Microstructural Evolution and Growth of Intermetallic Compounds at the Interface between Solid Cast Iron and Liquid Al–Si Alloy

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Abstract: Compound casting has received a great deal of attention as a useful method for fabricating a single complicated part from dissimilar metallic materials. For example, in the case of automobile cylinder blocks, research is being carried out to compound cast iron with aluminum alloys. In order to manufacture such as composite parts, it is essential to control the intermetallic compound formed at the interface between two metals. In this research, the type and growth behavior of compounds formed at the interface between cast iron and aluminum, or Al–Si alloy, were investigated. It was observed that the AlFe 5 phase was mainly formed at the interface between the pure aluminum melt and the solid cast iron, and the thickness of the interfacial compound increased proportionally with increasing contact time. On the other hand, more complex phases were formed at the interface between the Al–Si alloy melt and the solid cast iron. In the case of a specimen having a contact time of 10 min, Al 5 FeSi, AlFe 5 Si and AlFe 2 phases appeared to occupy the largest portion of the interfacial compound region. The total thickness of the interfacial compounds also increased in proportion to the contact time.

Keywords: compound casting; intermetallic compound; cast iron; Al–Si alloy

1. Introduction

As the demand for weight reduction continues to increase, especially in the automotive industry, efforts to replace previous iron-based parts using aluminum alloys are also on the rise. However, aluminum alloys have lower mechanical properties and heat resistance than iron-based alloys, in spite of their many advantages. Because the relatively low properties of aluminum alloys are limiting their expansion, much attention has been focused on a technique for manufacturing an iron-based alloy, and an aluminum alloy as a composite part, in order to overcome the low properties of the aluminum alloy. There are various methods of manufacturing composite parts, but among them, the compound casting method, which is capable of manufacturing a complicated shape part, and has excellent economical merit, has received a great deal of attention [1–3].

In the case of the cast joining of an iron-based alloy and aluminum alloy, a reaction occurs between a solid iron and a liquid Al alloy. First, the iron may be partly dissolved in an aluminum alloy melt, depending on the process conditions [4–6]. It is also expected that various intermetallic compounds will form and grow between Fe/Al interfaces [1–3,7–17]. It is known that the Al 5 Fe 2 and AlFe phases are mainly formed at the interface between the steel and the pure aluminum melt [7,12–17], while various AlFeSi phases, such as Al 5 FeSi and Al 5 FeSi, are known to be mainly formed at the interface between the steel and the Al–Si alloy melt [10,11,16].
Thus far, a lot of investigation has been conducted on the interfacial compounds formed between the steel and the molten aluminum or aluminum alloy. However, in the case of a cast iron/aluminum melt, since cast iron has a different composition and microstructure from steel, it may be different in the types of interfacial compounds.

In order to obtain sufficient bonding between dissimilar metallic materials to occur, it is necessary to form a compound at an interface, but when a large amount of compound is formed, deterioration in its ductility and an interference with heat transfer may occur. The formation conditions of such interfacial compounds are expected to change depending on the composition of Fe-based alloys and aluminum alloys, but the results of research on cast iron with Al-Si alloy, which is widely applied industrially, are insufficient.

This study was conducted as basic research for the development of the compound casting technology of cast iron and Al-Si alloy, which is widely used in the automotive industry. In addition, the main focus was to investigate the types and growth behaviors of the intermetallic compounds formed at the interface.

2. Materials and Methods

As shown in Figure 1, the cast iron insert (diameter of 5 mm, height of 10 mm, cylindrical shaped) was placed in the graphite mold with a diameter of 35 mm. Pure aluminum (99.9 wt.%, NeoDM, Daejeon, Korea) and commercial A356 alloy ingots were heated in a graphite crucible at a high frequency induction furnace, and then prepared molten metal of 720 °C was poured into the graphite mold. The mold was maintained at a temperature of 720 °C for a given time in a furnace, and then was naturally cooled in air. The chemical compositions of the cast iron and the A356 alloy are shown in Table 1. The observation and thickness measurement of the compounds of the cast iron/aluminum interfaces of the specimens were carried out using an optical microscope (OM) and a scanning electron microscope (FE-SEM, SU5000, Hitachi, Tokyo, Japan), equipped with energy dispersive X-ray spectrometer (EDS, JEOL, Tokyo, Japan). The metallographic specimens were polished and then etched using a 0.5% hydrofluoric acid solution in water. Backscatter diffraction (EBSD, TSL Hikari Super Program, EDAX, Mahwah, NJ, USA) analysis was also carried out to investigate the intermetallic compounds at the interface.

Table 1. Chemical compositions of gray cast iron (insert) and A356 casting alloy (wt.%).

| Alloy      | C   | Si  | Mg | Mn  | Fe  | Al  |
|------------|-----|-----|----|-----|-----|-----|
| Cast Iron  | 2.65| 0.31| -  | 0.15| Balanced | -  |
| A356       | -   | 6.96| 0.31| -   | -   | Balanced |

![Table 1. Chemical compositions of gray cast iron (insert) and A356 casting alloy (wt.%).](image)

Figure 1. Schematic diagram of cast iron/liquid aluminum interaction experiment.
3. Results and Discussion

3.1. Cast Iron/Pure Aluminum Interface

Figure 2 shows SEM micrographs of the interface between the cast iron insert and pure aluminum melt formed during isothermal heating with different holding time. Apparently, an AlFe compound was formed at the interface, and its thickness was increased with increased holding time. It is noteworthy that even when the holding time was 0 min, an interfacial compound with significant thickness (about 30 µm) was formed. This is partly due to the fact that the cooling rate of the aluminum melt was not fast, because the melt was poured into the crucible preheated to 720 °C. Although the EDS analysis is not quantitatively accurate, it seems reasonable to judge that it is one of the two AlFe compounds, Al$_5$Fe$_2$ and Al$_3$Fe, from the analysis results in Table 2. According to the literature [1,7,15,16], most interfacial compounds are likely to be of the Al$_5$Fe$_2$ phase.

It is also interesting to note that the iron dissolution occurred clearly from the cast iron insert, and the iron content in the aluminum matrix near the interface was observed to increase even when the hold time was short (Table 2). In Figures 2 and 3, it is also observed that the needle-shaped phases are formed at the aluminum matrix slightly away from the interface. A closer observation of the AlFe compounds was made, as shown in Figure 4. Although more detailed investigation is required, the iron fraction of the AlFe phase tended to be a little increased as the holding time increased. These needle-shaped compounds have been reported to form during the cooling of the aluminum melt with high Fe content, and the phase is believed to be Al$_3$Fe [7].

![Figure 2. SEM-EDS analysis of our cast iron/liquid aluminum interface isothermally heated at 720 °C for different times: (a) 0 min, (b) 1 min, (c) 5 min, (d) 10 min (C refers to the interfacial compound region and A refers to the aluminum base region near the interface in Table 2).](Image)
Table 2. Concentrations (at.%) of intermetallic compounds (C) at the interface of cast iron/liquid aluminum and aluminum matrix (A) near the interface with respect to the holding time (the numbers in parenthesis indicate weight percent).

| Location | Location |
|----------|----------|
|          | 0 min     | 1 min     | 5 min     | 10 min    |
|          | C         | A         | C         | A         | C         | A         |
| Fe       | 23.0      | 0.2       | 23.5      | 0.7       | 26.5      | 0.2       | 26.4      | 0.2       |
|          | (38.2)    | (0.4)     | (38.9)    | (1.4)     | (42.8)    | (0.4)     | (42.6)    | (0.3)     |
| Al       | 77.0      | 99.8      | 76.5      | 99.3      | 73.5      | 99.8      | 73.6      | 99.8      |
|          | (61.8)    | (99.6)    | (61.1)    | (98.6)    | (57.2)    | (99.6)    | (57.4)    | (99.7)    |

Figure 3. SEM micrographs and EDS line profile from the cast iron/liquid aluminum interface isothermally heated at 720 °C for different times: (a) 1 min and (b) 5 min.

Figure 4. SEM-EDS analysis of Fe-containing phases formed a little away from the cast iron/liquid aluminum interface isothermally heated at 720 °C for different times: (a) 1 min and (b) 5 min.

The thickness of the compound formed and grown at the solid iron and liquid aluminum interface depends upon the diffusion rate of the elements and the dissolution rate of the growing compound into liquid aluminum [4–6,16]. The rate of growth of the interlayer thickness \( \frac{dx}{dt} \) that involves some melting at the interface can be expressed as in Equation (1) [4]. If the melt convection is not large, the dissolution rate \( k \) in Equation (2) becomes a very small value, because the diffusion layer thickness \( \delta \) is much larger than the diffusion coefficient \( D \). As a result, if variables other than time are fixed, the increase in the thickness of the compound depends only on the holding time \( t \), in which case, the diffusion rate controls the thickness increase rate, as shown in Equation (3).

\[
\frac{dx}{dt} = \frac{k_1}{x} \left( \frac{C \cdot k}{\rho \cdot \eta} \right) \exp \left( -\frac{kSt}{v} \right)
\] (1)
\[ k = \frac{D}{\delta} \]  
\[ x \propto \sqrt{t} \]  

where \( k \) is the interlayer growth-rate constant, \( C_s \) is the saturation concentration, \( k \) is the dissolution rate constant, \( \rho_{int} \) is the density of an intermetallic compound, \( \phi \) is the content of the more refractory metal in an intermetallic compound, \( s \) is the solid surface area, \( v \) is the melt volume, \( t \) is the holding time, \( D \) is the diffusion coefficient of the dissolved metal, and \( \delta \) is the diffusion layer thickness [4–6,16].

Figure 5 shows that the thickness of the interfacial compound is generally proportional to the holding time. In the specimen with a holding time of 0 min, the interfacial compound was mainly formed during cooling. Therefore, the trend of increasing the thickness of the interfacial compound, except for the case of 0 min of holding time, is shown in Figure 5b, and it can be clearly seen that the thickness of the compound is proportional to the root of the holding time. Therefore, the theoretical parabolic relationship between thickness and holding time seems to be consistent with the experimental results, and it is concluded that the growth of the compound is determined by the diffusion rate under these experimental conditions.

**Figure 5.** Thickness changes of compounds formed at the cast iron/liquid aluminum interface with heating time at 720 °C: the specimen with a holding time of 0 min is included (a) and not included (b).

### 3.2. Cast Iron/Al–Si Alloy Interface

The reaction experiment between the cast iron insert and the Al–Si alloy melt was carried out in the same manner as the experiment between the cast iron and the pure aluminum melt. Figure 6 shows the difference of interfacial microstructures with the change of holding time. It is clearly observed that an interfacial compound was formed even at a short holding time, and the thickness of the compound was slightly increased as the holding time was increased. However, it can be seen from Figure 7 that there is a clear difference in the growth rate of the thickness of the interfacial compound. In the study of the formation of interfacial compounds due to the reaction of steel and Al, it has been reported that Si addition reduces the compound thickness increase [15,17]. Figure 7 also indicates that the total thickness of the interfacial compound is not accurately proportional to \( \sqrt{t} \), and this may be related to the presence of two or more compounds at the interface.

In the case of the interfacial compound, the results of EDS analysis in Table 3 indicate that Si is included in addition to Fe and Al. Therefore, this compound is considered to be different from the Al–Fe phase, and it is believed to be close to the Al\(_{13}\)FeSi (Al\(_{13}\)FeSi) or Al\(_{13}\)FeSi phase when compared with the results in the literature [3,17]. In SEM micrographs in Figure 6, the interfacial compound appears to consist of one phase, but two or more phases may be present in combination. The EBSD analysis result in Figures 8 and 9 led to the conclusion that the major interfacial compounds consist of Al\(_{13}\)FeSi and Al\(_{13}\)FeSi [12–18], depending on the holding time. In specimens with relatively short
holding times, Al₅Fe₂ and Al₃Fe₄ phases were not observed, and the interfacial compounds were mainly composed of Al₅Fe₂Si and Al₃Fe₄Si phases. On the other hand, a considerable amount of Al₅Fe₂ and some Al₃Fe₄ phases were also formed in the specimen with a long holding time (10 min), as shown in Table 4. Therefore, it is concluded that Al₅Fe₂Si and Al₃Fe₄Si, are initially formed, but AlFe compounds, such as Al₅Fe₂ and Al₃Fe₄, require sufficient time to form.

Figure 6. SEM-EDS analysis of cast iron/liquid Al-Si alloy interface isothermally heated at 720 °C for different times: (a) 0 min, (b) 1 min, (c) 5 min, (d) 10 min (C refers to the interfacial compound region and A refers to the aluminum base region near the interface in Table 3).

Figure 7. Thickness changes of compounds formed at the cast iron/liquid Al (pure) and at the cast iron/Al-Si alloy (A356) interface with heating time at 720 °C.
Table 3. Concentrations (at.%) of intermetallic compounds (C) at the interface of cast iron/liquid Al–Si alloy and aluminum matrix (A) near the interface with respect to the holding time (numbers in parentheses indicate weight percent).

| Time   | 0 min | 1 min | 5 min | 10 min |
|--------|-------|-------|-------|--------|
| Location | C   | A     | C   | A     | C   | A     | C   | A     |
| Fe     | 15.6  |      | 16.1 |      | 17.5 |      | 15.2 |      |
|        | (27.4)|      | (28.3)|      | (30.3)|      | (26.8)|      |
| Al     | 67.1  | 97.8  | 67.7 | 92.3  | 68.6 | 98.1  | 66.8 | 98.6  |
|        | (57.2)| (97.7)| (57.3)| (92.1)| (57.4)| (98.1)| (57.1)| (98.6)|
| Si     | 17.1  | 1.8   | 16.0 | 7.2   | 13.7 | 1.5   | 17.8 | 0.9   |
|        | (15.1)| (1.9) | (14.1)| (7.5) | (12.0)| (1.6) | (15.8)| (1.0) |
| Mn     | 0.2   |      | 0.2  |      | 0.2  |      | 0.2  |      |
|        | (0.3) |      | (0.3)|      | (0.3)|      | (0.3)|      |
| Mg     | -     |      | -    |      | 0.4  |      | 0.4  |      |
|        | (0.4) |      | (0.4)|      | (0.4)|      | (0.4)|      |

Figure 8. EBSD phase distribution maps of compounds formed at the cast iron/liquid Al–Si alloy interface isothermally heated at 720 °C: (a) for 1 min, (b) 5 min.

Figure 9. EBSD phase distribution map of compounds formed at the cast iron/liquid Al–Si alloy interface isothermally heated at 720 °C for 10 min.
Table 4. Phase fraction (number) of the compound region at the interface of cast iron/liquid Al–Si alloy shown in Figure 9.

| Phase          | Al₄.₅FeSi | Al₈Fe₂Si | Al₁₅Fe₄ | Al₃Fe₂ | Al | Ferrite |
|----------------|-----------|----------|---------|--------|----|---------|
| Fraction       | 0.19      | 0.19     | 0.14    | 0.21   | 0.13| 0.14    |

As in the case of pure aluminum, an iron-containing compound is also formed in the Al–Si alloy region slightly away from the interface (Figure 10). Of course, the Al–Si alloy contained about 7% Si and a little Mg, so that the eutectic Si and Mg₂Si phases were also formed.

The needle-shaped iron-containing compound is postulated to be the Al₄.₅FeSi (Al₅Fe₂Si) phase, and it has been well known to be harmful on the mechanical properties of aluminum alloys [19,20]. This result is indicative of the fact that the mechanical properties of the aluminum alloy region of the Fe/Al compound cast component may be degraded.

Figure 10. SEM-EDS analysis of cast iron/liquid Al–Si alloy interface isothermally heated at 720 °C for different times: (a) 5 min, (b) 10 min.

4. Conclusions

The reaction between the cast iron insert and the aluminum melt formed a compound at the interface, which was observed to form a significant thickness in a relatively short holding time. The interfacial compound is assumed to be mainly within the Al₅Fe₂ phase, and its thickness increase rate is approximately proportional to the root of the holding time, so it seems to be mainly determined by the diffusion rate of the elements. Meanwhile, at the aluminum matrix, slightly away from the interface, a compound, which is regarded as the Al₃Fe phase, was formed in a needle shape.

Experiments using Al–Si alloy melt showed similar results to those using pure aluminum melt. That is, the thickness increase rate of the compound region tended to be proportional to the root of the holding time, but the growth rate was significantly lower than that of pure aluminum. In addition, due to the alloying element Si, the main interfacial compounds were observed to be the Al₄.₅FeSi, Al₈Fe₂Si and Al₅Fe₂ phases. It was also found that the type and the relative amount of the interfacial phases varied with the holding time. In the aluminum alloy matrix, a little away from the interface, the acicular Al₄.₅FeSi phase, along with the eutectic Si and Mg₂Si phases, were observed.

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