Analysis of Microstructural Characteristics in Near-Eutectic Ni-P Amorphous Alloy Postprint

Authors: Peng Chao, Peng Ping

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Abstract

The rapid solidification process of liquid metal Ni100-xPx (x=19.0, 19.4, 19.6, 19.8, 20.0, 21.0) alloys at a cooling rate of 5×1012K/S was simulated using the molecular dynamics method, and the local atomic structures were characterized using Voronoi polyhedron indices ?n__3,n__4,n__5,n__6 ? and cluster-type indices (Z n__i?(ijkl)__i ···). The results show that the cluster characteristics of Ni atoms are mainly high-coordination (Z 12) Frank-Kasper clusters and their distorted variants, with a typical chemical short-range order of NiZ-2P3, and basic clusters can form medium-range order structures through IS linkages; whereas the local structures of P atoms, in addition to Z=10 BSAP polyhedra, also contain a large number of high-coordination (particularly Z=12) Frank-Kasper structural motifs, with a typical chemical short-range order of Ni12P; moreover, the shell atoms of P-centered basic clusters are entirely Ni, and they can only form extended clusters through VS, ES, and FS linkages. BSAP polyhedra and their related structures were confirmed to have a significant influence on the formation of Ni100-xPx amorphous alloys, with their population being maximum at the eutectic point x=19.6; the further the deviation from the eutectic point, the smaller the proportion, which is consistent with the variation trend of the glass-forming ability of Ni100-xPx alloys at different compositions. This may be the reason why Ni-P alloys have the strongest glass-forming ability at the eutectic point.

Full Text

Atomistic Simulation for Local Atomic Structures of Amorphous Ni-P Alloys with Near-eutectic Compositions

PENG Chao, LI Yuan, DENG Yonghe, PENG Ping
School of Material Science & Engineering, Hunan University, Changsha 410082, China
Correspondent: PENG Ping, professor, Tel: 13873119465, Email: ppeng@hnu.edu.cn

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Abstract

Ni100-xPx alloys with near-eutectic compositions exhibit strong glass forming ability (GFA), yet the microstructure prototypes and their evolution during various solidification processes remain unclear. To elucidate their unique structures, we performed a series of molecular dynamics simulations of the rapid solidification process for liquid Ni100-xPx (x=19.0, 19.4, 19.6, 19.8, 20.0, 21.0) alloys at a cooling rate of 5×10^12 K/s, and characterized their local atomic configurations at 300 K using Voronoi polyhedron index ⟨n1, n2, n3, n4⟩ and cluster type index methods. The results reveal that the local atomic structures of Ni atoms are predominantly Frank-Kasper clusters with high coordination (Z 12) and their distorted configurations, with typical chemical short-range orders of NiZ-2P3. These basic clusters can aggregate into medium-range orders (MROs) through intercross-sharing (IS) linkages. The majority of P-centered clusters are bi-capped square Archimedean anti-prism (BSAP) polyhedrons, though numerous Frank-Kasper clusters with higher coordination also exist in the amorphous Ni100-xPx alloys. Their typical chemical short-range orders are Ni12P. In these short-range orders (SROs) centered by P, all shell atoms are found to be Ni, and no MRO can be detected except for their extended clusters linked by vertex-sharing (VS), edge-sharing (ES) and face-sharing (FS). BSAP polyhedrons and their correlative structures play a crucial role in the formation of amorphous Ni100-xPx alloys. Their quantity demonstrates a significant impact on the glass transformation of rapidly solidified Ni100-xPx alloys. Notably, the number of BSAP polyhedrons and their deformed structures reaches a maximum at the eutectic composition point x=19.6 among Ni100-xPx alloys, and the farther x deviates from the eutectic composition, the smaller the proportion of BSAP polyhedrons and their related structures among all P-centered clusters. This trend is consistent with the variation tendency of GFAs of Ni100-xPx alloys, which may be responsible for the strongest GFA at the eutectic composition point of Ni100-xPx alloys.

KEY WORDS amorphous Ni-P alloy, molecular dynamics, BSAP cluster, glass forming ability

Introduction

Since Brenner and Riddle first electrodeposited Ni-P alloy coatings in the 1950s, the structure and properties of Ni-P alloys have been extensively investigated. Due to their amorphous structure at high phosphorus contents, these alloys lack crystal defects such as grain boundaries and dislocations, as well as chemical segregation. Combined with their inability to form passive surface films, they exhibit excellent chemical and mechanical properties including corrosion and wear resistance, leading to widespread applications in machinery, chemical, electronics, and petroleum industries.

Ni100-xPx alloys possess strong glass forming ability. In addition to amorphous coatings obtained through electrodeposition, amorphous ribbons can be prepared via rapid quenching over a wide composition range (x=16.5~21.0 at%). According to the Ni-P equilibrium phase diagram compiled by Nash, these compositions are located near the eutectic point (x=19.0, Tm=1143 K), though this eutectic point has been questioned. Schmetterer et al. determined the actual eutectic temperature for Ni-rich alloys to be 1163 K, while Huang et al. further identified the eutectic composition as x=19.6. Building on this, Huang et al. prepared Ni100-xPx amorphous alloy ribbons with different P contents and measured the reduced glass transition temperature Trg=Tg/Tm (where Tg and Tm are the glass transition temperature and equilibrium melting point, respectively) and critical thickness Dc. They found that both Dc and Trg reached maximum values at the eutectic point x=19.6, confirming that transition metal-metalloid (TM-M) alloys exhibit the strongest glass forming ability near the eutectic composition.

Numerous studies have demonstrated that glass forming ability is closely related to the local atomic structure within alloys. Bennett and Wright found that the nearest neighbor coordination number in Ni100-xPx (x=19, 21, 23, 24, 26) amorphous coatings is typically 13.0. Based on crystallization phase structures, Lu et al. proposed the existence of two types of ordered atomic clusters: Ni-Ni type structural units forming Ni solid solutions and Ni-P type units forming Ni3P compounds. Using EXAFS (extended X-ray absorption fine structure) spectroscopy combined with Monte Carlo simulations, Luo and Ma revealed that Voronoi polyhedrons in Ni80P20 amorphous alloys are primarily <0,2,8,0> bicapped square Archimedean anti-prism (BSAP) clusters. While these studies have provided initial insights into the microstructural features of Ni100-xPx amorphous alloys, they did not address the relationship between local atomic structure and glass forming ability. Since Ni100-xPx alloys exhibit the strongest glass forming ability at x=19.6, unique structural features likely exist at this composition. Therefore, this work aims to conduct a comparative investigation of local atomic structures in Ni100-xPx amorphous alloys near the eutectic point.

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1. Calculation Conditions and Methods

Molecular dynamics (MD) simulations were performed using the LAMMPS package. The simulation system consisted of a cubic box containing 10,000 atoms randomly placed according to Ni100-xPx (x=19.0, 19.4, 19.6, 19.8, 20.0, 21.0) atomic ratios. The simulations employed an isothermal-isobaric (NPT) ensemble, periodic boundary conditions, and an EAM (Embedded Atom Method) interatomic potential. The initial temperature was set to 1600 K with a time step of 1 fs. Temperature and pressure (P=0) were controlled using a modified Nose-Hoover method, and the Verlet algorithm was used to integrate the equations of motion.

The system was first equilibrated at constant temperature for 1 ns, then cooled to 300 K at a rate of 5×10^12 K/s. Data were recorded every 5 K to capture atomic coordinates and velocities at each temperature. The microstructures at different solidification stages were characterized using pair distribution functions, Voronoi polyhedron index, and cluster type index methods.

2.1 Pair Distribution Function

[Figure 1: see original paper] shows the pair distribution functions g(r) for rapidly solidified Ni100-xPx alloys at 300 K. As seen in Figure 1a, the second peak of the total pair distribution function gtot(r) splits into two sub-peaks, exhibiting a clear shoulder characteristic of typical amorphous structures, with an average coordination number Z 12.4 per atom. Figure 1b further reveals that Ni atoms have approximately 10.5 Ni neighbors and 2.4 P neighbors on average, while the first peak of the partial pair distribution function gP-P(r) is absent, indicating a very low probability of direct P-P bonding (the average P neighbor count for P atoms is effectively zero).

2.2 Voronoi Polyhedron Index Method Analysis

While pair distribution functions provide only one-dimensional statistical distributions, they cannot precisely describe three-dimensional atomic configurations. Therefore, we first employed the Voronoi polyhedron index method (VPIM) to characterize the local atomic structure of Ni100-xPx amorphous alloys. Similar to Wigner-Seitz cells, Voronoi tessellation uses the smallest closed polyhedron ⟨n1, n2, n3, ···ni···⟩ formed by perpendicular bisector planes between neighboring atoms to describe the local configuration around a central atom, where ni represents the number of i-edged faces and Σni = Z denotes the coordination number. In practice, parameter corrections are applied to eliminate distortions caused by small edges and faces, typically using a 4-index notation ⟨n1, n2, n3, n4⟩ to characterize cluster structure types.

Table 1 lists the types and quantities of major Voronoi polyhedrons in Ni100-xPx amorphous alloys at x=19.6; other compositions show similar distributions. The table reveals diverse Voronoi polyhedrons in Ni100-xPx amorphous alloys, predominantly those with 12 and 13 coordination, such as <0, 2, 8, 2>, <0, 1, 10, 2>, and <0, 3, 6, 4>. Most P-centered Voronoi clusters appear only at Z<12, while high-coordination structures (Z 12) are essentially Ni-centered clusters. The system also contains numerous icosahedra <0, 0, 12, 0> and their distorted variants (e.g., <0, 2, 8, 2> and <0, 3, 6, 3>), but these are mostly Ni-centered rather than P-centered. P-centered Voronoi clusters are primarily <0, 2, 8, 1>, <0, 2, 8, 0>, <0, 3, 6, 1>, and <0, 3, 6, 0>. Three-dimensional visualization reveals that <0, 2, 8, 0> is a standard BSAP polyhedron, <0, 3, 6, 1> is a distorted BSAP polyhedron, <0, 3, 6, 0> is a tri-capped trigonal prism (TTP) polyhedron, and <0, 2, 8, 1> is a defective icosahedron <0, 0, 12, 0>, as shown in Figure 2 [Figure 2: see original paper].

2.3 Cluster Type Index Method Analysis

The cluster type index method (CTIM) is a local structure characterization approach based on Honeycutt-Andersen (H-A) bond-type analysis, typically using the notation (Z n1/ijkl n2/ijkl···) to scale cluster properties. Here (ijkl) represents the H-A bond-type index, ni denotes the number of (ijkl) bond pairs between the central atom and its neighbors, and Z = Σni is the coordination number.

Analysis reveals over 330 distinct CTIM clusters in near-eutectic Ni100-xPx amorphous alloys. Table 2 shows the distribution of typical CTIM clusters with quantities exceeding 30 for x=19.6. The data indicate that Ni-centered clusters exhibit various structural morphologies but are dominated by high-coordination Frank-Kasper clusters and their distorted configurations, such as Z=14 clusters (14 2/1441 8/1551 4/1661), (14 1/1441 10/1551 3/1661), and (14 3/1441 6/1551 5/1661); Z=13 clusters (13 1/1441 10/1551 2/1661) and (13 3/1441 6/1551 4/1661); and Z=15 clusters (15 2/1441 8/1551 5/1661) and (15 1/1441 10/1551 4/1661). While P-centered clusters are primarily low-coordination (Z<12), numerous high-coordination (Z 12) Frank-Kasper clusters also exist, such as Z=12 (12 2/1441 8/1551 2/1661) and Z=13 (13 3/1441 6/1551 4/1661). The typical chemical short-range order for Ni-centered CTIM clusters is NiZ-2P3, corresponding to Ni12P3, Ni11P3, and Ni13P3 for Z=14, 13, and 15, respectively, plus minor amounts of Ni13P2, Ni12P2, and Ni12P4. Pure NiZ+1 clusters are absent (Table 3).

Graphical analysis shows that regardless of coordination number, the shell atoms of P-centered CTIM clusters are exclusively Ni. Statistical results for P-centered CTIM clusters show quantities of 82, 200, 504, 286, 64, and 4 for Z=10, 11, 12, 13, 14, and 15, respectively, indicating a dominant chemical short-range order of Ni12P with minor Ni13P and Ni11P. These P-centered clusters mostly exist as isolated units, with only a small fraction forming extended clusters through vertex-sharing (VS), edge-sharing (ES), or face-sharing (FS). No medium-range structures formed by intersect-sharing (IS) patterns were observed (Table 4). In contrast, Ni-centered clusters can form large extended clusters and MROs through IS linkages, and even bond with P-centered clusters to form Ni-P mixed clusters, as shown in Figure 3 [Figure 3: see original paper]. Although P-centered clusters cannot form MROs, the Ni atoms in their shells account for 7,992 out of 8,040 total atoms (99.4%).

Therefore, subsequent analysis of local atomic structure features in Ni100-xPx amorphous alloys will focus on P-centered clusters.

2.4 Local Structural Analysis of P Atoms in Ni100-xPx Amorphous Alloys

Table 5 presents the types and quantities of Voronoi polyhedrons centered on solute P atoms. The data show that over 80% of P-centered Voronoi polyhedrons are accounted for in all Ni100-xPx alloys, with <0, 2, 8, 0>, <0, 3, 6, 1>, <0, 2, 8, 1>, and <0, 3, 6, 0> being the most abundant. The standard BSAP polyhedron <0, 2, 8, 0> and its distorted variant <0, 3, 6, 1> constitute approximately 35% of the total, confirming that BSAP polyhedrons are indeed characteristic structures of P-centered clusters in Ni100-xPx amorphous alloys. Additionally, significant populations of TTP polyhedrons <0, 3, 6, 0> and defective icosahedra <0, 2, 8, 1> exist. Other polyhedrons such as icosahedra <0, 0, 12, 0> and <0, 4, 4, 1>, <0, 4, 4, 2>, <0, 3, 6, 2>, and <0, 4, 4, 3> are relatively scarce. Graphical analysis reveals that these non-mainstream Voronoi polyhedrons are primarily Frank-Kasper clusters with Z=9 and Z=11 and their distorted configurations.

Figure 4 [Figure 4: see original paper] further shows the variation of <0, 2, 8, 0> and <0, 3, 6, 1> polyhedron counts with P content. The proportion of standard BSAP polyhedrons and their distorted structures varies with composition but reaches a maximum at the eutectic point x=19.6, mirroring the trend of glass forming ability characterized by Trg in Ni100-xPx alloys. This suggests a correlation between P-centered BSAP polyhedrons and glass forming ability: higher populations of BSAP polyhedrons correspond to stronger glass forming ability.

Given that CTIM provides more detailed information about local atomic structure variations than VPIM (for instance, VPIM cannot distinguish between fcc and hcp basic clusters using <0, 12, 0, 0>, whereas CTIM can differentiate them as (12 12/1421) and (12 6/1421 6/1422)), we further analyzed typical P-centered CTIM clusters in Ni100-xPx amorphous alloys. The results (Table 6) show that while standard BSAP clusters (10 2/1441 8/1551) and their distorted variants (10 4/1551 2/1422 4/1431), (10 1/1441 5/1551 1/1541 3/1431), (10 1/1441 2/1551 1/1421 2/1541 4/1431) are not particularly abundant, the local atomic configurations of P are dominated by Z=12 clusters such as (12 2/1441 8/1551 2/1661), (12 2/1441 4/1551 2/1661 2/1541 2/1431), (12 3/1441 6/1551 3/1661), and (12 8/1551 2/1541 2/1431), as well as Z=11 clusters (11 2/1441 8/1551 1/1661), (11 1/1441 6/1551 2/1541 2/1431) and Z=13 clusters (13 3/1441 6/1551 4/1661). Although this variety complicates feature extraction, the total quantities of these clusters, particularly Z=10 and Z=12 CTIM clusters, show a composition dependence similar to BSAP polyhedrons (Figure 5 [Figure 5: see original paper]), indicating that the typical local structure of solute P can indeed characterize the glass forming ability of Ni100-xPx alloys.

Further analysis reveals that many Z=10 BSAP polyhedrons and their distorted structures in Table 5 actually correspond to higher-coordination (Z 11) CTIM clusters such as (11 2/1441 8/1551 1/1661) and (12 2/1441 8/1551 2/1661), where some coordinating atoms corresponding to polygonal faces are eliminated by VPIM parameter corrections (Figure 6 [Figure 6: see original paper]). Therefore, the composition dependence of BSAP polyhedrons in Figure 4 should match that of Z=10 and Z=12 CTIM clusters in Figure 5.

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3. Conclusions

1. Near-eutectic Ni100-xPx (x=19.0, 19.4, 19.6, 19.8, 20.0, and 21.0) amorphous alloys contain numerous short-range order structures. Ni atoms predominantly form high-coordination (Z 12) Frank-Kasper clusters and their distorted configurations with typical chemical short-range orders of NiZ-2P3. P atoms exhibit local configurations dominated by Z=10 BSAP polyhedrons but also contain substantial high-coordination (particularly Z=12) Frank-Kasper structures, with most BSAP polyhedrons in VPIM analysis having Z>10 and typical chemical short-range orders of Ni12P.
2. The shell atoms of P-centered basic clusters are exclusively Ni, which can only form extended clusters through VS, ES, and FS linkages. In contrast, Ni-centered basic clusters can additionally form MROs through IS linkages and even bond with P-centered clusters to create Ni-P mixed clusters.
3. The proportion of P-centered BSAP polyhedrons and their related structures in near-eutectic Ni100-xPx amorphous alloys reaches a maximum at the eutectic point x=19.6, consistent with the composition dependence of glass forming ability characterized by reduced glass transition temperature Trg and amorphous ribbon thickness Dc. This suggests that P-centered BSAP clusters significantly influence the formation of Ni100-xPx amorphous alloys, and their population may serve as a metric for evaluating the glass forming ability of rapidly solidified Ni100-xPx alloys.

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