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High-temperature wettability and interactions between Hf-containing NbSi-based alloys and Y_2O_3 ceramics with various microstructures

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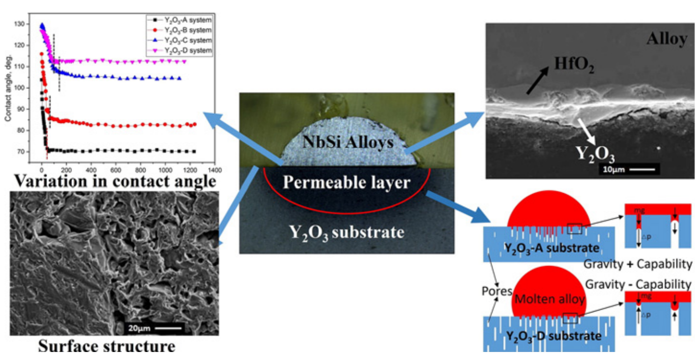
HIGHLIGHTS

- Mechanisms of high-temperature wettability and interactions between the molten NbSi alloys and Y_2O_3 ceramic were clarified.
- Y_2O_3 with open porosity level of about 20.7% was the most beneficial for DS casting of high-purity NbSi alloys.
- Wetting kinetics analyses indicated that the movement of triple line was driven by the formation of 1–5 μm HfO_2 layer.
- The equilibrium contact angle changed from 70.2° to 112.6° with the porosity of Y_2O_3 increasing from 3.6% to 27.2%.

GRAPHICAL ABSTRACT



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ABSTRACT

To obtain improved mould shells with appropriate microstructures for directional solidification casting of NbSi-based alloys, Y_2O_3 ceramics with various microstructures were designed and characterized. The mechanisms of high-temperature wettability and interactions between NbSi-based alloys and Y_2O_3 ceramics were studied. The results showed that there was a characteristic transition in the wettability between the molten alloys and ceramics when the Y_2O_3 microstructure changed. The spreading rate gradually decreased and the apparent equilibrium contact angle changed from 70.2° to 112.6° with the level of open porosity of Y_2O_3 ceramics increasing from 3.6% to 27.2%. Wetting kinetics analyses indicated that the movement of the triple line during the wetting process was driven by the formation of continuous HfO_2 reaction layer in the alloy-ceramic interface. The content of HfO_2 particles inner the alloy matrix with the level of open porosity of Y_2O_3 ceramics increasing from 3.6% to 27.2% initially gradually decreased and subsequently increased. Y_2O_3 with open porosity level of about 20.7% were the most beneficial for directional solidification casting of high-purity Hf-containing NbSi-based alloys.

1. Introduction

NbSi-based alloys, which find potential applications as high-temperature structural materials for new-generation aeroengine components with high thrust-weight ratios, have been extensively studied because of their high specific strength and good performance at temperatures of 1200–1400 °C [1–3]. However, their high brittleness and low room-temperature fracture toughness limit their practical applications [4,5]. Accordingly, Sekido [6] suggested that NbSi-based alloys with well-aligned regular structures and excellent mechanical properties could be obtained using ceramic moulds via the directional solidification (DS) technique. Bewlay [1] found that the use of alloying

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elements Hf and Ti, which are also known as active elements, was effective for improving the creep behavior and oxidation resistance of NbSi-based alloys. However, during the DS process, interfacial reactions occur between the active elements in the NbSi-based alloys and the ceramic moulds. Reaction products precipitate at the alloy-ceramic interface and within the alloy matrix [7,8], consequently deteriorating the microstructure and mechanical properties of the resultant NbSi-based alloys. Therefore, when manufacturing mould shells, it is important to select a thermodynamically stable and chemically inert refractory material.

Based on theoretical analysis, Y_2O_3 is considered the most thermodynamically stable among common refractory materials (SiO_2, MgO, Al_2O_3, CaO, ZrO_2, and Y_2O_3) [9,10]. As reported by Wang et al., pure Y_2O_3 and Y_2O_3 doped with CaO + ZrO_2 and MgO + ZrO_2 were used for the casting of NbSi-based alloys, and the pure Y_2O_3 crucible performed better [11]. Even Y_2O_3 has been widely used in the DS casting of NbSi-based alloys [12–17], reactions between the Y_2O_3 moulds and molten NbSi-based alloys are inevitable. Ma et al. showed that Y_2O_3 reacted with Hf to form HfO_2 when NbSi-based alloys were directionally solidified in Y_2O_3 moulds [8,18]. Wang et al. also reported that a reaction layer consisting of HfO_2 and Y_2O_3 was observed when NbSi-based alloys were re-melted in Y_2O_3 moulds [11]. However, although some reaction products between NbSi-based alloys and Y_2O_3 moulds have been detected, the associated kinetic mechanisms have not been fully understood and the information of wetting behavior under high temperature between them is severely deficient.

As reported by N. Sobczak, study of the wetting behavior in liquid metal/solid ceramic systems at elevated temperatures was critical for high-temperature liquid phase materials processing stimulated by the needs of modern metallurgy and foundry industry [19]. The wettability of ceramics with regard to molten metal is of importance to understand the reaction mechanisms between them. For instance, the mechanisms of the interfacial reactions between molten metals of Al, Ni, Sn and ceramics of MgO, Al_2O_3, B_4C/TiC were analyzed by means of sessile drop wetting technique [20–23]. A recent work by Wu et al. showed that ceramic substrates with rough surfaces significantly promoted the wetting and spreading of molten alloys in non-reactive systems, however, they resulted in poor wettability in reactive systems [24]. Thus, the surface structure of ceramics is of importance with regard to the wetting process. Lin also studied the effect of porosity in Al_2O_3 ceramic substrates on the brazing and interfacial reactions with a AgCuTi alloy [25]. The surfacial reaction did not show appreciable dependence on the porosity of the ceramic substrates when the brazing procedure was under 0.4 N normal load and the liquid alloy could not spread spontaneously on the substrates.

In general, the temperature gradient in DS furnace could be as great as 3000–7000 K/m and the DS moulds need to be quenched rapidly (within 2 s) from the furnace hot zone to a molten Ga-In-Sn alloy pool. The ceramic moulds for DS are required to have an excellent thermal shock resistance. Therefore, the ceramic moulds with appropriate microstructures are essential [8]. This will contribute to controlling appropriate levels of wettability and reducing interfacial reactions with the NbSi-based alloys during casting to obtain high-quality NbSi-based alloy castings. Unfortunately, the influence of microstructures of ceramics on high-temperature wettability and interactions between NbSi-based alloys and Y_2O_3 moulds during DS process have rarely been investigated.

Table 1
Parameters related to Y_2O_3 substrates with various microstructures.

Number	Y_2O_3-A	Y_2O_3-B	Y_2O_3-C	Y_2O_3-D
Open porosity level	3.6%	11.8%	20.7%	27.2%
Grinding time (min)	40	20	10	5
PVA binder (5 wt%)	0.6 g	0.6 g	0.6 g	0.6 g
Molding pressure (MPa)	260	260	160	120

Therefore, in the present study, four Y_2O_3 ceramic substrates with open porosity level of 3.6%, 11.8%, 20.7%, and 27.2% were self-designed and characterized. The influence of various microstructures of Y_2O_3 ceramics on the high-temperature wettability and interfacial reactions with regard to NbSi-based alloys and Y_2O_3 ceramics were studied. Meanwhile, the corresponding mechanisms were clarified.

2. Experimental procedure

The NbSi-based alloys, with a nominal composition of Nb-16Si-22Ti-2Al-2Hf-17Cr (at.%), were prepared by the arc-melting method with appropriate amounts of high purity metals, Nb (99.87%), Si (99.50%), Ti (99.76%), Al (99.99%), Hf(99.9%), and Cr (99.98%), as raw materials, in a water-cooled Cu crucible. The $3 \times 3 \times 3$ mm^3 cubes (about 0.16 g) obtained from the central part of the master alloy were used for the wetting experiments. The Y_2O_3 ceramic substrates in dimensions of Φ21 mm × 10 mm were prepared by the dry-pressing method and sintered at 1913 K. The raw material consisted of a mixture of 5 μm, 60–80 mesh, 200 mesh and 325 mesh Y_2O_3 particles (purity 99.9%). The various microstructures of the substrates were controlled by changing grinding period and molding pressure [26,27],and the specific parameters were designed and given in Table 1. The open porosity level of each as-obtained substrate was measured according to GB/T 1966–1996 [28]. The surface line roughness (Ra) of each ceramic substrate was measured using a LEXT OLS4000 3D laser (12.5 mm, performed 3 times). The microstructure of the ceramic surface was observed using a scanning electron microscope (SEM, JSM6010, Japan).

The wetting experiments were performed using an improved sessile-drop equipment, as shown in Fig. 1, which offered a distinct advantage in measuring initial contact angles and preventing the occurrence of the interfacial reactions between NbSi-based alloys and Y_2O_3 ceramic substrates during heating. The whole experimental process was in a deeply purified Ar (99.999%) atmosphere to prevent active elements from evaporation and oxidation, and the oxygen partial pressure in this condition should be lower than 10^{-8} Pa [29]. When the temperature was 1873 K, the alloy, which was located at the top of the equipment, was dropped. The spreading process was recorded by the CCD camera. The contact angles were directly measured from the captured drop profiles using an axisymmetric-drop-shape analysis (ADSA) program with an error within ±1° [30,31].

After the wetting experiment, the solidified samples were embedded in resin and then cut and polished for microstructural observations. The microstructure of the alloys was analyzed by SEM. The chemical

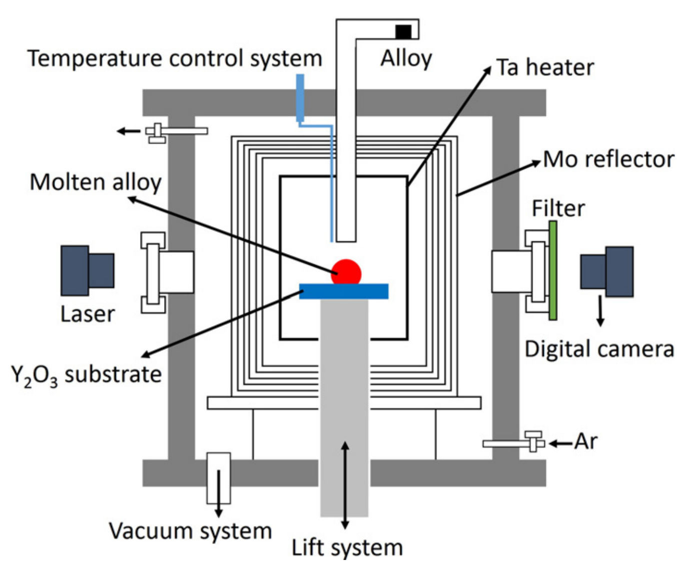

Fig. 1. Schematic diagram of the improved sessile drop equipment.