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Description of millisecond Ohmic heating and forming of metallic glasses

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Abstract

A quantitative description of the millisecond capacitive discharge heating and forming process built on a finite-element simulation platform is introduced. The platform incorporates thermodynamic and rheological models that extend beyond the supercooled liquid regime accessible by conventional calorimetry and rheology, accessing the regime that has just recently been uncovered via millisecond Ohmic heating. For the first time, a description of the dynamic glass transition is introduced and incorporated into the platform. The platform accurately simulates the process evolution and the thermodynamic and rheological response of the metallic glass, providing excellent agreement with experiments. Features such as the rapid temperature response, a dynamic glass-transition accommodated by a broad enthalpy recovery, a remarkable temperature and deformational uniformity, and an enthalpy trend that validates the adiabatic constraint, are accurately simulated. The platform is considered a useful tool for modeling the dynamic response and process evolution of metallic glasses under rapid uniform heating.

Keywords: Amorphous metals; Thermoplastic forming; Rheology; Calorimetry; Capacitive discharge

1. Introduction

The discovery of the metallic glass in 1960 [1] led to the broadening of the field of glass physics by introducing a "vitrified" metallic material with thermodynamic and kinetic properties similar to previously known silicate and molecular glasses, but with fundamentally different mechanical, electronic and optical properties. For example, like silicate or molecular liquids, metallic glass-forming liquids exhibit a hyper-Arrhenius increase in viscosity upon cooling from the molten state, accompanied by a loss of configurational entropy and enthalpy and a rise in isoconfigurational rigidity, until a kinetically "frozen" state is attained below a glass-transition temperature where the metastable liquid vitrifies (falls out of configurational equilibrium). However, unlike silicate or molecular glasses, metallic glasses are electronically and optically "metallic" like ordinary metals, and exhibit certain mechanical properties (e.g. fracture toughness) that are closer to those of crystalline metals than silicate glasses.

Several of the metallic bulk-glass forming compositions developed over the last decade are capable of forming glasses with critical casting thickness up to several centimeters [2]. Their glass-forming ability is associated with critical cooling rates from the equilibrium melt as low as $1\ \mathrm{K\ s^{-1}}$ or less, and time–temperature-transformation (TTT) diagrams with crystal incubation times at the crystallization "nose" up to tens of seconds or more [3]. The relatively slow crystallization kinetics has enabled the study and characterization of thermodynamic and rheological properties of the metallic undercooled liquid state previously not possible with "marginal" metallic glass formers. While the crystallization kinetics of bulk metallic glasses

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are substantially slower than those of the early metallic glasses, they are not sluggish enough to enable access to the entire undercooled liquid region on typical experimental time scales. Owing to a relatively rapid crystallization on cooling and particularly on reheating, thermodynamic and rheological data cannot be gathered throughout the entire metastable undercooled liquid. Consequently, experimentally inaccessible temperature ranges in measuring crystallization kinetics, viscosity and specific heat capacity exist where crystallization is most rapid [4,5]. Silicate or other molecular glasses with slower crystallization kinetics have TTT diagrams with "nose times" of thousands or even millions of seconds rather than tens of seconds or less, enabling experimental access over the entire metastable undercooled liquid region. Attaining experimental access through the entire undercooled liquid region of the metallic glass is thus of great importance for extending the comprehensive fundamental understanding of the physical behavior of these materials.

A method of rapidly heating and shaping an amorphous metal using a rapid capacitor discharge has recently been introduced [6]. This method utilizes Joule heating to uniformly heat, rheologically soften and thermoplastically shape metallic glasses rapidly (typically with processing times in the millisecond range). The method utilizes the discharge of electrical energy (typically 100 J–100 kJ) stored in a capacitor to heat a sample of metallic glass uniformly and rapidly to a predetermined "process temperature" within the supercooled liquid region in a time scale of several milliseconds or less. This process proceeds from the observation that metallic glass, by virtue of its being a frozen liquid, has a relatively high electrical resistivity that depends weakly on temperature, which results in highly dissipative, efficient, spatially uniform and nearly adiabatic heating of the material. By uniformly heating a bulk metallic glass in milliseconds, the stability of the supercooled liquid against crystallization is dramatically extended. Such a process has provided experimental access to physical properties such as enthalpy and viscosity in the entire range of the metastable liquid, as this range is no longer limited by crystallization on such short time scales.

In the present study, a quantitative description of the rapid capacitive discharge process and a thorough analysis of the physical properties of the metallic glass over the inaccessible temperature regime are attempted. Specifically, a finite-element model is developed to describe the effective coupling between electrical energy dissipation, heat generation and fluid flow under the application of electrical discharge and deformational force. The heating and flow response of the metallic glass is simulated and contrasted to experimental data gathered under identical conditions. Thermodynamic and rheological models are presented that have been extended to apply beyond the temperature ranges associated with typical experiments into regimes typically inaccessible under conventional heating rates. Moreover, for the first time a model for the glass transition has been developed in order to simulate the dynamic glass transition under these ultra-high heating rates. When incorporated into a finite-element simulation, these models yield excellent agreement with the thermodynamic and rheological data gathered in the experiments of Johnson et al. [6]. The finite-element model introduced here can be thought of as a computational platform for implementing process and property simulation studies that extend outside the supercooled liquid regime accessible by conventional heating.

2. Modeling transport phenomena

The analytic model in the present study was built to conform closely to the experiment of Johnson et al. [6]. The samples used in those experiments were as-cast Vitreloy 1 ($Zr_{41.2}Ti_{13.8}Cu_{12.5}Ni_{10}Be_{22.5}$) rods with radius ~2 mm and length ~2 cm. The rods were placed between parallel copper electrodes, heated by capacitive discharge and deformed between two parallel ceramic discs with holes in the center. Various voltages and compression forces were used. Ceramic discs were used due to their very low thermal conductivity so that the heat dissipated into the environment is negligible and the adiabatic assumption is ensured. The central holes in the ceramic discs have also aided in alignment and uniformity of deformation. The sample's effective deforming length between the ceramic discs was ~8 mm. In the present model, a cylindrical domain 2 mm in radius and 8 mm in length effectively simulates the samples in the experiments of Johnson et al. [6].

Considering the cylindrical geometry of the sample, a two-dimensional model with axial symmetry is constructed using the COMSOL Multiphysics finite-element analysis software. A mesh with ~10,000 triangular elements was generated. The element size and growth rate were adjusted to optimal values, and the ratio between the element area minimum and maximum was adjusted to 0.002. The mesh elements at the boundaries were small enough to capture the steep gradients in strain rate, while the total element number still ensured the overall calculation load was manageable on a stand-alone computer. Since the sample deformed severely in the experiment, the arbitrary Lagrange–Euler method was adopted to generate a moving mesh. The model accommodated three component mechanisms coupled together: electrical conduction, heat generation and viscous deformation.

2.1. Electrical conduction

The circuit implemented in the experiment of Johnson et al. [6] was an effective RLC circuit with the time constant $\tau = L/R$ and damping factor $= (R/2)\sqrt{C/L}$, where R is the total resistance comprising the sample resistance R_s and system resistance R_0, L is the inductance and C the capacitance. The following values were reported [6]: $C \approx 0.264$ F, $L \approx 2.4$ μH, $R_s \approx 3$ mΩ and $R_0 \approx 2.85$ mΩ. Together, these values yield a damping factor $\zeta \approx 1$. Therefore, the circuit was near its critical damping point. This gives the fastest