



Vitrification, crystallization, and atomic structure of deformed and quenched Ni₆₀Nb₄₀ metallic glass

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ABSTRACT

A Ni₆₀Nb₄₀ alloy was vitrified from fcc Ni and bcc Nb by cold rolling and by rapid solidification of an alloy ingot. The Ni₆₀Nb₄₀ that was vitrified by rolling has a lower crystallization onset temperature with single crystallization reaction compared to the two-stage crystallization at higher temperature for quenched Ni₆₀Nb₄₀. Fluctuation electron microscopy and hybrid reverse Monte Carlo modeling show that the as-rolled Ni₆₀Nb₄₀ contains fewer icosahedral-like Voronoi clusters and more crystal-like and bcc-like Voronoi clusters, which are inherited from the bcc Nb. The crystal-like and bcc-like medium range order clusters may be the structural origin for lower crystallization temperature in the as-rolled glass. It is demonstrated that hybrid reverse Monte Carlo simulations incorporating fluctuation microscopy and an empirical interatomic potential are insensitive to the starting structure.

1. Introduction

Bulk metallic glasses (BMGs) are typically synthesized by solidification from an undercooled liquid [1–5], but a variety of techniques have also been developed based on solid state reactions, such as mechanical alloying (e.g. ball milling, cold rolling) [6–11] and thermally induced multilayer interdiffusion [12,13]. Solid state reaction requires large negative heats of mixing between constituent components as the driving force. Then, to form an amorphous state, the input energy must be high enough to allow rapid and substantial interdiffusion of atoms, while avoiding nucleation of crystalline intermetallic compounds during the reaction time. Schwarz and Johnson [14] first reported the formation of metallic glass through solid-state interdiffusion in a multilayer structure of Au and La crystalline metals, which was followed by other alloys such as Mg₄₅Y₁₅Cu₄₀ [10], Ni₆₀Nb₄₀ [11], and Al₉₂Sm₈ [15].

Due to the non-equilibrium nature of metallic glass, the structures and properties are highly related to the processing history. For example, in amorphous Zr₅₅Al₁₀Cu₃₀Ni₅, a mechanically alloyed sample exhibits a higher crystallization temperature T_x but a narrower supercooled liquid region ΔT_x than a quenched glass of the same composition [6]. An amorphous Al₉₂Sm₈ alloy shows an even more dramatic difference [14]. The deformation induced Al₉₂Sm₈ amorphous alloy shows different crystallization behavior from melt-quenched samples. After annealing, the melt-quenched Al₉₂Sm₈ sample forms Al nanocrystals, but the as-deformed alloy is not susceptible to primary crystallization and

maintains an amorphous structure to significantly higher temperature and longer time. Fluctuation electron microscopy (FEM) experiments indicate that nanoscale Al-like medium range order (MRO) drives primary crystallization in the quenched glass, and that the deformed glass contains no Al-like MRO [15]. However, the structure of the deformed glass is not determined, leaving the structure associated with the glass state of the deformed alloy unclear.

Ni-based Nb containing amorphous alloys, which exhibit high fracture strength and high corrosion resistance [16,17], can be formed within a very wide composition range of 30–60 at% Nb by rapid solidification [18]. The Ni₆₀Nb₄₀ alloy is considered to be a good glass former in this binary alloy since it can be vitrified up to 2 mm thickness by quenching [19]. In addition to solidification, amorphous Ni₆₀Nb₄₀ can also be prepared by mechanical alloying (ball milling) from nickel and niobium powders [11]. However, during ball milling different powder particles experience different levels of deformation, so the inhomogeneity makes it difficult to relate the atomic structure to the thermodynamic properties.

Here, we report amorphous Ni₆₀Nb₄₀ fabricated by cold rolling and melt spinning. Both as-rolled and as-spun Ni₆₀Nb₄₀ alloy display an amorphous structure by X-ray and electron diffraction, but their crystallization behavior is quite different. The as-rolled Ni₆₀Nb₄₀ crystallized at a lower temperature and more crystalline phases were identified compared to as-spun sample after primary crystallization. The atomic structure was characterized by the combination of FEM and hybrid reverse Monte Carlo (HRMC) modeling and high-resolution

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transmission electron microscopy (HRTEM) for both alloys. HRTEM identified nanometer-scale residual crystallinity in the as-rolled sample. FEM plus HRMC demonstrated that even between the identifiable nanocrystals, more crystal-like and bcc-like MRO clusters, with 4- and 6-fold rotational symmetry and identifiable bcc local order, exist in as-rolled Ni₆₀Nb₄₀ compared to as-spun Ni₆₀Nb₄₀.

2. Experimental and computational methods

2.1. Sample preparation of Ni₆₀Nb₄₀

For as-spun Ni₆₀Nb₄₀ BMG, Ni and Nb of 99.9 at% purity or higher slugs were weighed to yield a composition of Ni₆₀Nb₄₀ that was arc-melted in a Ti-gettered, high-purity argon atmosphere to make a master ingot. The ingot was then remelted and injected onto a copper wheel spinning at 55 m/s tangential wheel speed to form 350 μm thick ribbons. For as-rolled samples, Ni and Nb foils of 99.8% purity with 25 μm in thickness were cut and weighed to reach Ni₆₀Nb₄₀ in the average composition. The Ni and Nb foils were stacked alternatively to form arrays as Ni/Nb/Ni/Nb... The cold rolling process was realized with a rolling mill using rollers 7.62 cm in diameter and 12.7 cm in length. After each rolling pass, the sample was compressed to half its thickness, and then folded before the next rolling pass. Every rolling pass took around 1 s and the number of rolling passes was used to represent the degree of deformation. Electron probe microanalysis after rolling confirmed that the Ni concentration was 59.5 ± 0.65 at% and Nb concentration was 40.5 ± 0.25 at%, which is quite close to the nominal composition of Ni₆₀Nb₄₀.

The as-rolled Ni₆₀Nb₄₀ became very brittle due to the mechanical deformation, so focused ion beam (FIB) was performed to prepare the TEM sample. For as-spun Ni₆₀Nb₄₀, both electropolishing and FIB were used to make TEM samples. For electropolishing, samples were prepared by a standard twin-jet electro-polisher with electrolyte of 10% H₂SO₄ in methanol [20]. The temperature range for electropolishing was between −20 °C and −25 °C and samples were electro-polished at 42 V. For FIB, TEM samples were prepared using a Zeiss Auriga cross beam FIB system. Firstly, a platinum protection layer with ~1 μm in thickness was deposited by ion beam and a sample membrane of about 15 μm × 10 μm × 2 μm in size was lifted out using 30 kV, 2 nA Ga⁺ ion milling. Then, the sample was attached to copper grid for further milling. The milling recipe was: sample was milled to ~200 nm thick by 30 kV, 80 pA Ga⁺ ion, then 30 kV, 20 pA Ga⁺ ion was applied until it became electron transparent in the 7 kV scanning electron microscopy (SEM) imaging. To remove surface damage, 2 kV, 100 pA Ga⁺ ion was used for final thinning until sample was electron transparent in the 3 kV SEM imaging.

2.2. Characterization

X-ray diffraction (XRD) spectra were collected using a Bruker D8 Discover diffractometer with a Cu-Kα micro X-ray source (λ = 1.541 Å). The 2θ angular range between 10° and 90° was explored at a scan rate of 2°/min with a Vantec 500 area detector. The diffracted intensity was obtained by integrating around the diffraction rings captured on the detector. Differential scanning calorimetry (DSC) measurements were conducted at 20 °C/min heating rate under a flow of purified argon in a TA Instruments Q100. The temperature accuracy of DSC experiment is ± 0.1 °C. The HRTEM was performed on a FEI Tecnai-TF30 microscope with field emission gun source, operated at 300 kV. Images with 2048 by 2048 pixels were recorded on a Gatan Ultrascan CCD with 1 s exposure time.

MRO occupies the length scale between short range order (SRO) and long-range order, which does not exist in glasses. SRO in the form of a pair distribution function can be measured experimentally by wide-area diffraction with x-rays, neutrons, or electrons. FEM is an effective method to detect MRO structure [21–24] from the normalized variance

V of a set of electron nanodiffraction patterns by calculating defined as,

$$V(k, Q) = \frac{\langle I^2(k, Q, r) \rangle}{\langle I(k, Q, r) \rangle^2} - 1, \quad (1)$$

where values within $\langle \rangle$ are averaged over the position r on the sample. I is the spatially-resolved diffracted intensity. R is the spatial resolution and can be defined by the Rayleigh criterion, $R = 0.61/Q$ [24], in which Q is the radius of the virtual objective aperture in the scanning transmission electron microscopy (STEM) mode. k is the scattering vector magnitude. V depends on the three- and four-atom correlation functions of the sample. The position in k of peaks in V is controlled by the interatomic spacing inside nanoscale structural heterogeneities, and the magnitude of V is controlled by their size, density, and internal order. One way to define MRO is in terms of preferred packing of tightly-bound nearest-neighbor clusters of atoms. The clusters then define the material's SRO, and how they fit together define its MRO. FEM is particularly sensitive to well-ordered MRO, but it also encodes some SRO-scale information, since preferred interatomic distances within the SRO clusters control the k position of at least some of the peaks in $V(k)$.

FEM experiments were performed on a probe aberration corrected FEI Titan at 200 kV. The microscope was operated in the microprobe STEM mode (μP-STEM mode) to obtain a wider range of Q . The camera length was 512 mm and GIF energy filtering with a slit width of 10 eV was used to remove inelastically scattered electrons. A highly coherent beam of 2 nm in diameter was formed with 0.74 mrad semi-convergence angle. The 512 by 512 pixel nanodiffraction patterns were acquired using Gatan US1000 CCD camera. To ensure that the step size of scanning was larger than the diameter of probe, a 30 by 30 nm² area at 1.3 Mx magnification was selected for 10 by 10 grid positions. The acquisition time for each diffraction pattern was 6 s. The sample thickness was calculated based on the elastic mean free path and the elastic scattering transmittance. Because the elastic scattering in amorphous materials obeys the Poisson statistics, it can be described by

$$t/\lambda = \ln(I_t/I_0), \quad (2)$$

where λ is the elastic mean free path, which is ~35 nm for the Ni₆₀Nb₄₀ metallic glass [25]; t is the sample thickness; I_t is the total number of incident electrons; I_0 is the number of the unscattered electrons.

2.3. Simulations

HRMC simulations [26] were performed to refine the atomic structure for both the as-spun and as-rolled states. The simulation was simultaneously constrained by minimizing the total energy E and the squared difference between the experimental variance $V_e(k)$ and simulated variance $V_s(k)$. The simulation minimized the cost function

$$\chi^2 = E + \alpha \sum_i \left[\frac{V_e(k_i) - \beta \cdot V_s(k_i)}{\sigma_{err}} \right]^2, \quad (3)$$

where σ_{err} is the uncertainty in $V_e(k)$. The weighting factor α was adjusted to ensure that the energy E and the variance discrepancy between simulation and experiment decreased at the same rate as the simulation proceeded. β is a correction factor to adjust the variance difference in magnitude between $V_e(k)$ and $V_s(k)$ caused by the thickness difference between the sample and the simulation model [21,26]. β is typically defined as $\frac{1}{3} \frac{t_{sim}}{t_{exp}}$. The experimental thickness t_{exp} calculated from the elastic mean free path and elastic electron transmittance [25] is 29 ± 2 nm and the simulation thickness t_{sim} is 2.83 nm, so here β is 0.0329. The embedded atom model (EAM) potential for Ni–Nb was developed by H. Sheng [27].

HRMC requires a starting structure for refinement. The equilibrium density for the starting structure was determined by molecular dynamics quenching of the liquid in the NPT ensemble using the EAM

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