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# Improvement and application of $Y_2O_3$ directional solidification crucible

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**Abstract** In order to satisfy the drastic temperature change and high chemical activity in directional solidification of Nb–Si based alloys, $Y_2O_3$ crucible is demanded to possess high thermal shock resistance and erosion resistance. This paper improved the sintering degree and density of $Y_2O_3$ crucible by optimizing the sintering temperature and time, and its practical application performance was investigated. $Y_2O_3$ grains gathered with the increase of sintering temperature and time, and the contact area enlarged, resulting in the open pores being changed into closed pores. The higher density caused the improvement of erosion resistance of $Y_2O_3$ crucibles. However, excessive density weakened the thermal shock resistance. Considering high-temperature strength, erosion resistance, thermal shock resistance and costs, optimum sintering temperature and time of $Y_2O_3$ directional solidification crucible were 1800 °C and 120 min, respectively, and the porosity was 20%. Improved $Y_2O_3$ crucible has been successfully applied to directional solidification of Nb–Si based alloys, and significantly reduced the oxygen contamination. Slight interaction occurred between Hf and $Y_2O_3$, but no obvious dissolution, penetration or erosion was found, showing good erosion resistance and thermal shock resistance.


## 1. Introduction

The next-generation aeroengine blades need higher temperature-bearing structure material to substitute nickel-based superalloy which has reached the limit of operating temperature. With higher melting point, relatively lower density and good processability, Nb–Si based alloys show great promise for application in aero-engine blades operating at 1200–1400 °C.[1] Directional solidification technology enables the alloy grains well-aligned in certain direction, so that the creep rupture life and thermal fatigue strength would be greatly improved.[2] At present, directional solidification is widely used in the preparation process of aeroengine blades.

However, directional solidification of Nb–Si based alloys excludes almost all of traditional crucible materials because

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of the high melting point ($\geqslant$1800 °C) and high active elements, such as Ti, Hf and Y.[3] The directional solidification crucibles not only require sufficient refractoriness and high-temperature strength, but also should have excellent thermal shock resistance which could bear rapid cooling from ~2000 °C to room temperature. Meanwhile, the crucible materials should present good erosion resistance and chemical stability, no reaction or only slight reaction occurring with the alloying elements.

Based on theoretical analysis, $Y_2O_3$ is found more thermodynamically stable than traditional ceramic materials, such as CaO, $ZrO_2$, MgO, $Al_2O_3$ and $SiO_2$.[4] Experimental results showed that relative stability of rare earth oxides follows an increasing sequence of $CeO_2$–$ZrO_2$–$Gd_2O_3$–didymium oxide–$Sm_2O_3$–$Nd_2O_3$–$Y_2O_3$.[5] Kuang et al.[6] found that pure $Y_2O_3$ was very promising in application to melting and casting high activity alloys among several refractory materials. Our previous research has applied $Y_2O_3$ to the vacuum induction melting process and investment casting process.[7] However, $Y_2O_3$ suffers from two drawbacks: inherently poor thermal shock resistance and high cost. Thus, crucibles/moulds coated by a $Y_2O_3$ protective coating seem to be effective and economical, but its operating temperature is completely limited by the basic crucible material.[8,9] Moreover, it seems inevitable that the molten alloys would interact with the crucible materials[10–12] and the intensity is closely related to the crucible material and its density.[13] Currently, sintering temperature of $Y_2O_3$ crucible is 1550 °C[14] −1650 °C,[15] much lower than its melting point ($\geqslant$2400 °C), which means that excessive porosity makes $Y_2O_3$ crucible suffer more erosion and dissolution by the molten alloys.[16]

The directional solidification of Nb–Si based alloys needs an improvement of $Y_2O_3$ crucible to increase density and erosion resistance against high active melt. Meanwhile, the crucible should have sufficient thermal shock resistance to meet the rapid temperature changes during the process. In this paper, optimized sintering degree and density of $Y_2O_3$ crucible were studied by controlling the sintering temperature and time, and the practical application performance was investigated.

## 2. Experimental

The crucibles were made by gelcasting with the dimensions of 20 mm × 180 mm. For gelcasting, acrylamide [$C_2H_3CONH_2$] (AM) was used as a monomer, N,N′-methylenebisacrylamide [$(C_2H_3CONH)_2CH_2$] (MBAM) as a coupling agent, N,N,N′, N′-tetramethylethylenediamine (TEMED) as a catalyst, ammonium persulphate as an initiator and ammonium polyacrylate as a dispersant. The $Y_2O_3$ powders were mixed in a weight proportion of 1:1:1 with particle sizes of $<$10 μm, 30–60 μm and 100–150 μm, respectively. Binder burnout was operated at 200–600 °C for 6 h. Sintering was performed in a self-made vacuum sintering furnace. The porosity was measured by Archimedes' principle.

Directional solidifications were performed in a Bridgman furnace. Before heating, the furnace chamber was evacuated to $6 \times 10^{-3}$ Pa and then backfilled with high purity argon up to 0.05 MPa. The heating temperature was 1900 °C and kept isothermally for 20 min. Then the bars were directionally solidified with a withdrawal rate of $1 \times 10^{-4}$ m/s.

Thermal shock resistance was evaluated by the times of thermal cycle. The samples were placed in a resistance furnace at 1400 °C for 30 min, and then cooled immediately in room-temperature water for 3 min. After drying in a thermostatic oven at 150 °C for 60 min, the samples were repeated with this process until the cracks appeared.

Scanning electron microscopy (SEM, FEI Quanta600, USA) was used to investigate the microstructure. Energy dispersive spectrometry (EDS, Oxford INCAPentaFET-x3) was used to analyze the chemical composition. The oxygen contents were measured by the inert gas infrared–thermal conductivity technique (IGI, LECO TC-436).

## 3. Results and discussion

### 3.1. Effect of sintering temperature on microstructure

Fig. 1 shows the microstructures of $Y_2O_3$ crucible at sintering temperature ($T$) of 1650 °C, 1800 °C and 1900 °C for 120 min, and the porosities are 38%, 20% and 17%, respectively. At the sintering temperature of 1650 °C, only fine $Y_2O_3$ powders are preliminarily sintered. Apparent gaps between fine powders and coarse powders demonstrate that they are not well combined yet. Once the crucible cracked during the directional solidification, the cracks will propagate along the gaps and fracture the crucible. At the sintering temperature of 1800 °C, fine powders and coarse powders are integrally sintered, and more contact areas are formed between them, suggesting that the strength of crucible improves correspondingly.[17] However, it could be found that there are still a certain number of pores existing in the inner of crucible. At the sintering temperature of 1900 °C, fine powders and coarse powders are entirely sintered. The interconnected pores in Fig. 1(b) become closed ones in Fig. 1(c) and its number declines. Besides, as shown in Fig. 2, the increasing sintering time ($t$) makes the microstructure of $Y_2O_3$ crucible present similar variation trend, and its porosities are 25%, 21% and 18% for 90 min, 120 min and 150 min, respectively. The above evolutional results suggest that the increased sintering temperature and time cause the powders gradually to gather, the particle contact area to expand and the binding force to enhance, resulting in the strength improvement of $Y_2O_3$ crucible.

Density closely relates to the erosion resistance of $Y_2O_3$ crucible.[18] During the melting, the melt will penetrate along the pores or cracks into the ceramic structure, and then interact with $Y_2O_3$. As a result, a metamorphic layer with different properties from original material is formed. When directional solidification proceeds, the dramatic temperature change leads to the metamorphic layer splintering and flaking into the alloy melt, and the deeper penetration implies thicker metamorphic layer and more contamination. The melt penetration depth ($x$) into the $Y_2O_3$ crucible can be estimated by[18]:

$$X = \sqrt{\frac{r\sigma\cos\theta}{2\eta}\tau} \tag{1}$$

where $\sigma$ is the surface tension of the melt, $\theta$ the wetting angle, $\eta$ the viscosity of the melt, $r$ the radius of pore, and $\tau$ the time. Under the conditions of the other parameters fixed, reduced radius of pore ($r$) could decrease the depth of penetration, thereby enhancing the erosion resistance of $Y_2O_3$ crucible