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酸素濃度が与える Fe-Cu 合金の相分離挙動への影響

Effect of oxygen concentration on phase separation behavior of Fe-Cu alloys

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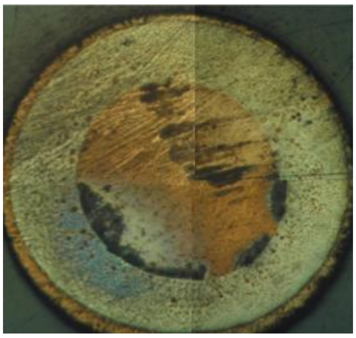


Abstract: Fe–Cu alloys, despite forming a peritectic system, exhibit a nearly flat liquidus line across a wide intermediate composition range. Upon rapid cooling, phase separation occurs, indicating a metastable liquid miscibility gap below the liquidus. However, conventional container-based processing introduces contamination and restricts supercooling due to heterogeneous nucleation. To overcome these limitations, this study employed a containerless gas-jet levitation method to melt and solidify Fe–Cu alloys (Fe–40 at%, 50 at%, 60 at% Cu) under low-oxygen environments.Microstructural analysis revealed that a Cu-rich phase consistently formed on the outer regions, and notably, Fe–50 at% Cu samples exhibited a distinct three-layer phase-separated structure. The highest occurrence of multiple-sphere morphologies (53.3%) and multiple latent heat release events (40.0%) was observed for the Fe–50 at% Cu composition. Cooling curve analysis showed both single and multiple latent heat release behaviors, attributed to differing solidification sequences of Fe-rich liquid droplets.These findings suggest that phase separation and multi-sphere structure formation are most pronounced near the equiatomic composition. Future studies will focus on the influence of oxygen concentration and other environmental factors to further understand the mechanisms driving these complex solidification behaviors.

1. Introduction

Fe–Cu alloys belong to a peritectic system; however, over a wide intermediate compositional range, they exhibit a liquidus line that remains nearly constant in temperature. When these alloys are rapidly cooled, phase separation is observed, suggesting the presence of a metastable solubility gap just below the liquidus line[1]. When melting and solidifying Fe–Cu alloys using a container, both Fe and Cu, as transition metals, are chemically active in the liquid state and readily react with the container material. As a result, compositional changes and property variations caused by contamination from container-derived impurities cannot be neglected. Furthermore, the container wall serves as a heterogeneous nucleation site, making it difficult to achieve large degrees of supercooling. To overcome these issues, the present study employed the gas-jet levitation method, a type of containerless melting technique. This approach enables non-contact holding and heating of the sample, thereby preventing container–sample reactions and contamination by impurities, while also allowing the realization of large supercoolings.

In this study, Fe–Cu alloys were melted and solidified using the gas-jet levitation method under environments with different oxygen concentrations. The purpose was to elucidate the liquid-phase separation phenomena in this system based on analysis of solidification microstructures and temperature data.

2. Experimental method

Alloy spheres of Fe–40 at% Cu (12 samples), Fe–50 at% Cu (15 samples), and Fe–60 at% Cu (12 samples), each 2 mm in diameter, were prepared by weighing high-purity Fe (99.5%) and Cu (99.9%) to the desired molar ratios. Sample preparation and experiments were carried out inside a glove box filled with Ar gas, with the oxygen concentration maintained at 30 ppm. Inside the glove box, a semiconductor laser, a radiation thermometer, a flow meter, and a sample stage were installed. The sample stage was made of boron nitride, which has a low coefficient of thermal expansion and excellent heat resistance. For levitating the samples, when the oxygen concentration was below 30 ppm, high-purity Ar gas was injected from below the sample stage toward the upper side of the sample. While levitated, the samples were melted by irradiating them with two semiconductor lasers. After melting, the laser irradiation was stopped, and the samples were solidified under rapid cooling while temperature data were recorded. The prepared alloy spheres were then polished, etched, and their internal microstructures were observed using an optical microscope. A schematic diagram of the experimental apparatus is shown in Figure. 1

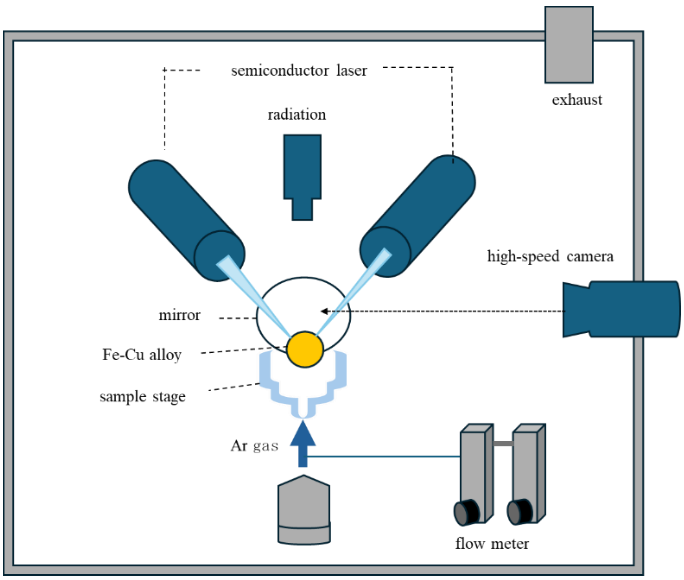
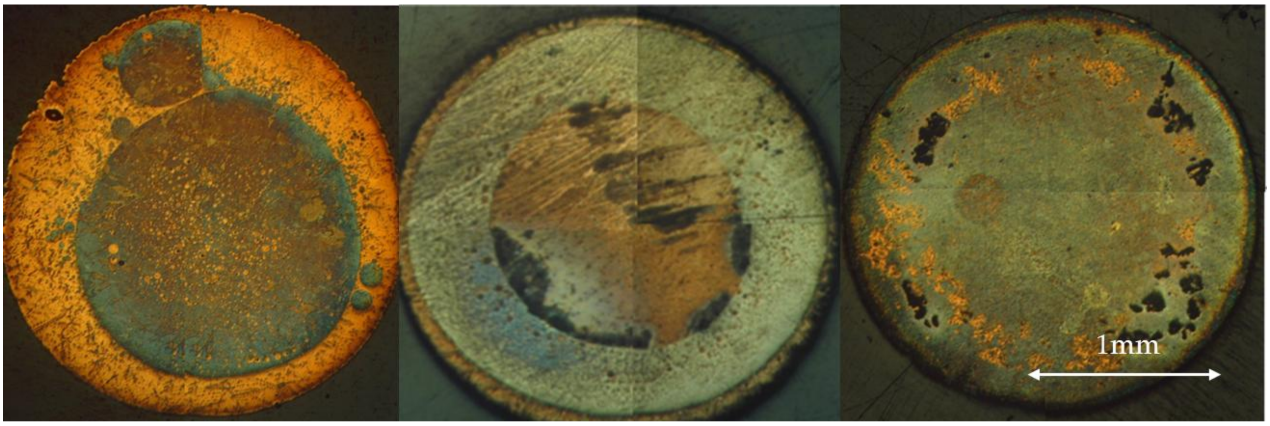


Figure1. A schematic diagram of the experimental apparatus

3. Experimental Results and Discussion

Figure 2 shows the cross-sectional microstructures of each composition captured using an optical microscope, and Figures 3 and 4 show the cooling curves of the obtained Fe–Cu alloys



Fe-40-at%Cu Fe-50-at%Cu Fe-60-at%Cu

Figure 2. Cross-sectional microstructures of Fe–Cu alloy spheres

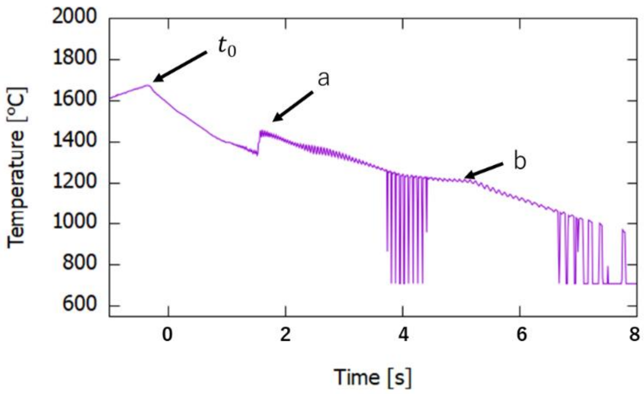


Figure 3. Cooling curves of Fe–Cu alloy spheres
(t_0: termination of laser irradiation,
a: solidification of the Fe-rich liquid phase,
b: solidification of the Cu-rich liquid phase)

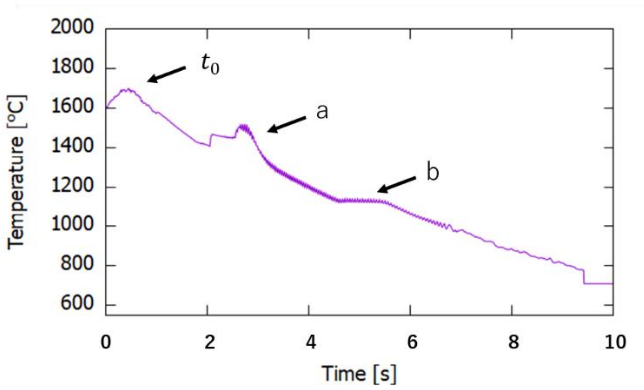


Figure 4. Cooling curves of Fe–Cu alloy spheres
(t_0: termination of laser irradiation,
a: solidification of the Fe-rich liquid phase,
b: solidification of the Cu-rich liquid phase)

Regarding the alloy microstructures, it was found that, for all compositions, a Cu-rich phase was distributed on the outer regions. In the case of Fe–50 at% Cu, a distinct three-layer phase-separated structure was observed, consisting of a thin outer Cu-rich phase, a broad Fe-rich phase in the intermediate region, and a circular Cu-rich phase in the innermost region. Among the fabricated samples, the proportion in which multiple-sphere structures were identified was 25.0% for Fe–60 at% Cu (3 out of 12 samples), 53.3% for Fe–50 at% Cu (8 out of 15 samples), and 33.3% for Fe–40 at% Cu (4 out of 12 samples).

With respect to the cooling curves, two distinct types were observed across all compositions, irrespective of the composition ratio: one exhibiting a single latent heat release during cooling (Figure 3) and the other exhibiting multiple latent heat release events (Figure 4).

The occurrence of multiple latent heat releases is considered to be due to differences in the solidification timing of multiple Fe-rich liquid droplets.

The proportion of samples exhibiting multiple latent heat release events varied with composition: 16.7% for Fe–60 at% Cu (2 out of 12 samples), 40.0% for Fe–50 at% Cu (6 out of 15 samples), and 16.7% for Fe–40 at% Cu (2 out of 12 samples). No correlation was found between latent heat release and the formation of multiple-sphere structures. Among the samples in which multiple-sphere structures were formed, the proportions that exhibited multiple latent heat release events were 25.0% (1 out of 4 samples), 50.0% (4 out of 8 samples), and 25.0% (1 out of 4 samples), respectively.

Considering these results, both the occurrence rate of samples exhibiting a triple-spherical structure and the proportion showing multiple latent heat release events during solidification were highest for Fe–50 at% Cu in the present experiments. This suggests that when the Fe and Cu contents are close to an equiatomic ratio, phase-separation phenomena and the formation of multiple-sphere structures become most pronounced.

In the future, we will involve conducting experiments under controlled environmental factors, such as oxygen concentration, to elucidate the mechanisms governing the formation of multi-phase-separated structures and multiple latent heat release behavior, their compositional dependence, and the role of oxygen in the development of the final microstructure.

References

1) T. Kobayashi and K. Nagayama: Journal of the Japan Institute of Metals, **81** (2017), 251page