_3\) inclusions in the 718 superalloy melt were always present in solid form even at the maximum experimental temperature of 1873 K. In contrast, TiN inclusions can be dissolved during melting at high temperatures and usually precipitate during subsequent solidification.2122

In view of the high melting point of Al\(_2\)O\(_3\) (2303 K), Al\(_2\)O\(_3\) inclusions in the 718 superalloy melt were always present in solid form even at the maximum experimental temperature of 1873 K. In contrast, TiN inclusions can be dissolved during melting at high temperatures and usually precipitate during subsequent solidification.2122 Figure 10 shows the Thermo-Calc Software–calculated equilibrium phase diagram of the 718 superalloy in the temperature range of 1873 K to 1073 K, demonstrating that the liquidus and solidus temperatures equal 1609 K and 1533 K, respectively, and that TiN inclusions begin to precipitate from the alloy melt at 1653 K. In the supergravity-promoted separation experiment, the rotation was started after the sample was heated at 1873 K for 40 min and continued until the temperature decreased to 1373 K. Once the rotation started, the Al\(_2\)O\(_3\) inclusions migrated towards the sample top driven by supergravity. When the temperature was cooled to 1653 K, TiN inclusions precipitated and began to migrate towards the sample top until the full solidification of the sample occurred at 1533 K.

![Phase diagram of the 718 superalloy containing 0.01334 mass% N and 0.844 mass% Ti. (Online version in color.)](image)

An experiment was implemented to verify the results of Thermo-Calc Software. The supergravity (\( G = 210 \)) was only applied at the isothermal stage (1873 K) for 5 min. After that, the rotation was stopped and the sample was cooled to 1373 K before being taken out from the centrifuge. The SEM images of various regions of the sample are presented in Fig. 11. It was seen that the Al\(_2\)O\(_3\) inclusions were concentrated at the sample top, while the TiN inclusions were distributed relatively evenly in the sample. Since the TiN inclusions dissolve at high temperatures and precipitate during the cooling stage, the TiN inclusions cannot be removed if the cooling stage of the sample was not applied by supergravity. This observation was in good agreement with the calculation of the Thermo-Calc Software.

Assuming that oxide and nitride inclusions are spherical, their motion in the viscous melt under the action of a centrifugal force can be described by Stokes’s law (Eq. (3)).

\[ \frac{dr}{dt} = \frac{d^2|\rho_L - \rho_P|}{18\eta} \omega^2 r \]

where \( dr/dt, \rho_L, \rho_P, \omega, r, d, \eta, \) and \( N \) are the moving velocity, liquid density, particle density, angular velocity, distance of the particle from the centrifugal axis, particle diameter, molten metal viscosity, and centrifuge rotation speed, respectively.

Under the condition \( A = \frac{d^2 \Delta \rho}{18\eta}, \Delta \rho = \rho_L - \rho_P \), Eq. (3) can be written as

\[ \frac{dr}{dt} = \frac{d^2 \Delta \rho}{18\eta} \omega^2 r = A \omega^2 r \]

If \( \omega \) is assumed to be constant, the following equation can be obtained:

\[ t = \frac{1}{A \omega^2} \ln \frac{r}{r'} = \frac{900}{A \pi N^2} \ln \frac{r}{r'}, \]

When the densities of Al\(_2\)O\(_3\) and TiN inclusions (\( \rho_{L1} = 3.97 \times 10^3 \) kg m\(^{-3}\), \( \rho_{P1} = 5.44 \times 10^3 \) kg m\(^{-3}\)), density of the 718 superalloy melt (\( \rho_L = 7.50 \times 10^3 \) kg m\(^{-3}\)), superalloy melt viscosity (\( \eta = 0.005 \) Pa s), \( N = 850 \) rpm, \( r = 0.27 \) m, and \( r' = 0.24 \) m are substituted into Eq. (5), one obtains

\[ t_1 = 3.79 \times 10^{-10} \frac{1}{d_1^2} \quad \text{and} \quad t_2 = 6.49 \times 10^{-10} \frac{1}{d_2^2} \]

At \( G = 210 \) (\( N = 850 \) rpm) and an average inclusion diameter of 3.72 \( \mu \)m, the time required for Al\(_2\)O\(_3\) and TiN

![SEM images of (a) top region and (b) middle region of the sample obtained with supergravity treatment (\( G = 210 \)) only at the isothermal stage (1873 K) for 5 min.](image)
inclusions to move from the position of 0.27 m to 0.24 m can be calculated as $t_1 = 0.46\text{ min}$ and $t_2 = 0.78\text{ min}$, respectively. Consequently, the corresponding time for a particle of $\text{Al}_2\text{O}_3$ surrounded by TiN varies between 0.46 and 0.78 min. Besides, as discussed by Miki et al., \textsuperscript{21} inclusions of different diameters can collide and grow during their movement. Therefore, most inclusions were efficiently driven to the sample top at $G = 210$, $t \leq 10\text{ min}$, with mainly small inclusions remaining at the lower part. The experimental and calculated moving velocities were concluded to be in good agreement.

It is also should be noted that, after supergravity separation, there were still a small amount of inclusions at the middle and bottom of the sample, which were almost all TiN inclusions. The dissolved Al is very active and prone to form $\text{Al}_2\text{O}_3$ inclusions with the dissolved oxygen in molten alloy, so $\text{Al}_2\text{O}_3$ inclusions can be effectively separated during the isothermal stage. There are two main reasons for the remaining of TiN inclusions. First, it is shown from Fig. 10 that in the solidification process about 25% of the TiN inclusions precipitated when the temperature was below liquidus temperature, and the increased solid fraction in the sample can decrease the moving velocity of these TiN inclusions. Second, a small amount of TiN inclusions still precipitated after the sample was solidified (below solidus temperature). These TiN inclusions cannot be removed in supergravity fields and remained in the sample. Reducing cooling rate and increasing gravity coefficient are two effective methods to remove the TiN inclusions from Inconel 718 superalloy.

5. Conclusions

Herein, we established a method of separating oxide and nitride inclusions from the Inconel 718 superalloy under the action of supergravity and arrived at the following conclusions.

(1) Supergravity is well suited for the removal of oxide and nitride inclusions from Inconel 718, inducing the migration of most inclusions to the sample top.

(2) The number density and average size of inclusions showed a clear gradient along the direction of supergravity, and the above gradient became steeper with increasing gravity coefficient and centrifugation time.

(3) The migration of inclusion particles was driven by inclusion–superalloy melt density differences, and the experimentally determined moving velocities could be well modeled by Stokes’s law.

(4) At $G = 210$, $t = 10\text{ min}$, the average oxygen and nitrogen contents of the superalloy could be reduced to only 8.7 and 34.1 ppm, respectively, which corresponded to respective removal efficiencies of 74.7% and 74.4%.

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