## Abstract

Metallic glasses (MGs) are energetically metastable and can posses a series of lower energy state depending on the state of pressure, temperature and stress. The nature of metastability deviates MGs from its ideal state, as the lowest possible energy state of MGs refers to the ideal glass, and naturally prepared MGs refers to the true glass. Each state of MGs is linked with the specific glass transition temperature  $(T_g)$ . In the thesis, the  $T_g$  and super-cooled liquid region ( $\Delta T_x$ ) of Zr<sub>55</sub>Cu<sub>30</sub>Ni<sub>5</sub>Al<sub>10</sub>MG ribbon and bulk glassy plate have been measured using conventional and step-scan modulated temperature differential scanning calorimeter (MTDSC). A wide variation of  $T_{\rm g}$ (±14.8 K) and  $\Delta T_x$  (±18.5 K) have been estimated using conventional DSC at a heating rate of 5-400 K/min in the present study as well the data reported in the literature. It has been shown that the  $T_g$  can be precisely (±1.5 K) measured using step-scan MTDSC from the reversible heat flow curve, which excludes the structural relaxation near to glass transition. Similarly, the onset of the glass transition  $(T_g^{onset})$ and crystallization  $(T_x)$  temperatures of Zr<sub>47.5</sub>Cu<sub>47.5</sub>Al<sub>5</sub> and Zr<sub>41.2</sub>Ti<sub>13.8</sub>Cu<sub>12.5</sub>Ni<sub>10</sub>Be<sub>22.5</sub> (Vitreloy-1) bulk metallic glasses (BMGs) were investigated using conventional differential scanning calorimeter (DSC) and step-scan MTDSC. A large scatter of the  $T_g^{onset}$  values with error of  $\pm 6.9$  K for  $Zr_{47.5}Cu_{47.5}Al_5$  and  $\pm 8.0$  K for Vitreloy-1 have been observed, at a heating rate of 20 K/min using conventional DSC. Moreover, the tuning of the  $T_g$  in Zr<sub>55</sub>Cu<sub>30</sub>Ni<sub>5</sub>Al<sub>10</sub> MG by rolling at room temperature has been investigated. The precise thermo-analytical measurements using step-scan MTDSC revealed a decrease of  $T_g$  by 16.9 K in ribbons and 7.1 K in bulk glassy plates upon 40% and 70% thickness reduction, respectively. The cold rolled plates exhibit higher yield strength up to  $\sigma_y = 1.66$  GPa and larger plastic strain ( $\varepsilon_p = 8.0\%$ ) before failure than that of as-cast plates ( $\sigma_v = 1.45$  GPa). Transmission electron microscopy and DSC studies suggest that a relaxed structure has evolved due to the deformation induced structural change upon rolling, which reduces the activation energy of the shear transformation zone and improves the inherent plasticity of the glassy phase. Furthermore, A large number of metallic glasses were rolled at room temperature to introduce configurational changes and the variation of  $T_{g}$ ,  $\Delta C_{p}$  at  $T_{g}$ , configurational entropy ( $\Delta S_{\text{conf.}}$ ), pressure coefficient of  $T_g$  ( $dT_g/dP$ ), and bending strength were measured to establish the configuration-dependent pressure coefficient. The results

show the pressure-configuration interplay in MGs/BMGs and a new avenue to tune  $T_g$  by rolling at room temperature, and to classify in terms of  $\Delta C_p$  and  $dT_g/dP$ .