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Preparation of *in situ* Al_3Nb-NbB_2-NbC/Al inoculant and its effect on microstructures and properties of weldable Al-Cu-Mn alloy

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ABSTRACT

The *in-situ* Al_3Nb-NbB_2-NbC/Al inoculant (Al-5Nb-0.75B_4C) ingots and ribbons were prepared with Al-Nb and B_4C powers to refine the Al-Cu-Mn alloy. The Al_3Nb-NbB_2-NbC/Al inoculant ribbons showed excellent inoculant effect, which modified the dendrite crystals to equiaxed crystals and refined the grains from ~100 µm to ~30 µm. The transmission electron microscopy (TEM) images of the Al-Cu-Mn alloys reveal that the θ′ phases were refined from ~83 nm to ~40 nm and their number densities were also increased significantly. The selected area electron diffraction (SAED) patterns indicate that there are orientation relationships (ORs) between α-Al and Al_3Nb, NbB_2 and NbC, which suggest that these particles possess nucleation potency to α-Al. After inoculated by 1% Al_3Nb-NbB_2-NbC/Al ribbons, the ultimate tensile strength and elongation of the Al-Cu-Mn alloy were improved from 435 MPa and 8.4% to 514 MPa and 11.5%, respectively. The welding microstructure and property of the Al-Cu-Mn alloy inoculated by 1% Al_3Nb-NbB_2-NbC/Al inoculant were also improved significantly.

1. Introduction

Al-Cu-Mn alloy is a kind of weldable aluminum alloy with light weight, high strength, good ductility, fracture toughness, excellent formability and satisfactory corrosion resistance, thus it is widely used in the aerospace, armored vehicle, radar and other industrial sectors [1–4]. In the process of welding, both the base metal and the filler metal will participate in the metallurgy process, so the microstructure and property of the filler metal directly determine the quality of the welding joint. Optimizing the microstructure of the filler metal can obviously improve its mechanical properties, thus improving its welding properties [5,6]. Grain refinement is the simple and effective methods to improve the microstructure and mechanical properties of the aluminum alloys.

Grain refinement by the addition of grain refiners has been widely used in industrial production, since it can facilitate the casting process and reduce cast defects, and consequently obtain excellent as-cast structure [7,8]. Al-5Ti-B is one of the most commonly used grain refiners and it is effective for most of the aluminum alloy. Up to now, many theories have been proposed about its refinement mechanism; however, there is no consensus on the exact mechanism [9]. Recently, Fan et al. [10] found that there is a $(112)_{Al3Ti}$ two-dimensional compound (2DC) on the $(0001)_{TiB2}$ surface, which can significantly increase the nucleation potency of the TiB_2 particles. Besides, for the Al-Ti-B refiners, they also possess some shortcomings, for instance, they can be poisoned by Si, Zr, V and Cr etc. [11–14]. In this study, the experimental Al-Cu-Mn alloy contains Zr, V and Cr elements, so the Al-5Ti-B is not the ideal refiner for it. There are also many other reports on the refinement of the Al-Cu based alloy, for example, Wang et al. studied the effect of Al-Ti-C master alloys on the Al-5Cu alloy and the results inidicated that the minimum grain size of the Al-5Cu alloy was refined to ~50 µm; however, the refined grains showed equiaxed dendrite structure rather than perfect equiaxed crystal [15]. Furthermore, Yu et al. found that Al_3Ti can react with Al_2Cu to form Ti(Al, Cu)$_2$, resulting in the refinement fading of the Al-5Ti-0.4 C master alloy [16]; in addition, TiC is not stable at high temperature, because it can react with liquid Al to form Al_4C_3, resulting in the refinement fading [17]. The rare earth element Sc was also used to refine the Al-Cu based alloys, it can modify the dendrite crystals to perfect equiaxed crystals and decrease the average grain size to ~40 µm [18]; however, Sc is generally very expensive, so that limits its application in industrial production.

Many studies have proved that if the ceramic particles can act as the heterogeneous nuclei, they must meet the following conditions [19,20]: (1) high chemical stability to avoid their interaction with the alloying elements; (2) high melting point to prevent their melting in the melt; (3) low lattice misfit between the substrates and the solid to reduce the

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interfacial energy between the substrates and the solid. The melt points of Al_3Nb, NbB_2 and NbC are 1680 °C, 3036 °C and 3490 °C, respectively [21], so their melt points are high enough that they do not melt in aluminum melt. Al_3Nb, NbB_2 and NbC have the similar lattice structures and constants with Al_3Ti, TiB_2 and TiC, respectively. Therefore, in order to overcome the shortcomings of the Al-Ti-B refiner, Bolzoni et al. prepared the Al-Nb-B inoculant with Nb powers and B powers, as well as KBF_4 flux; meanwhile, studied the effects of the Al-Nb-B inoculants on the Al-Si alloys [22–24]. It suggested that the Al-Nb-B inoculants could obviously refine the Al-Si alloys and the poisoning phenomenon caused by the Si element did not occur. Therefore, we attempted to use the Al-2Nb-2B master alloy to refine the Al-Cu-Mn alloy, but its refining effect was non-ideal. The reasons for the above differences can be speculated as follows: in the Al-2Nb-2B master alloy, the content of the boronis is relatively high; therefore, apart from forming NbB_2, there is also some excess boron to form AlB_2 with Al. It has been proved that B can refine the Al-Si alloy effectively through creating a layer of SiB_6 at the interface between AlB_2 and Al. However, without the presence of Si, the AlB_2 cannot refine the α-Al alone [25,26]. In the Al-Cu-Mn alloy, the content of Si is trace, so the AlB_2 can not work. On the other hand, the poor wettability of B in liquid Al increases the preparation difficult of Al-Nb-B refiners; furthermore, the Nb/B ratio is not easy to control, due to the oxidation of Nb and the incomplete transformation of B [24,27]. Therefore, through the above analysis, we prepared Al-5Nb-0.75B_4C inoculant with Al-Nb and B_4C powers using vacuum arc furance. Meanwhile, its effects on the microstructure and properties of the Al-Cu-Mn alloy were also studied in detail.

The Al-Cu-Mn alloy is a kind of weldable aluminum alloy, so in this paper, effect of the Al_3Nb-NbB_2-NbC/Al inoculant on its welding microstructure and property are also studied.

2. Materials and methods

The Al_3Nb-NbB_2-NbC/Al inoculant was prepared with commercial-purity Al (99.7%, all compositions quoted in this work were in wt% unless otherwise stated), Al-Nb and B_4C powers using vacuum arc furnace. The inoculant ingot was made into ribbons using vacuum rapidly quenched furnace.

Al-Cu-Mn alloy was prepared with commercial-purity Al, commercial-purity Cu (99.9%), commercial-purity Ti (99.9%), Al-Mn, Al-Mg, Al-V and Al-Zr. These raw materials were melted with the resistance furnace at 750 °C. When the alloy was melted thoroughly, pour it into the steel mold to cast into ingot. For the purpose of assessing the effectiveness of the inoculant, different amounts (0.5% and 1%) of inoculant ribbons were added into the Al-Cu-Mn alloys under the same conditions. The metallographic specimens were sectioned from the center region of the uninoculated and inoculated Al-Cu-Mn ingots, respectively. After being mechanical polished, the specimens were anodized at 25 V in a 25 g/L HBF_4-distilled water solution for optical metallography. The experimental Al-Cu-Mn alloys were solution treated at 525 °C for 24 h followed by hot-rolled at 450 °C and the deformation was 80%, then held for 1 h at 525 °C followed by water quenching, finally, the artificial aging treatment was performed at 165 °C for 14 h.

TIG welding was used in the welding experiment, which used the as cast uninoculated and inoculated Al-Cu-Mn alloys as filler metals, and chose T4-2A12 aluminum alloy sheet with a thickness of 3 mm as the base metal. The welding parameters were as follows: welding current was 120 A, travel speed was 120 mm/min, wire diameter was 2 mm, gas flow rate was 15 L/min and electrode diameter was 3.2 mm. The cross sections of joints perpendicular to the welding direction were used to analyze the microstructure.

The X-ray diffraction (XRD) was used to identify the phases of the inoculant. The scanning electron microscopy (SEM) with EDAX energy-dispersive spectrometry (EDS) was used to observe the microstructure of the inoculant and analyze the element composition of the second phase particles in the inoculant. For the purpose of surveying the

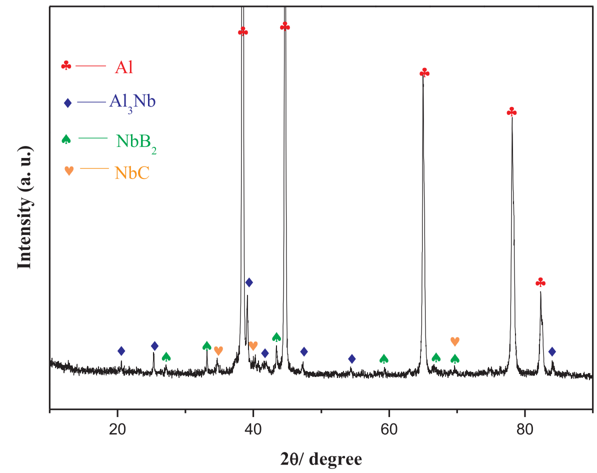

Fig. 1. XRD patterns of Al_3Nb-NbB_2-NbC/Al inoculant.

inoculant effect of the inoculant, the high scope video microscope (HSVM, KH-2200) was used. The transmission electron microscope (TEM) was used to analyze the ORs between α-Al and Al_3Nb, NbB_2 and NbC, and analyze the effect of the inoculant on the θ′ phases. The mechanical properties were tested by universal testing machine and microhardness tester.

3. Results and analysis

3.1. Microstructure and phase composition of the inoculant

Fig. 1 shows the XRD patterns of the prepared inoculant, it can be seen that it mainly contains α-Al, Al_3Nb, NbB_2 and NbC phases.

Fig. 2 is the SEM images of Al_3Nb-NbB_2-NbC/Al inoculant ingot. Fig. 2a is the low magnification SEM image of the inoculant, where it can be seen that there are many particles distribute in the Al matrix and some of them exist with agglomeration. In order to further observe the distribution and morphology of these second phase particles, the high magnification SEM images are shown in Fig. 2b, c and d; Fig. 2e, f, and g are the corresponding EDS analysis of different particles. According to the XRD and EDS results, it can be confirmed that these particles are Al_3Nb, NbB_2 and NbC particles. The large irregular block particles are Al_3Nb and their sizes are about 7–8 μm; the NbB_2 and NbC particles are smaller than Al_3Nb, whose sizes are about 1–3 μm. In Fig. 2c, it also can be noticed that the NbB_2 particles are prone to agglomerate, however, when there NbC particles around them (Fig. 2b), this tendency decrease.

In Fig. 2, it can be seen that the second phase particles especially for the NbB_2 particles agglomerate seriously, thus the inoculant ingot was prepared to ribbons by vacuum rapidly quenched treatment. In order to explain the difference between the inoculant ingots and ribbons in microstructures, the SEM images of them are shown Fig. 3. Compared with Fig. 3a, in Fig. 3b, the NbB_2 aggregations are broken up and most of the particles are decreased to 200 ~ 500 nm, though there are also some large particles. The reasons for the above changes are speculated as follows: first, the rapid cooling make the particles have not a chance to grow up during the process of solidification; second, the big and clustered ceramic particles are mechanically broken in the process of vacuum rapid quenching treatment. Therefore, compared with the inoculant ingots, the number density of the nucleation particles in the inoculant ribbons are increased and when the inoculant ribbons are added to the Al-Cu-Mn alloy melt, the nucleation particles can distribute in the melt more uniformly, resulting in the improvement of inoculant effect.