NODULARIZATION OF α-AlFeSi COMPOUNDS IN PURE ALUMINUM

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ABSTRACT The aim of this research is to establish the mechanism of nodularization of α-AlFeSi constituents based on the hypothesis that this phase particles form in hydrogen bubbles during the solidification. In order to prove this hypothesis, the following experiments were done: 1) the observation of structure of α-AlFeSi nodules, 2) the effect of hydrogen bubbles on the formation of α-AlFeSi nodules. As a result, it became clear that thc morphology of α-AlFeSi nodules, which forms during solidification is a spherical shape with a hollow shell. Therefore this structure indicates that α-AlFeSi particles nucleates on the inner wall of the hydrogen bubbles which evolve during solidification. Furthermore , the size and density of α-AlFeSi nodules changed with the cooling rate of solidification and their variations agreed with those of the hydrogen bubbles. Thus, this hypothesis that α-AlFeSi nodules form in hydrogen bubbles was proved.

Keywords: *α-AlFeSi, nodularization, pure aluminum, hydrogen bubble, porosity*

1. INTRODUCTION

Most commercial aluminum alloys contain primary constituents, which form during solidification due to impurities. The most important impurity in this regard is iron, which combines with aluminum and silicon to form intermetallic compounds during solidification. The Al-Fe phases which appear in pure aluminum are Al_3Fe, Al_6Fe and α-AlFeSi[1] and so on. The structure and composition of these phases have already been studied[2]-[5]. But, there are only few studies about the morphology of these phases. It is known that α-AlFeSi compounds in pure aluminum are spherical, but it has not been clear why α-AlFeSi compounds have spherical shapes.

Nodular graphite in cast iron is known to have a spherical shape, and many studies about the origin of nodular graphite in cast iron[6] have been reported. Many hypotheses suggested about nodular graphite cannot reasonably explain the actual phenomena. On the other hand, the gas-bubble theory[7], which suggested that graphite precipitates inside gas bubbles in molten cast iron and then the gas bubbles are filled with the graphite, can reasonably explain the spherical shape of nodular graphite.

It is well known that hydrogen causes porosities in pure aluminum[8]. Hydrogen exist as interstitial atoms in solid aluminum. But its solubility in the solid state is very small and about one twentieth of that in the liquid state. So porosities evolve during solidification resulting from the decrease of solubility. Based on the gas bubble theory, the reason why α-AlFeSi nodules with a hollow shell form can be explained as porosities, namely, hydrogen bubbles are not filled all over with α-AlFeSi particles. When the size and density of porosities change, it is expected that those of α-AlFeSi nodules follow the changes of porosities.

The aim of this research is to establish the mechanism of nodularization of α-AlFeSi constituents on the hypothesis that this phase particles form in hydrogen bubbles. In order to prove this hypothesis, the following experiments were done : 1) the observation of the structure of α-AlFeSi nodules, 2) the effect of hydrogen bubbles on the formation of α-AlFeSi compounds.

2. EXPERIMENTAL PROCEDURE

Commercial purity aluminum containg 0.13%Si, 0.13%Fe as impurities were used in this work. The pure aluminum was melted in a carbon crucible at 800℃, and solidified in the mold made of copper with water cooling pipes illustrated in Fig.1. In this mold, the cooling rates during solidification can be controlled by changing the thickness of the spacer. Cast slabs were divided in two parts perpendicularly to the thickness; one part was used for the observation of microstructures after extracting particles by phenol method, the other was for the measurement of hydrogen content in the slabs. Specimens etched by 0.5% HF were used for the observation of the microstructures, and an ultrahigh vacuum method was used for the determination of the hydrogen contents.

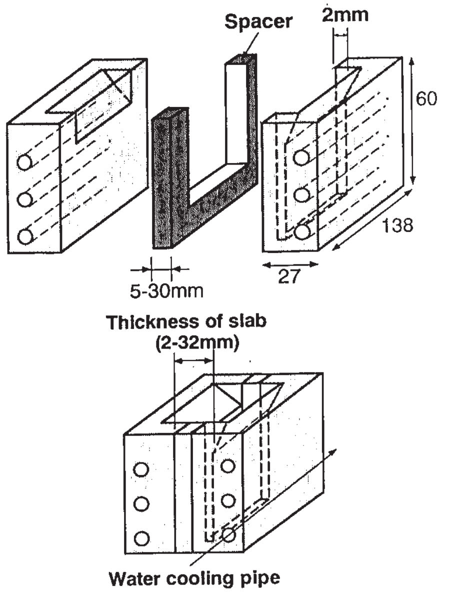

Fig.1 Dimmensions of the mold and spacer made of copper. Cooling rate was controlled by changing the thickness of the spacer.

2.1 Structure of α-AlFeSi nodules

Specimens cut from slabs were solved by phenol method, and extracted α-AlFeSi nodules on filters were observed by SEM. Further microtomed specimens were prepared for TEM in order to observe the cross-section of α-AlFeSi nodules.

2.2 Effect of hydrogen bubbles on the formation α-AlFeSi nodules

Two methods were employed to examine the effect of hydrogen on the formation of α-AlFeSi nodules.

(1) The changing cooling rates during solidification

Aluminum melted at high temperature can contain a large amount of hydrogen. During solidification, hydrogen is liberated according to the decrease of solubility and gas bubbles evolve in the molten aluminum. These bubbles float up and go out from the melt surface. But, in fast cooling rate during solidification, it is expected that the bubbles are captured in the melt and then the porosities increase. Therefore, the cooling rates was controlled by changing the thickness of the spacer illustrated in Fig.1. It is known that the relation between cooling rate(C) and dendrite arm spacing(D) can be indicated by Eq. 1[9]. In this equation, α and A are constant. In order to estimate the cooling rates, the data of α=0.33

and A=33.4[10] were substituted into Eq.1.

$$D \times C^{\alpha} = A \qquad (1)$$

The α-AlFeSi nodules extracted from slabs were observed by SEM, and their diameter was measured by SEM. Furthermore, in order to examine the size and density of the hydrogen bubbles by cooling rates during solidification, high purity aluminum (99.99%) containing a large amount of hydrogen was used. Cooling rates were controlled by changing the thickness of the spacer, and the center of the slabs were examined under optical microscopy (OM) to estimate the average diameter of the hydrogen porosities.

(2) Increasing hydrogen contents in the molten aluminum

It is expected that the molten aluminum containing a large amount of hydrogen tends to evolve many hydrogen bubbles during solidification. In order to increase the content of hydrogen in the molten aluminum, potatoes were added into the melt. Hydrogen entered into the molten aluminum by the reaction of aluminum with water in potatoes. This molten aluminum was cast in the mold with 30mm spacer. The estimation of porosities was carried out in the same method previously described.

3. RESULTS AND DISCUSSION

3.1 Structure of α-AlFeSi nodules

Figure 2 shows the SEM micrograph of the compounds extracted by phenol method from a pure aluminum(Al-0.13%Si-0.45%Fe). It is obvious that Al-Fe compounds are plate-like, and the α-AlFeSi nodules were spherical. Figure 3 shows the microtomed section of α-AlFeSi nodules. It is clear that α-AlFeSi nodules are spherical shape with a hollow shell, and the outer shell consists of many particles. The gas bubble theory suggested in cast iron can explain this structure reasonably. The schematic model of the formation of α-AlFeSi nodules is shown in Fig.4. The α-AlFeSi particles which are single crystals, nucleate into the hydrogen gas bubbles and grow along the inner wall of hydrogen bubbles. Then these particles coagulate as polycrystals and form spherical nodules with a hollow shell.

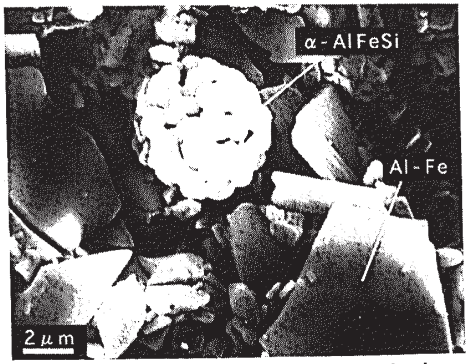

Fig.2 SEM micrographs of spherical α-AlFeSi nodule and plate-like Al-Fe compounds extracted by the phenol method from a slab.

3.2 Effect of the cooling rate during solidification on the formation of α-AlFeSi nodules and hydrogen bubbles.

Figure 5 shows the microstructures of as-cast slabs and the SEM images of extracted α-AlFeSi nodules from them which were cast by various cooling rates during solidification. The density of α-AlFeSi increased shown in Fig.5(A)-(C), while their size decreased with increasing the cooling rates

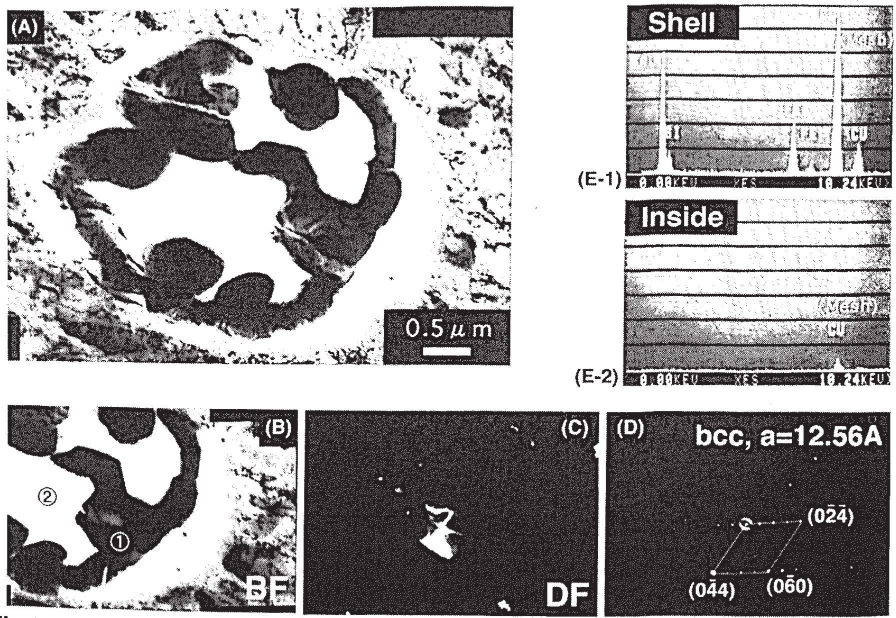

Fig.3 TEM micrographs of microtomed section of α-AlFeSi nodule. (A)and(B):microtomed section of α-AlFeSi (bright field images), (C):dark field image, (D):selected area diffraction pattern of B-①, (E-1):EDX spectrum from outside shell of α-AlFeSi (B-①), (E-2): EDX spectrum from inside shell of α-AlFeSi (B-②).

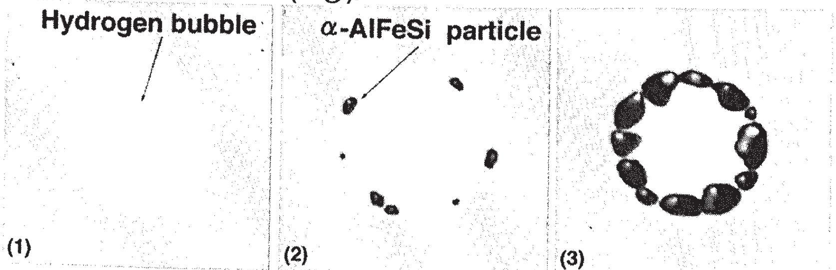

Fig.4 Schematic model for the formation of α-AlFeSi nodule. (1) evolution of hydrogen bubble during solidification. (2) nucleation of α-AlFeSi particles on the inner wall of hydrogen bubble. (3) bubble is gradually filled from the wall into the center with α-AlFeSi particles.

shown in Fig.5(D)-(F). Figure 6 shows hydrogen bubbles in high purity aluminum slabs cast by different cooling rates. The size of hydrogen bubbles decreased and their density increased with increasing the cooling rates. The size and density of bubbles in high purity aluminum are different from commercial purity aluminum because the evolution of hydrogen bubbles is affected by impurities or trace elements. In this work, two kinds of purity aluminum were used in order to examine the effect of cooling rates on α-AlFeSi nodules and hydrogen bubbles. Thus, the size and density of α-AlFeSi

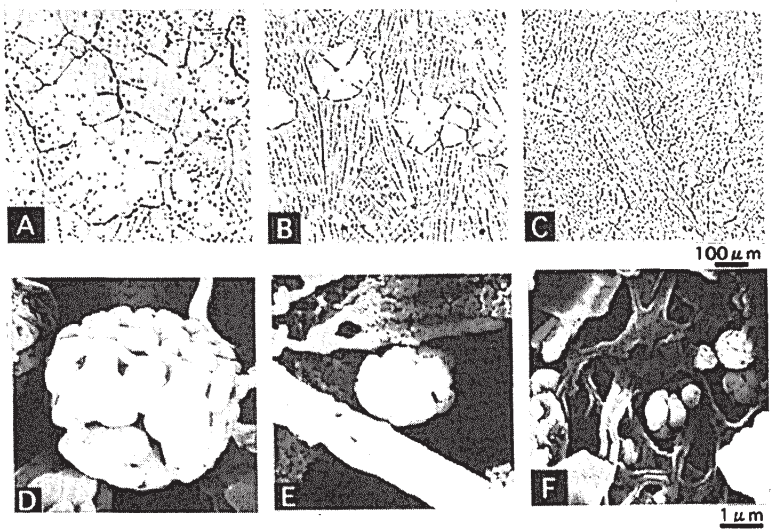

Fig.5 Effect of cooling rates during solidification on the DAS and the size and density of α-AlFeSi nodules. (A)-(C):OM, (D)-(F):SEM of α-AlFeSi nodules.
Cooling rates during solidification: (A)(D):0.1Ks^{-1}, (B)(E):0.6Ks^{-1}, (C)(F):34Ks^{-1}

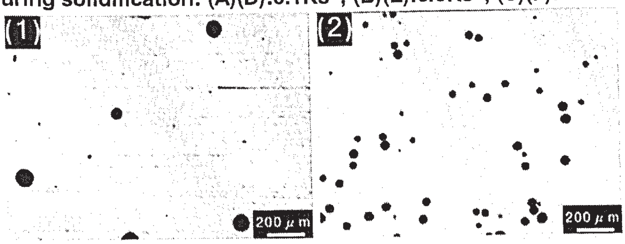

Fig.6 Effect of cooling rates during solidification on the size and density of porosities in 99.99% high pure aluminum. Cooling rates during solidification: (1):0.1Ks^{-1}, (2):34Ks^{-1}

nodules did not agreed fully with those of hydrogen bubbles. Conseqently it is concluded that the size and density of α-AlFeSi nodules change according to the variation of hydrogen bubbles in which α-AlFeSi particles nucleate.

3.3 Effect of solubility of hydrogen in the molten aluminum on α-AlFeSi formation.

The hydrogen content in slab produced by adding potatoes into the melt was 0.57cm^3/Al 100g, while it was 0.27cm^3/Al 100g in a slab without potatoes. Figure 7 shows α-AlFeSi extracted from slabs which have different contents of hydrogen. Figure 8 shows the microstructures of these slabs. It is obvious that the average size of α-AlFeSi nodules did not change, but the density of α-AlFeSi nodules increased with hydrogen contents in slabs. It is considered that the hydrogen bubbles increase with the hydrogen content in the molten aluminum, and as a result the density of α-AlFeSi nodules also increase.

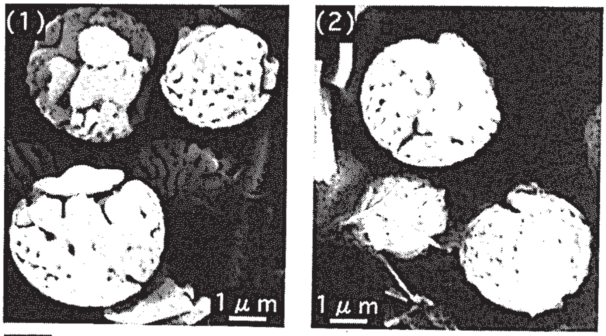

Fig.7 Scanning electron micrographs of typical α-AlFeSi nodules extracted from slabs by phenol method.
Content of hydrogen
(1): 0.57cm^3/Al100g
(2): 0.27cm^3/Al100g

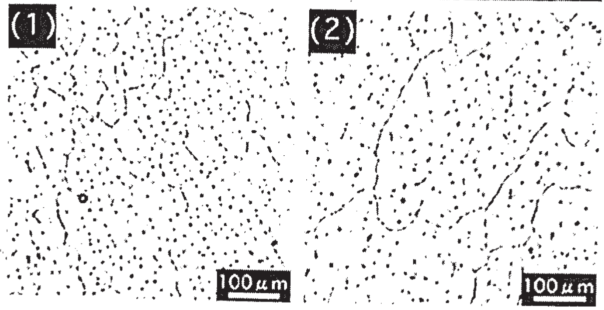

Fig.8 Effect of hydrogen content in slabs on the density of α-AlFeSi nodules.
Content of hydrogen
(1): 0.57cm^3/Al100g
(2): 0.27cm^3/Al100g

4. CONCLUSIONS

(1) α-AlFeSi compounds are spherical nodule with hollow shell, and the outer shell consists of coagurated α-AlFeSi particles.

(2) The variation in the size and density of α-AlFeSi nodules changed by the cooling rate during solidification agreed with those of hydrogen bubbles.

(3) The density of α-AlFeSi nodules and the hydrogen bubbles which evolved during solidification increased with the hydrogen content in the molten aluminum.

(4) For the above results, it is considered that α-AlFeSi nodules nucleate in hydrogen bubbles during solidification. This is the reason why α-AlFeSi nodules are spherical with a hollow shell.

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