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Effect of hydrogen on interfacial reaction between TiAl alloy melt and graphite mold

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ABSTRACT

The effect of hydrogen on the interfacial reaction between Ti–47Al alloy melt and graphite mold was studied by plasma-arc melting in hydrogen atmosphere. Especially, reduction of recarburization which is harmful to the mechanical properties of TiAl alloys has been investigated. The carbon concentration in the interface of Ti–47Al alloy, as well as the corresponding reaction distance decreases rapidly with increasing hydrogen partial pressure. Activated hydrogen atoms dissociated in high-temperature arc probably take part in the interfacial reactions, represented by $[C] + 4[H] = CH_4$.

1. Introduction

TiAl alloys (TiAl intermetallics) have been considered to be the most promising alternative light-weight heat-resistant material to conventional superalloys with the aim to reduce the rotational and reciprocating engine components [1–4]. However, it is hard to apply conventional hot forging process to valve production using TiAl alloys due to its inherent poor machinability and deformability [5]. So far, precision casting has been reported as a candidate manufacturing process for TiAl because of its low cost and the ability to form any shape [5]. However, the casting mold is so limited for TiAl alloys, because the titanium in molten TiAl alloys is extremely active and can react with nearly all the common casting molds, resulting in surface-level chemical reaction.

Only several special kinds of mold can be used for the casting of TiAl alloys, such as Y_2O_3, ZrO_2, CaO and graphite. The use of Y_2O_3 and ZrO_2 has been limited by their high cost, and CaO is apt to absorb moisture. Among them, the graphite is cheap and has a good formability, so it is an ideal alternative as the casting mold for TiAl alloy [6]. But the graphite has an interfacial reaction with TiAl alloy melt leading to recarburization [7] and form a thick carbon-rich layer including α brittle phase [8]. Although sand blasting or acid pickling is used to remove the layer, it is difficult to control because of the non-uniform layer due to the different reaction condition of the interface. The layer will affect the property of TiAl castings if it is not got rid of completely. Conversely, the casting precision will be affected if too much is removed [8]. In this regard, reducing the interfacial reaction is of great significance for the application of graphite mold.

Some researchers have studied its interfacial reaction under different experimental conditions. For example, Luo et al. [9] studied the interfacial reactions between Ti–47Al alloy and graphite crucible during directional solidification, and Tan et al. [10] investigated the reaction of graphite and Al–Ti alloys. These limited researches, however, just focused on the reaction of Ti and graphite, instead of providing effective methods on controlling the interfacial reaction so far. In fact, it is difficult to reduce its interfacial reaction due to the high melting point, vacuum melting and strong affinity for carbon. But to broaden the usage of graphite mold, a simple and practical method to reduce its interfacial reaction has been required.

Hydrogen may be attractive for controlling the interfacial reaction between the TiAl alloy melt and graphite mold. Hydrogen was once regarded as a detrimental element for inducing embrittlement. However, extensive research recently revealed that hydrogen has many positive effects, such as modifying the structures and purifying the molten alloys [11]. Unlike the commonly added elements, the hydrogen can be easily removed by vacuum annealing [11–16]. But most studies ignore its effect on casting properties, especially the interfacial reaction between alloy melt and mold.

In this study, a novel method, based on the hydrogen-involved interfacial reaction, was introduced to the TiAl alloy melt to

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reduce the recarburization of the graphite mold. The effect of hydrogen on the interfacial reaction between Ti–47Al (at.%) alloy melt and graphite mold was examined and its mechanism was discussed.

2. Experimental

The main apparatus for melting, hydrogenation and fluidity test is a plasma-arc melting furnace. It consists of a non-consumable tungsten electrode, a water-cooled copper crucible, and a graphite mold. The water-cooled crucible was used to avoid the reaction and contamination of the alloy during melting.

Ti–47Al binary alloys were selected for this study. Ti–Al alloy ingots were prepared first by arc melting under a pure argon atmosphere. Ti sponge and high-purity Al (>99.99%) were used as the starting materials. The alloy ingots were melted four to five times to improve chemical homogeneity.

The hydrogenation process was conducted as previously studied [17]. The Ti–47Al alloy ingots were remelted in a mixture of hydrogen and argon. During the melting process, the hydrogen dissociated and diffused into the melt until saturation. Four levels of hydrogen partial pressure were chosen, which were 0, 5, 10 and 15 kPa, respectively. The hydrogen concentration absorbed by the TiAl alloy melt can be determined according to a previous study [18].

When the hydrogen was saturated in the alloy melt, the alloy melt was poured into a graphite mold, as shown in Fig. 1(a). The melting was under an identical condition each time to provide a same melting superheat. Fig. 1(b) shows the casting, in which the selected samples were cut from the middle using an electric discharge machine. The place for measurement of carbon concentration is along the center line perpendicular to the interface, shown by the white line in Fig. 1(b).

The images of the castings were observed using a digital camera. Microstructure characterization and chemical composition were analyzed by scanning electron microscopy (Quanta 200, FEI) with an energy disperse spectrometer (EDAX genesis).

3. Results

Fig. 2 shows the profile of Ti–47Al castings melted and subsequently poured with different hydrogen partial pressures. The surface of the specimen without hydrogen presents dark-colored. It is mainly due to the carbonization reaction between the TiAl melt and graphite. However, the surface color becomes lighter with increasing hydrogen partial pressure, indicating the effect of hydrogen on the interface (surface of castings). The flow marks on the surface are caused by the chilling action of graphite mold.

The carbon concentration distribution of Ti–47Al alloys along the normal direction with different hydrogen partial pressure was examined, as shown in Fig. 3. Table 1 lists the carbon concentration of each detected point in Fig. 3, in which the overall content was measured. It can be seen that the carbon concentration in the interface is very high; however, it becomes lower with the increasing distance from the interface. This is caused by the absorption and diffusion of carbon in the alloy melt from the graphite mold, which involves the reaction of carbon and alloy melt. The carbon diffusion becomes increasingly difficult towards the internal alloy. When the distance from the interface reaches a certain value, the carbon will vanish. This distance is defined as reaction distance, which can be characterized as the reaction extent between the alloy melt and mold. After hydrogenation, on the one hand, the carbon concentration in the interface is reduced with increasing hydrogen partial pressure. When the hydrogen partial pressure is 15 kPa, the carbon concentration is decreased from 33.55% to 17.66%, about 47.4% of decreasing amplitude. On the other hand, the reaction distance, meanwhile, decreases with increasing hydrogen partial pressure, as shown in Fig. 4. It is reported that graphite can react with molten Ti to form carbide, or some of which can be dissolved into the alloy, occurring at the interface [10]. Therefore, it is considered that the interfacial reaction between the graphite mold and TiAl alloy melt is restrained by the hydrogen.

4. Discussion

In order to discuss the hydrogen-induced decarburization mechanism, it is necessary to understand what causes recarburization of TiAl alloy. It can be summarized in three aspects: deciduous carbon from the mold, dissolved carbon, and reacted carbide. After the alloy melt is poured into the mold, they contact with each other within a short time, so the recarburization is mainly occurring in the interface. First, some graphite may fall off into the alloy melt from the mold wall by the heat or physical impact of alloy melt. The deciduous graphite particles move into the alloy melt according to the Brownian movement until the viscosity of the

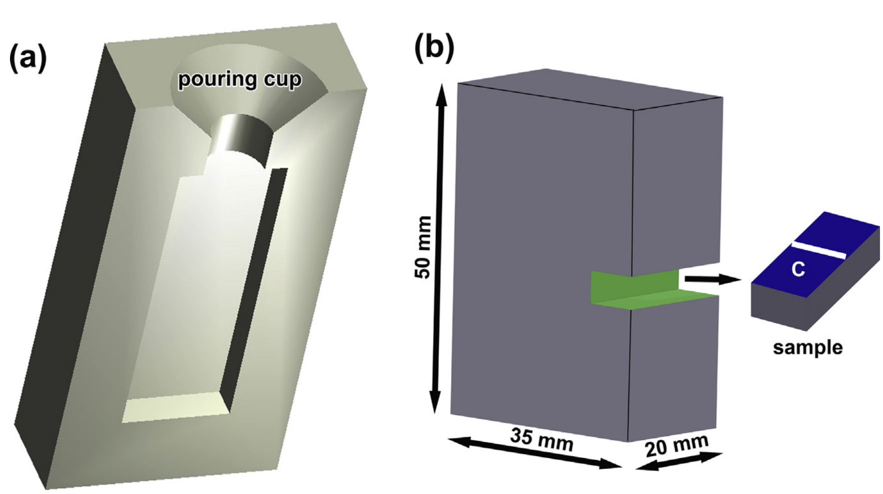

Fig. 1. Graphite mold (a) and casting and the selected sample (b).