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Molecular dynamics analysis of plastic deformation and mechanics of imprinted metallic glass films

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ABSTRACT

The effects of mold cavity geometry and imprinting temperature on imprinted amorphous ZrCu films are studied using molecular dynamics simulations based on the many-body embedded-atom potential. The effects are investigated in terms of shear strain, flow field, loading force, relationship between pattern filling height and mold displacement, and pattern elastic recovery. The simulation results show that imprinting should be conducted at high temperatures (slightly above the glass transition temperature), followed by unloading at 300 K. Imprinting at higher temperatures shortens the pattern filling time, decreases the required loading force, and lowers the shear strain concentration inside the films. Unloading at 300 K leads to a low pattern elastic recovery, which increases with increasing temperature. Imprinting using a groove pattern on the mold requires a larger loading force and less filling time compared to those required when using a tip-like pattern on the mold. For imprinting using a tip-like pattern on the mold at temperatures of 300–475 K, shear transformation zones form inside the films in the form of a ring surrounding the mold.

1. Introduction

Nanoimprinting lithography (NIL) [1,2] is a useful technique for fabricating various micro- and nanocomponents at low cost, high resolution, and high throughput. Patterns made using NIL have many attractive practical applications, such as in micro- and nanostructure integration systems [3], optical devices [4], biological devices [5], and micro- and nanomold fabrication [6]. In NIL, pattern transfer is a mechanical deformation process operated under suitable temperature and pressure. A mold with nanoscale patterns is directly replicated onto thin films through imprinting and release.

Metallic glasses (MGs), whose atomic arrangement is disordered, are promising mold materials [6] for NIL to replace silicon and quartz molds due to their high hardness and strength, large elasticity, excellent viscous flow formability in the supercooled liquid region, and sufficient wear resistance [7–10]. A typical service life of MG molds for imprinting polyethylene (PE) was estimated to be about 200 years [11]. However, the imprinting of MGs may be accompanied by crystallization, which induces embrittlement and degrades the mechanical properties of MGs [12].

The understanding of the plastic deformation and mechanics of MGs is essential for the design and fabrication of nanomolds and components. Molecular dynamics (MD) simulation is a powerful tool for studying materials and mechanics at the nanoscale. Atomic simulation avoids experimental noise and can be used to analyze atomic trajectories, thermodynamics, and mechanical properties. Zhu et al. [13] modeled deformed $Cu_{46}Zr_{54}$ MG films under NIL. They found that when the film thickness approaches the width of the pattern grooves, the mold filling time and amount of material used can be reduced. Wu [14] simulated the forming mechanism and mechanics of $Cu_{50}Zr_{25}Ti_{25}$ MGs and found that a shear transformation zone (STZ) forms at the substrate surface underneath the mold and that the STZ area increases with increasing imprinting depth. Great pattern transfer can be obtained when unloading is conducted at room temperature rather than at high temperatures.

The present work investigates the plastic deformation and mechanics of $Zr_{50}Cu_{50}$ MG films during the NIL process utilizing MD simulation. The effects of mold cavity geometry and imprinting temperature on imprinted $Zr_{50}Cu_{50}$ MG films are studied. The results are discussed in terms of shear strain, flow field, loading force, relationship between pattern filling height and mold displacement, and pattern elastic recovery.

2. Model and methodology

A $Zr_{50}Cu_{50}$ alloy block with dimensions of 30 nm (length) × 30 nm (width) × 30 nm (height) was arranged as a face-centered

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cubic (fcc) structure for MG preparation. Each fcc unit cell consisted of two Zr atoms and two Cu atoms. Periodic boundary conditions (PBCs) were applied in three dimensions and the isobaric-isothermal ensemble (NPT) was used in the simulation. The Nosé-Hoover thermostat was used to control the temperature and maintain the system pressure at zero [15]. The time step was set as 1 fs. The block was heated from 0 to 2650 K (above the melting temperature [16]) at a constant heating rate of 0.5 K/ps and then quenched to 300 K at a high cooling rate of 1 K/ps to obtain a $Zr_{50}Cu_{50}$ MG block.

The NIL physical model comprises a $Zr_{50}Cu_{50}$ MG film and a mold, as shown in Fig. 1. The ZrCu MG film has dimensions of 14.5 nm (length) × 5.4 nm (width) × 12.6 nm (height), corresponding to 112,564 atoms. The film was obtained by cutting the $Zr_{50}Cu_{50}$ MG block. The mold composing of silicon atoms was assumed to be an ideally rigid object to simplify the NIL analysis and focus on the film deformation. Two types of pattern (tip-like and groove patterns) on the mold were used for studying the effect of mold cavity geometry on the NIL process, as shown in Fig. 1 (a) and (b). The two types of pattern had the same dimensions, namely 6.2 nm (length) × 5.4 nm (width) × 4.5 nm (height).

Two kinds of atom were set in the film, namely Newtonian atoms and fixed atoms. The fixed atoms of the four layers at the film bottom were used to support the whole system. PBCs were applied in the X and Y directions and the constant-volume, constant-temperature ensemble (NVT) was employed in the simulation. The mold had a constant downward displacement of 3×10^{-5} nm per time step for imprinting, followed by a holding process for 50 ps. Finally, the mold was instantly unloaded to simulate an ideal unloading process. The potential function adopted in this work was a realistic many-body embedded-atom (EAM) potential [17], which is suitable for describing the interaction of Zr and Cu atoms. The interaction between the mold and the film was described by the Lennard-Jones Potential [13]. The EAM potential equation is as the following:

$$E_i = \frac{1}{2}\sum_j \phi(r_{ij}) + F(\rho_i) \quad (1)$$

where E_i is the well-defined atomic site energy of the ith atom, $\phi(r_{ij})$ is a pairwise potential, $r_{ij} = |x_i - x_j|$ is the distance between atoms i and j, $F(\rho_i)$ is the embedding energy, and $\rho_i = \sum_j f_i(r_{ij})$ is the electron cloud density at the site of atom i. The parameters of the EAM potential of Zr-Zr and Cu-Cu were developed by Mendelev et al. [17]. The parameters of the LJ potential of Si-Zr and Si-Cu are listed in Table 1 [13], where the parameters were calculated using the Lorenz-Berthelot mixing rule [18]. The parameters of the LJ potential of Si-Zr and Si-Cu describe the interaction between the Si mold and ZrCu MG film.

Fig. 2(a) and (b) shows the partial pair distribution ($g(r)$) and the variation of the average potential energy per atom versus temperature of the MG film. In the $g(r)$ curve, the first-neighbor bonds evaluated from the position of the first peak are about 2.8 Å and the second peak is splitting, which are consistent with the experimental and theoretical values [19,20], indicating that the EAM potential function for describing the MG films is appropriate. The intersection point in the potential energy-temperature curve indicates the glass transition temperature (T_g) region of the MG film (about 680 K), as shown in Fig. 2(b).

3. Results and discussion

3.1. Tip-like pattern on mold

Fig. 3 shows variation of the imprinting process using the tip-like pattern on the mold at various mold displacement (D) values at a temperature of 300 K. At D = 4.6 nm, the van der Waals (VDW) attractive force drives a few MG atoms at the film surface to adsorb onto the mold. As the mold begins to imprint onto the film at a D value of 5.8 nm, the shear strain values of the MG atoms (light blue atoms) underneath the mold increase. The number of atoms with high shear strain values and high shear strain magnitudes increases with increasing D value. With a further increase in D value (6.3–8.4 nm), the number of atoms with high shear

Table 1
Parameters of LJ potential of Si-Zr and Si-Cu.

Species	ε (eV)	σ (Å)
Si-Zr	0.1137	3.3789
Si-Cu	0.0846	3.0820

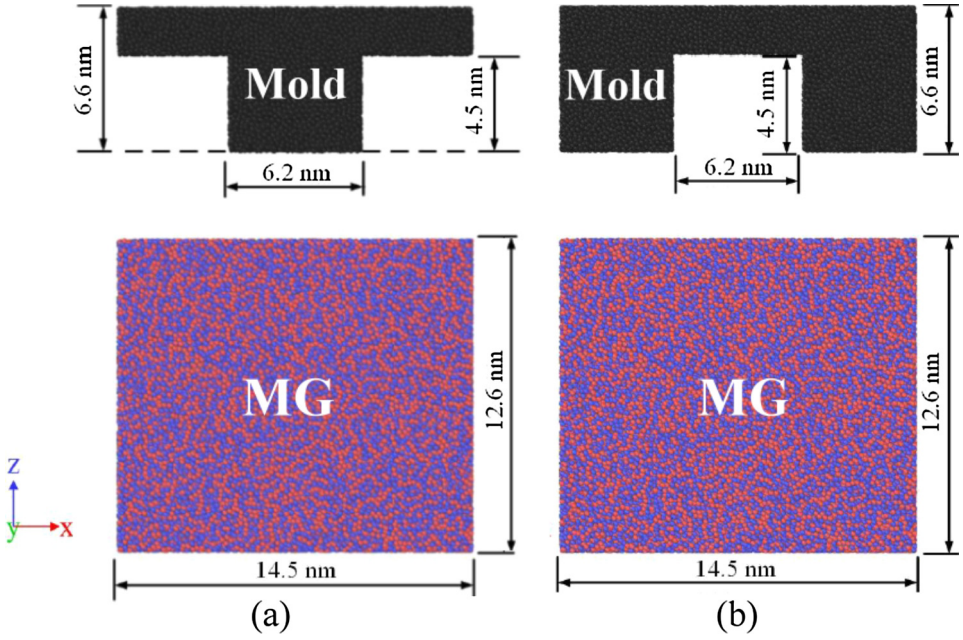

Fig. 1. Schematic of nanoimprinting physical model of $Zr_{50}Cu_{50}$ MG film (unit: nm). (a) Tip-like and (b) groove patterns on mold.