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熱エネルギー貯蔵材料開発に向けた相分離合金の液相分離現象のその場観察と熱物性計測

In-situ observation of liquid-phase separation phenomenon and thermophysical property measurement of phase-separated alloys for the development of thermal energy storage materials

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1. Introduction

Thermal energy storage (TES) is a key technology for stabilizing the fluctuating supply of renewable energy sources [1-3]. Among TES methods, latent heat storage using high-temperature materials offers high energy density and a constant operation temperature. However, a major challenge is that such materials often exhibit high chemical reactivity at elevated temperatures, while those with large latent heat tend to have low thermal conductivity. Recently, miscibility gap alloys (MGAs) have been proposed as promising high-temperature latent heat storage media due to their ability to form encapsulated structures through liquid-liquid phase separation during solidification [4]. These structures can potentially enhance thermal stability and reduce degradation of container material. Despite these advantages, the phase separation dynamics and their relationship with thermophysical properties remain insufficiently understood. This study aims to elucidate the mechanism of liquid-phase separation in Fe–Cu alloys under containerless conditions and evaluate their heat of fusion, in order to optimize MGA microstructures for TES applications.

2. Experimental Procedure

Spherical samples of Fe–Cu alloys were levitated in an Ar–3%H_2 atmosphere using a closed-type aerodynamic levitator (ADL) to suppress oxidation. The sample was melted with a 100 W CO_2 laser and subsequently cooled at controlled rates. The solidification process was observed in situ using a high-speed camera operating at 1000 fps. The oxygen partial pressure in the chamber was monitored by a zirconia-type oxygen sensor to ensure minimal oxidation. After solidification, the microstructures were examined by optical microscopy and SEM–EDS to determine the morphology and composition of the phase domains.

3. Results and Discussion

In situ observations revealed that the Fe-rich liquid domains appeared first, followed by the formation of Cu-rich liquid droplets. The Fe-rich domains gradually coalesced and, at higher Fe concentrations, completely encapsulated the Cu-rich phase. This sequence is consistent with the calculated asymmetry in the Gibbs energy of mixing (ΔG_{mix}) for the Fe–Cu liquid, where the Fe-rich side exhibits a steeper slope, favoring its earlier nucleation.

The Fe75Cu25 alloy formed under normal gravity using the ADL exhibited a random dispersion of Cu-rich domains within the Fe-rich matrix. In contrast, the same composition processed under microgravity using the electrostatic levitation furnace aborded at the iss (ELF) showed a concentric distribution of Cu-rich domains. These morphological differences suggest that convective flow under normal gravity disturbs the concentration field governed by ΔG_{mix}, resulting in a disordered dispersion of the Cu-rich phase. This phenomenon will be further investigated through numerical simulations incorporating the thermophysical properties of molten Fe–Cu alloys obtained from the ELF experiments.

4. Conclusions

Containerless processing using aerodynamic levitation enabled in-situ observation of the liquid–liquid phase separation in Fe–Cu alloys, as well as accurate measurement of their heats of fusion. The sequence of phase separation, driven by the asymmetry of ΔG_{mix}, results in microstructures ranging from encapsulated to dispersed Cu-rich domains, depending on the cooling conditions. Future work will focus on controlling droplet mobility during solidification to tailor microstructures for improved thermal cycling stability in thermal energy storage applications.

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References

1. IRENA, Innovation outlook: Therml Energy Storage, International Renewable Energy Agency (2020).
2. T. Okazaki: Renew. Energ., 151 (2020) 563.
3. Odenthal, F. Klasing and T. Bauer: Energ. Proc., 135 (2017) 14.
4. A. Rawson, E. Kisi, H. Sugo and T. Fiedler: Int. J. Heat. Mas. Trans., 77 (2014) 395.