Thermoplastic Forming and Related Studies of the Supercooled Liquid Region of Metallic Glasses

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Abstract

The thermoplastic formability (TPF) of metallic glasses was found to be related to the calorimetrically measured crystallization temperature minus the glass transition temperature, $T_g - T_x = \Delta T$. Alloy development in the ZrTiBe system identified a composition with $\Delta T = 120$ °C. Many alloys with $\Delta T > 150$ °C and one alloy, $Zr_{35}Ti_{30}Be_{27.5}Cu_{7.5}$, with $\Delta T = 165$ °C were discovered by substituting Be with small amounts of fourth alloying elements. The viscosity as a function of temperature, η(T), and time temperature transformation (TTT) measurements for the new alloy are presented and combined to create ηTT plots (viscosity time transformation) that are useful in determining what viscosities are available for a required processing time. ηTT plots are created for many alloys used in TPF in the literature and it is found that for processes requiring 60 - 300 s, $Zr_{35}Ti_{30}Be_{27.5}Cu_{7.5}$ provides an order of magnitude lower viscosity for processing than the other metallic glasses. Injection molding is demonstrated with $Zr_{35}Ti_{30}Be_{27.5}Cu_{7.5}$ and the part shows improved mechanical properties over die cast specimens of the same geometry. Changes of slope in η(T) measurements were observed and investigated in some quaternary compositions and found to be present in ternary compositions as well. Traditionally metallic glasses show a single discontinuity in heat capacity at the glass transition temperature. Alloys with the changes in slope of η(T) were found to show two discontinuities in heat capacity with the changes in slope of η(T) roughly correlating with the observed T_g values. These two T_g values were assumed to arise from two glassy phases present in the alloy. Further heat capacity analysis found systematic trends in the magnitude of the heat capacity discontinuities with composition and the single phase compositions of a metastable miscibility gap were discovered.

Microscopic evidence of the two phases is lacking so we must limit our claims to evidence of two relaxation phenomena existing and can't definitively claim two phases.

The alloy development led to the discovery of alloys with densities near Ti that are among the highest strength to weight ratio materials known. Alloys with corrosion resistances in simulated sea water 10x greater than other Zr based glasses and commonly used marine metals were discovered. Glasses spanning 6 orders of magnitude in corrosion resistance to 37% w/w HCl were discovered. Corrosion fatigue in saline environments remains a problem for these compositions and prevents their utility as biomaterials despite good evidence of biocompatibility in *in vitro* and *in vivo* studies.

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