11th International Conference on Advanced Materials

Rio de Janeiro Brazil
September 20 - 25

ICAM2009

Crystallization behavior of a Zr-based bulk metallic glass at high heating rates

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Abstract – Laser processing is a powerful technique for the creation of metallic components with amorphous or uniquely tailored, highly non-equilibrium microstructures. Rapid, highly localized laser heating of a Zr-based bulk metallic glass results in the formation of an amorphous melt zone surrounded by a crystalline heat affected zone (HAZ). Because the high heating rate quickly bypasses the nucleation peak, the HAZ is composed of widely spaced, ~10 μm diameter spherulites formed from isolated nuclei. X-ray diffraction and microscopy comparisons with specimens prepared at lower heating rates reveal that the spherulites have a crystal structure distinct from the equilibrium crystalline phases.

The dimensions of as-cast bulk metallic glass (BMG) components are limited by the critical cooling rates necessary to produce fully amorphous structures, thus restricting the widespread use of many otherwise promising alloy compositions [1]. Laser additive manufacturing techniques present a unique opportunity to produce large-scale amorphous metallic components because of the inherently localized heat input and high cooling rates [2]. In order to take full advantage of this processing technique, the effects of the high heating rate on the structural evolution of the glass must be understood.

In the present study, we use the Laser Engineered Net Shaping (LENS^{TM}) process to deposit a Zr-based metallic glass forming powder on amorphous and crystalline substrates with the same nominal composition and examine the resulting structural changes in both the deposit and underlying substrate. An amorphous melt zone surrounded by a crystalline heat affected zone (HAZ) was observed during single-layer deposition on glassy substrates, as illustrated in Figure 1(a). The extent and morphology of the HAZ depend on the laser heat input, with very little crystallization observed for heat inputs less than ~7 J/mm^2. Under all processing conditions, the majority of the HAZ consists of ~10-20 μm diameter crystalline domains and/or spherulites, shown in Figure 1(b). TEM analysis indicates that the spherulites have an orthorhombic crystal structure and are distinct from the crystalline phases formed under equilibrium heating conditions. Finite element modeling (FEM) of the laser heating process indicates that crystallization is observed in regions where the peak temperature is within 200 K of the melting temperature (~1100 K) and the heating rate is on the order of 10^3 K/s. Rapid heating to this significant fraction of the melting temperature is expected to suppress crystal nucleation, such that growth dominates the crystallization process in the HAZ. The FEM further confirms that the rapid heating is followed by rapid cooling, with the temperature falling below the glass transition temperature within 0.5 s. This further limits the potential for nucleation of new crystals and suggests that crystallization begins during heating rather than cooling.

To further investigate the effect of heating rate on crystallization behavior, as-cast amorphous specimens were heated to just below their melting temperature at heating rates ranging from 0.5 to 50 K/s and immediately cooled at a constant cooling rate of 1.5 K/s. The resulting microstructures were investigated by x-ray diffraction and TEM. Bright field TEM images shown in Figure 2 reveal the formation of nanoscale crystals at lower heating rates and microscale spherulites at higher heating rates. Although they are much smaller in size, the spherulites are similar in structure to those observed in the HAZ. The reduction in the number and more extensive growth of crystals at the higher heating rate is consistent with the laser deposition observations. Further investigations of the crystallization kinetics are ongoing.

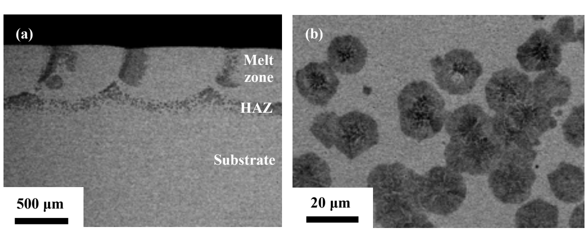

Figure 1: Cross-sectional SEM images of laser processed material. **a)** Amorphous melt zone surrounded by crystalline HAZ; and **b)** individual spherulites in the HAZ.

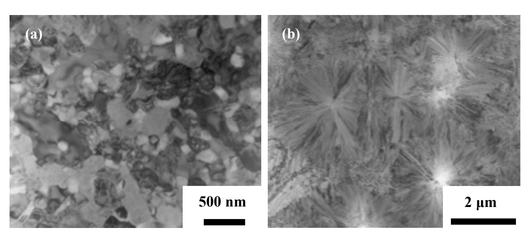

Figure 2: Bright field TEM images of the crystallized Zr-based BMG annealed at different heating rates. **a**) 0.5 K/s and **b)** 5 K/s.

References

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