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Eutectic-to-metallic glass transition in the Al–Sm system

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Abstract

A systematic experimental study is carried out to investigate microstructure selection over a very wide range of undercooling at the interface in the Al–Sm system by using a combination of the Bridgman, laser and melt spinning techniques, which give increasing interface undercooling. Eutectic microstructure forms at low undercooling, while metallic glass forms at very large undercooling. Experiments are designed to obtain a sharp transition from the eutectic to glass during the growth of eutectic as the interface undercooling increases with growth. The eutectic spacing at the transition is characterized, and the results are analyzed by using a model of eutectic growth that incorporates non-equilibrium effects at the interface. It is shown that a very large undercooling at the interface, required for glass formation, is obtained due to the combined effects of the sharp decrease in the diffusion coefficient, or the sharp increase in viscosity of the liquid, with undercooling in this system and the large undercooling required for the attachment kinetics at the compound–liquid interface in the eutectic structure.
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Keywords: Glass transition; Eutectic solidification; Metallic glass; Dendritic solidification; Non-equilibrium

1. Introduction

Microstructure selection during solidification under far-from-equilibrium conditions has been very important in the design of very fine-scale microstructures that play a key role in materials performance. Of specific importance is the selection of different stable and metastable phases, including the formation of metallic glass, with increasing departure from equilibrium conditions. Thus, the understanding of the fundamental physics of transformation pathways by which different microstructures evolve as a function of increasing departure from equilibrium has been the focus of research in recent years. The main emphasis of this work is to systematically investigate phase and microstructure selection in a fragile glass-forming system that undergoes liquid-to-glass or crystalline-to-glass transitions with increasing undercooling.

Experimental studies have shown that deep eutectic systems with low solubility in both phases are often prime candidates for the formation of metallic glass under the large undercoolings that are generally present in melt spinning experiments [1–3]. For example, melt spinning experiments in several Al–R systems (R = rare earth element) have shown the formation of metallic glass within a limited composition range in the hypereutectic region [1]. Since eutectic microstructures form in these systems at a lower undercooling, while the glass phase forms at a very large undercooling, it is important to investigate different microstructures that evolve between these two growth regimes so that one can establish the physics of transitions, including the transition to the glass phase. The aim of this work is to carry out a systematic study of the evolution of microstructure as a function of velocity or interface undercooling for different compositions in the Al–Sm system,

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which is a fragile glass-forming system. Previous investigations in this system have largely been focused on obtaining the amorphous phase under rapid solidification conditions in which very high cooling rates are used to suppress nucleation of the crystalline phase. To obtain an insight into the physics that leads to glass formation, we take a different approach in this study by designing experiments in which a direct transition from a eutectic to metallic glass phase occurs during the dynamics of growth process in which interface undercooling is increased systematically. In this case, a sharp interface is present between the eutectic and glass, and the conditions present at the interface can be characterized through the experimental measurements of eutectic spacing, interface temperature and the interface velocity at the transition. The physics of the transition is then evaluated by using the model of eutectic growth under rapid solidification conditions.

To systematically examine the selection of microstructures over a wide range of growth conditions, a combination of several experimental techniques has been used to obtain increasing departure from equilibrium. These techniques include the Bridgman technique of directional solidification, the laser scanning technique [4–6] and the melt spinning technique. Both the Bridgman growth and the laser melting techniques result in directional solidification, while in melt spinning the growth occurs from the undercooled melt. In the laser technique, the interface undercooling increases with the distance from the bottom to the top of the solidified pool [7–9], so that a sharp transition interface will be observed when appropriate undercooling is reached. In melt spinning, the wheel speed can be adjusted to give a cooling rate that triggers nucleation of a single phase or eutectic. These nuclei subsequently grow in the liquid that is being increasingly undercooled so that the growth will stop when the undercooling at the interface reaches that required for the transition to glass.

Three sets of experimental measurements are carried out to obtain relevant information. (1) Eutectic spacing variations with interface velocity in three different compositions are measured over the entire range of velocities where a stable eutectic structure is observed under directional growth conditions. (2) Since it is difficult to measure interface temperatures at very high velocities, the experimental conditions for the transition from eutectic to Al dendrite are determined first in laser scanning experiments, and then analyzed by using the competitive growth model for directional solidification that predicts that the interface temperature at the eutectic interface is the same as that at the dendrite tip at the transition velocity. The tip temperature of the Al dendrite is calculated by using the rapid solidification model, which includes the non-equilibrium effects at the interface [10,11]. The eutectic interface temperature value obtained at the transition velocity is then used in the eutectic growth model [12–14] to evaluate the non-equilibrium effects present at the eutectic interface. (3) In melt spinning experiments, where a direct transition from eutectic to glass is observed, the eutectic spacing at the transition interface is measured along with the variation in eutectic spacing with the growth of the eutectic nodule prior to the transition. The eutectic growth model is then used to characterize the velocity and undercooling values at the transition interface, and the physics that lead to large undercooling at the interface is evaluated. It is shown that a large undercooling at the interface, required for the transition to glass, can be attributed to the strong coupling between the following two effects. (i) In fragile glass-forming systems, the viscosity of the melt increases sharply, so that the diffusion coefficient, D, decreases sharply, as the undercooling is increased [15]. For eutectic growth, the decrease in D causes the interface undercooling to increase since a larger undercooling will be required for the solute transport between the two phases [12,13]. (ii) The non-equilibrium effects at the interface, required to drive the atomic attachment process for the compound phase in the eutectic, requires undercooling that depends on (V/D). Thus, once the non-equilibrium effects become important, the interface temperature is reduced, leading to a lower value of D, which in turn further increases the undercooling at the interface. This interplay between the non-equilibrium undercooling and the diffusion coefficient variation with temperature leads to a sharp increase in the undercooling that can become sufficiently large to cause the transition to glass formation. The importance of this physics on the transition to glass formation is discussed.

2. Experimental

Al–Sm alloys with compositions of 11.0, 15.0, 25.0 and 38.0 wt.% Sm were selected for study in the present work. These alloys correspond to hypoeutectic, eutectic and hypereutectic compositions, as seen in the phase diagram in Fig. 1 [16]. The relevant system parameters are listed in Table 1. The master alloys were prepared by melting

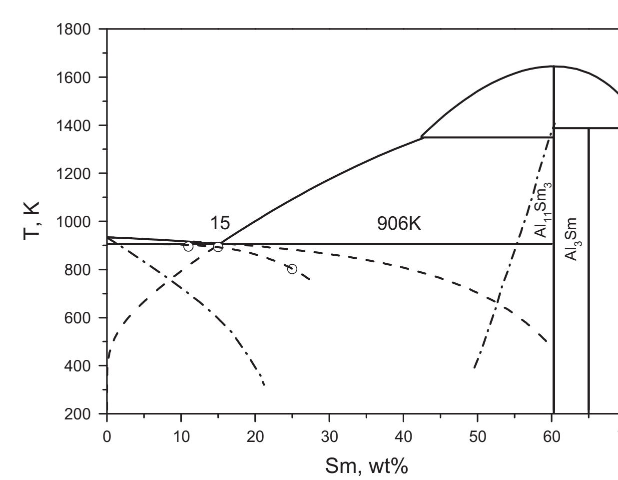

Fig. 1. Al-rich-side phase diagram of Al–Sm alloy. The liquidus line and their extensions are calculated by ThermoCalc by using the data in Ref. [16]. The data with circles represent the limit of eutectic growth or the coupled growth boundary. The T_o lines for the two phases are shown by the dot-dashed lines.