EFFECT OF PROCESSING PARAMETERS ON MICROSTRUCTURE AND MECHANICAL PROPERTIES OF CAST TiAl BASED ALLOYS

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Abstract

Effect of several processing parameters such as casting technology, arrangement of gating system during centrifugal casting and post-solidification heat treatments on the microstructure and mechanical properties of intermetallic Ti-45Al-8Nb-0.2C-0.2B and Ti-46Al-8Nb (at.\%) alloys was studied. Reproducibility of room-temperature mechanical properties are compared statistically by using two parameter form of Weibull relation. Room-temperature tensile properties of centrifugally and gravity cast ingots are reported and measured ductility is related to different oxygen content in the analyzed ingots. Various types of post-solidification heat treatments consisting of high temperature isostatic pressing, solution annealing, cooling at various cooling rates from solution annealing temperature and stabilization annealing were performed. The optimal heat treatment leading to the highest creep resistance is identified and related to corresponding microstructure. The parameters of kinetic equation for minimum creep rate are defined for temperature range from 973 to 1073 K and applied stresses varying from 200 to 400 MPa. Large attention is paid to primary creep stage, plastic deformation to minimum creep rate and time to 1\% creep deformation.

1. INTRODUCTION

The continuous demand for weight reduction and higher engine efficiencies in automotive, aerospace and energy industries pushes the materials towards their limits. Therefore, these industries have a strong need for developing novel light-weight materials which can withstand temperatures up to 800°C, while maintaining acceptable mechanical properties. Intermetallic TiAl based alloys are among the most promising candidates to fulfill the required specifications for structural applications [1-3]. Especially, TiAl based alloys with high Nb-contents based on the general composition Ti-(42-48)Al-(6-10)Nb (at.\%) have attracted much attention because of their high creep strength, good ductility at both room and elevated temperatures and excellent oxidation resistance [4-6]. The high Nb content reduces the stacking fault energy in TiAl, retards diffusion processes and modifies the structure of the oxidation layer [4,6]. Reduction of diffusivity facilitates massive transformation during cooling from the $\alpha$-phase field [7]. In the alloys modified by boron, boron ensures a grain refining effect during solidification and tends to form very stable borides which is also beneficial in case of heat-treatments conducted at high temperatures [8]. Here, the borides retard grain coarsening by pinning of the grain boundaries [8]. In addition, the borides favour the formation of the lamellar microstructure ($\alpha \rightarrow \alpha + \gamma$) over the massive transformation ($\alpha \rightarrow \gamma_m$) by heterogeneous nucleation of $\gamma$-lamellae [8]. Carbon improves the strength by solid solution hardening and precipitation hardening by carbides which are stable at operating temperatures [9].

The aim of this paper is to evaluate the effect of processing parameters such as casting technology and heat treatments on the microstructure and mechanical properties of cast TiAl
based alloys with the chemical composition Ti-45Al-8Nb-0.2B-0.2C and Ti-46Al-8Nb (at.%). The studied alloys have been selected as a potential material for the investment casting of components for gas turbine and automotive industry.

2. EXPERIMENTAL PROCEDURE

2.1. Centrifugal casting

Cylindrical ingots with a diameter of 16 mm and length of 110 mm from Ti-45Al-8Nb-0.2B-0.2C (at.%) alloy were centrifugally cast (CC) using a horizontal centrifugal caster “LINN Supercast” under argon atmosphere. The caster requires induction melting of the ingot material in ceramic crucibles which is followed by centrifugal casting into preheated ceramic shell moulds. Both the crucibles and the ceramic shell moulds were composed of a front layer based on Y$_2$O$_3$ and back-up layers based on Al$_2$O$_3$. The moulds were produced by several dipping/sanding cycles of adequately shaped wax patterns which was followed by de-waxing and firing the ceramic shells. During induction melting the alloy was heated to a temperature by 20°C higher than the liquidus temperature of the alloy and then cast into moulds preheated to 800°C. Two types of mould filling systems were used to optimize centrifugal casting process: (i) direct and (ii) indirect mould filling. Fig. 1 shows examples of two types of as-cast ingots prepared with direct and indirect filling systems of ceramic shell moulds. After casting the ingots were cooled to a temperature of 1200°C and transferred to a furnace with a temperature of 800°C for 1 h.

![Fig. 1. Arrangement of mould filling systems for centrifugally cast ingots: (a) direct mould filling, (b) indirect mould filling.](image)

2.2. Heat treatments of centrifugally cast ingots

In order to achieve various interlamellar spacing in the as-cast ingots from Ti-45Al-8Nb-0.2B-0.2C (at.%) alloy, the ingots were subjected to three different types of thermomechanical heat treatments:

- **Type A** - hot isostatic pressing (HIP) at 1260°C and applied pressure of 200 MPa for 4 h which was followed by cooling at rates of 10 K/min in the range from 1260 to 800°C and 5 K/min in the range from 800 to 300°C.

- **Type B** - solution annealing at 1340°C for 6 min followed by cooling at rates of 15 K/min in the range from 1340 to 1300°C and 4 K/min at temperatures lower than 1300°C. After solution annealing the ingots were HIP-ed at 1260°C and applied pressure of 200 MPa for 4 h and then they were cooled at rates of 10 K/min in the range from 1260 to 800°C and at 5 K/min in the range from 800 to 300°C.

- **Type C** - solution annealing at 1340°C for 6 min followed by cooling in air. After solution annealing the ingots were HIP-ed at 1260°C and applied pressure of 200 MPa for 4 h and
then cooled at rates of 10 K/min in the range from 1260 to 800°C and at 5 K/min in the range from 800 to 300°C.

2.3. Centrifugal and gravity cast ingots for comparative mechanical testing

Cylindrical ingots with the chemical composition Ti-46Al-8Nb (at.%) were prepared by centrifugal casting with a diameter of 13 mm and length of 110 mm and by gravity casting with a diameter of 16 mm and length of 200 mm into ceramic shell moulds. Induction skull melting (ISM) was used to melt the ingot material to ensure minimum contamination during gravity casting. Details concerning ISM and gravity casting can be found elsewhere [10]. Gravity cast (GC) ingots were subjected to the first step of HIP-ing to close porosity and machined to diameter of 13 mm and length of 95 mm. Both types of ingots were subjected to solution annealing at 1360°C for 1 h and oil quenched to room temperature. After solution annealing, HIP-ing at 1310°C and applied pressure of 150 MPa for 4 h followed by cooling at a rate lower than 10 K/min was applied.

2.4. Mechanical testing

Cylindrical tensile specimens with a diameter of 8 mm and gauge length of 40 mm were lathe machined, high surface quality of gauge section was achieved by grinding and finally by polishing with diamond paste. All room- and high-temperature tensile tests were performed at a strain rate of 1x10^-4 s^-1. During testing, the strain was monitored continuously with a high-temperature extensometer Maytec until the specimen fracture. The extensometer was mounted to a specimen gauge region using ceramic arms touching the specimen surface at a defined load. For high-temperature tensile tests, the specimens were heated in a resistance furnace to a temperature of 850°C at a heating rate of 9 K/min and stabilised at this temperature for 20 min. The test temperature was controlled with a precision of ± 1°C using a thermocouple touching gauge region of the specimen. After fracture, the furnace was open and tensile specimen was cooled to room temperature in air.

Cylindrical creep specimens with a gauge diameter of 6 mm and gauge length of 30 mm were lathe machined from the cast ingots. Final roughness of gauge section was achieved by grinding. Constant load creep tests were performed at temperatures ranging from 700 to 800°C and initial applied stresses ranging from 200 to 400 MPa. The creep temperature was controlled with two thermocouples touching the specimen gauge section and held constant with a precision of ± 1°C for each individual test. The creep deformation was measured with high-temperature extensometers attached to the ledges of creep specimens.

2.5. Microstructure characterization

Microstructure evaluation was performed by optical microscopy (OM), scanning electron microscopy (SEM), backscattered electron microscopy (BSE) and energy-dispersive X-ray spectroscopy (EDX). Samples for metallographic observations were prepared by grinding on abrasive papers and by polishing on diamond pastes. After polishing, the samples were etched in a reagent of 150 ml H_2O, 9 ml HNO_3 and 4.5 ml HF. Size of the grains and interlamellar spacing were measured by computerized image analyser.

3. RESULTS AND DISCUSSION

3.1. Effect of heat treatments on microstructure

Fig. 2 shows the typical microstructure of heat treated Ti-45Al-8Nb-0.2B-0.2C (at.%) ingots used for tensile testing. The microstructure consists of lamellar grains with a mean grain size of 43 μm. The grains contain lamellae of α_2(Ti₃Al) and γ(TiAl) phases with a mean interlamellar α_2-α_2 spacing of λ = 810 nm. Due to alloying with boron, numerous ribbon like
boride particles were formed in the form of local networks, as seen in Fig. 2a. Besides boride particles, Y$_2$O$_3$ particles resulting from ceramic crucible-melt interactions were also found, as seen in Fig. 2b. As shown in Fig. 2c, the cast microporosity was not fully removed by HIP-ing and some pores with a size up to 10 µm still remained in some ingots.

Since the creep deformation behaviour is very sensitive to microstructural parameters, all tested ingots were subjected to quantitative metallographic analysis. Log-normal distribution function was used to determine mean values of interlamellar $\alpha_2$-$\alpha_2$ spacing $\lambda$. Fig. 3 shows the typical microstructure of the ingots with measured mean interlamellar spacing $\lambda$ for Type A (Fig. 3a), Type B (Fig. 3b) and Type C (Fig. 3c) of the applied heat treatments. Fig. 4 shows log-normal distribution curves for grain size $d$ in all ingots subjected to creep testing. The applied heat treatments led to very high reproducibility of measured interlamellar spacing which was significantly affected by the applied cooling rates from solution annealing temperature. On the other hand, the mean grain size for ingots "B" (see Fig. 1a) ranging from 80 to 133 µm is higher than that for the ingots "A" and "C" ranging from 29 to 53 µm, as shown in Fig. 4. This indicates that during casting the cooling rates in shell moulds for ingots "B" were somehow lower than those for the other "A" and "C" ingots.

3.2. Effect of mould filling on tensile properties

During room-temperature tensile testing of Ti-45Al-8Nb-0.2B-0.2C (at.%) alloy all specimens showed zero or very limited tensile ductility to fracture. Due to limited tensile
ductility a probabilistic approach to the prediction of fracture strength is appropriate [11, 12].

Weibull statistical methods are often used when evaluating the properties of flaw-sensitive materials. A Weibull analysis assumes that properties are related to flaws that are distributed at random throughout the material. Therefore, average properties will decrease as the sample volume is increased because a large amount of material has greater probability of containing a large flaw than a smaller volume. This behaviour is valid not only for ceramic but for metallic materials as well. The situation is less severe in a material that exhibits substantial straining before failure since flaw may not lead to any significant reduction in strength if there is sufficient ductility to reduce the stress concentration associated with the flaw. In metallic materials showing certain level of ductility, three parameter type of Weibull plot is usually applied. TiAl based alloys lie somewhere between the brittle behaviour of ceramics and ductile behaviour of metallic materials. In this case, the threshold stress $\sigma_m$ is taken as the yield strength of material. Since the studied Ti-45Al-8Nb-0.2B-0.2C (at.%) alloy showed no plastic deformation to fracture during room-temperature tensile testing, offset yield strength could not be determined. Assuming very brittle type of failure and constant volume of all tensile specimens, the probability of failure $F$ is defined by two parameter form equation as follows:

$$F = 1 - \exp \left[ - \left( \frac{\sigma}{\sigma_0} \right)^{-m} \right]$$

where $\sigma$ is the failure stress, $\sigma_0$ is a characteristic strength defined as the stress at which the probability of failure $F$ is 63.2%. Fig. 5 shows two parameter Weibull plot for all tested tensile specimens. The Weibull modulus determined by regression analysis is 18.2 and 17.8 and the characteristic stress is 648 and 712 MPa for specimens prepared by indirect and direct mould filling, respectively. The correlation coefficients of these fits $r^2$ are better than 0.97. Lower characteristic stress for specimens prepared by indirect mould filling can be explained by layer porosity observed in the as-cast ingots.

All specimens subjected to high-temperature tensile testing at 850°C showed 0.2% offset yield strength (YS) ranging from 480 to 490 MPa, ultimate tensile strength (UTS) from 564 to 602 MPa and tensile elongation to fracture from 0.63 to 1.04 %. However, due to limited number of tensile specimens (5 pieces) the value of Weibull modulus could not be
determined to evaluate statistically the reproducibility of casting and heat treatment processes for the studied alloy.

Fig. 6 shows the typical fracture features of specimens after room-temperature tensile tests. As seen in Fig. 6a, the fracture path follows also the grain boundaries in a direction perpendicular to the fracture surface. Fig. 6b shows the fracture surface with the lamellar grain and few boride particles. Fig. 6c shows longitudinal section in the vicinity of the fracture surface. This figure clearly indicates that the cracks, which were observed on the fracture surface in Fig. 6a, propagate along the interlamellar interfaces.

3.3. Effect of casting technology on room-temperature tensile properties

The typical microstructure of the ingots from Ti-46Al-8Nb (at.%) alloy is shown in Fig. 7. The microstructure is similar to that recently published by Hu et al. [13]. It should be noted that no statistical difference in the microstructure was found between the ingots produced by centrifugal and gravity casting after the heat treatments. The results of tensile tests are summarized in Table 1. While the CC ingots show relatively reproducible values of UTS ranging from 477 to 580 MPa, tensile ductility is limited and exceptionally achieved a value of 0.29%, which was sufficiently high to determine 0.2% offset yield strength. On the other hand, GC ingots show large variation in UTS ranging from 148 to 604 MPa indicating some large defects in the microstructure leading to a premature fracture during tensile testing. Generally, room-temperature ductility of GC ingots ranging from 0 to 0.53%
Table 1. Measured room-temperature 0.2% offset yield strength (YS), ultimate tensile strength (UTS) and plastic elongation to fracture for centrifugally cast (CC) and gravity cast (GC) ingots from Ti-46Al-8Nb (at.%) alloy

<table>
<thead>
<tr>
<th>Specimen No.</th>
<th>YS (MPa)</th>
<th>UTS (MPa)</th>
<th>Elongation (%)</th>
</tr>
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<tbody>
<tr>
<td></td>
<td>CC</td>
<td>GC</td>
<td>CC</td>
</tr>
<tr>
<td>1</td>
<td>-</td>
<td>-</td>
<td>505</td>
</tr>
<tr>
<td>2</td>
<td>-</td>
<td>540</td>
<td>533</td>
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<tr>
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<td>-</td>
<td>583</td>
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<td>562</td>
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</tr>
<tr>
<td>9</td>
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<td>-</td>
<td>542</td>
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</tbody>
</table>

was higher than that measured for CC ones. Better ductility of GC ingots can be explained by a lower oxygen content (about 700 wtppm) when compared to that one in CC ingots (about 1000 wtppm).

3.4. Effect of heat treatments on creep properties

Fig. 8 shows variation of creep rate with the strain for Ti-45Al-8Nb-0.2B-0.2C (at.%) alloy at 750°C. During primary creep stage the creep rate decreases with increasing strain. After reaching a minimum at a strain ranging from 1.6 to 2.4%, the creep rate increases with increasing strain. At studied applied stresses, no steady-state creep stage is observed. The primary creep stage is followed directly by a tertiary creep, as seen in Fig. 8. The minimum creep rate \( \dot{\varepsilon}_{\text{min}} \) and applied stresses \( \sigma \) were fitted to the power law

\[
\dot{\varepsilon}_{\text{min}} = A\sigma^n
\]

where \( A \) is a constant and \( n \) is the stress exponent. Fig. 9 shows the variation of the minimum creep rate with the applied stress for all tested specimens at 750°C. Using linear regression analysis of the creep data, the stress exponent was determined to vary from 5.2 to 5.9. From Fig. 9 is clear that the Type A and Type B of heat treatments characterized by the smallest and highest interlamellar spacing (see Fig. 3) exhibit the lowest and highest minimum creep rates, respectively. Decrease of interlamellar \( \alpha_2-\alpha_2 \) spacing \( \lambda \) decreases the minimum creep rate at a given applied stress and temperature. On the other hand, variation in the grain size in the range from 29 to 133 \( \mu \text{m} \) has no effect on the minimum creep rate indicating that the creep is not controlled by sliding along the grain boundaries at the temperature of 750°C.

Type A of the heat treatment was in addition combined with relaxation annealing at 950°C for 0, 6, 12 and 25 h. Fig. 10 shows the variation of the minimum creep rate with the applied stress for all relaxation annealed specimens. Using linear regression analysis of the creep data, the stress exponent was determined to vary from 5.2 to 7.3. Fig. 11 shows dependence of time to 1% creep deformation on the applied stress at 750°C. It is clear from this figure that the specimens annealed for 12 h show the longest time to 1% creep deformation when compared to that of other specimens at the applied stress of 300 MPa. This difference decreases at higher applied stresses and is negligible at 400 MPa. Microstructure
Fig. 8. Creep deformation curves at 750°C for Ti-45Al-8Nb-0.2B-0.2C (at.%) ingots subjected to Type A of heat treatments.

Fig. 9. Dependence of minimum creep rate on the applied stress for Ti-45Al-8Nb-0.2B-0.2C (at.%) ingots during the creep at 750°C.

Analysis of the annealed specimens before creep testing by the means of transmission electron microscopy showed that the increase of time to 1% creep deformation is connected with a change of dislocation microstructure and a decrease of mobile dislocations.

Due to the longest time to 1% creep deformation at lower applied stresses, further detailed creep study was focused on the creep specimens subjected to relaxation annealing at temperature of 950°C for 12 h. The stress-minimum creep rate-temperature data were also fitted to the Bailey-Norton power law expression

\[
\dot{\varepsilon}_{\text{min}} = B \sigma^n \exp \left(-\frac{Q_a}{RT}\right)
\]

where \( B \) is a constant, \( Q_a \) is the apparent activation energy for creep, \( R \) is the universal gas constant and \( T \) is the absolute temperature. The apparent activation energy for creep \( Q_a \)

Fig. 10. Dependence of minimum creep rate on the applied stress for Ti-45Al-8Nb-0.2B-0.2C (at.%) ingots at 750°C. The annealing times and stress exponents are indicated in the figure.

Fig. 11. Dependence of time to 1% creep deformation on the applied stress for Ti-45Al-8Nb-0.2B-0.2C (at.%) ingots at 750°C. The annealing times are indicated in the figure.
calculated for five different applied stresses at three different temperatures is 406 ± 9 kJ/mol. Regression analysis of the creep data yields an equation for the minimum creep rate in the form

\[
\dot{\varepsilon}_{\text{min}} = 0.362 \sigma^{5.5} \exp \left( -\frac{406000 \text{ J/mol}}{RT} \right)
\]

The correlation coefficient \( r^2 \) of this fit is equal to 0.97. Eq.(4) allows to predict the minimum creep rate as a function of the applied stress and temperature. However, the validity of this equation is limited to the studied creep conditions. This equation cannot be used for lower applied stresses or different temperatures without knowing the mechanisms controlling the creep deformation at assumed creep conditions.

The creep characteristics including the stress exponent and activation energy for creep of the studied alloy can be compared with literature data for various TiAl-based alloys where the stress exponent was determined to vary from about 2 at low stresses to about 10 at high stresses. It should be noted that the stress exponent of 5.5 measured in this study is lower that of 6.8 reported by Recina et al. [14] for intermetallic alloy with a nominal composition Ti-48Al-2W-0.5Si (at.%) with pseudoduplex microstructure or that of 7.3 determined by Lapin and Nazmy [15] for Ti-46Al-2W-0.5Si (at.%) alloy. On the other hand it is comparable with a value of 5 determined by Lapin [16] for Ti-45.2Al-2W-0.6Si-0.7B (%) alloy. For the activation energy for creep, we can refer to the values ranging from 230 to 430 kJ/mol reported in the previous studies. The apparent activation energy for creep of \( Q_a = 406 \) kJ/mol is higher than that of 288-312 kJ/mol and 395 kJ/mol determined for self-diffusion of Ti and Al in TiAl, respectively [17]. For self-diffusion of Ti and Al in TiAl, we can refer to the values of 250-295 kJ/mol and 358 kJ/mol, respectively [17].

4. CONCLUSIONS

The study of the effect of several processing parameters such as casting technology, arrangement of gating system during centrifugal casting and post-solidification heat treatments on the microstructure and mechanical properties of Ti-45Al-8Nb-0.2C-0.2B and Ti-46Al-8Nb (at.%) intermetallic alloys suggest following conclusions:

1. Change of gating system during centrifugal casting into shell ceramic moulds has no effect on the reproducibility of room-temperature mechanical properties which were evaluated statistically using Weibull statistics. Decrease of UTS of ingots prepared by indirect mould filling is caused by layer porosity within the ingots.

2. Room-temperature ductility of GC ingots is superior to that measured in CC ingots due to lower oxygen content. On the other hand, CC ingots showed higher reproducibility of room-temperature tensile properties.

3. Various types of post-solidification heat treatments consisting of high temperature isostatic pressing, solution annealing, cooling at various cooling rates from solution annealing temperature and stabilization annealing were performed. It was found that a decrease of \( \alpha_2-\alpha_2 \) interlamellar spacing leads to higher creep resistance of CC ingots. Relaxation annealing affects time to 1% creep deformation which is related to a change of dislocation structure and density of mobile dislocations.

4. The parameters of kinetic equation for minimum creep rate are defined for temperature range from 973 to 1073 K and applied stresses varying from 200 to 400 MPa for Ti-45Al-8Nb-0.2C-0.2B (at.%) alloy.

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