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Weighted Voronoi tessellation technique for internal structure of metallic glasses

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Abstract

Voronoi tessellation technique has been widely used to describe the atomic structure of amorphous materials, such as short-range order of icosahedra, because of the relative simplicity of calculation. In the ordinary Voronoi tessellation technique, a face is placed halfway between equal-sized atoms. However, as the atoms are not of the same size, bisection cannot be the exact plane to represent the polyhedron. It also produces errors during estimation of volume of each atom and the number of faces on the polyhedron. In this paper, we solve this problem using Richards's method, a kind of weighted Voronoi tessellation technique. The results show over 50% differences in the number of faces and the atomic volume for the binary metallic glasses.

Keywords: B. Glasses, metallic; D. Microstructure; E. Simulations, atomistic

1. Introduction

Since the first synthesis of the system with amorphous phase, metallic glasses have been widely studied for a few decades. Especially, both experimental and theoretical studies focus on the structural characteristic of metallic glasses [1]. It has been acceptable that despite lack of long-range order as in the crystals, there exists short-range order formed due to the heterogeneous internal structure of glass materials. In addition, the earlier MD study has suggested that a possible structural unit of amorphous material is the icosahedra [2]. Many studies indicate that the macroscopic structure of metallic materials depends on their internal structures, so that it is important to precisely analyze their structures. The well-known, ordinary Voronoi tessellation technique is usually used to probe them in the researches for the formation of amorphous alloys using molecular dynamics [3]. In the ordinary Voronoi tessellation technique, the Voronoi polyhedron is assigned to each atom. Then the distribution of the atom with the given number of faces indicates the local structure around the corresponding atom. A common method for calculating volume and studying packing of spheres is originally developed by Voronoi [4]. In biology, calculating the volume of atom also has significant meaning because precise volume calculation of protein is required for prediction about thermostability of cavity-filling mutants [5]. In the ordinary Voronoi tessellation technique, a face is placed halfway between atoms. Then, the edges of the polyhedron made by Voronoi tessellation are formed by the intersection of these faces. However, as the atoms are not of the same size, bisection cannot depict the exact plane to represent the polyhedron. It produces errors of volume calculation of each atom and the number of faces on the polyhedron. Therefore, several researchers propose methods to divide plane subject to atomic size. Richards suggests an alternative way that the ratio of the distance between the atoms and the plane equals the ratio of the atomic radii [6]. In the radical plane method, Gellatly and Finney position the plane in the intersection circle of the atoms [7]. Gerstein et al. first introduce non-planar boundaries, called Gerstein spheres, between atoms, such that the ratio of distances to

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the centers of the neighboring atoms is constant [8]. In this paper, we introduce the Richard's method employed in biology and apply it to the internal structural analysis of the metallic glasses. Then, the results are compared with the ordinary Voronoi tessellation technique.

2. Computational model

The binary systems consisting of 1300 of A atoms and 700 of B atoms are subjected to periodic boundary condition and constant pressure. Molecular dynamics study using modified Lennard–Jones potential is adopted. The A atom is modeled as Zr atom. B atom is a fictitious atom with smaller atomic radius than A atom. The size of B atom is changed, to describe the effect of atomic size. The interatomic potential between A atoms is modeled by the modified Lennard–Jones potential for Zr atom. And the other potentials are defined by potentials similar to the A–A interactions but whose shape and position are scaled by the size of the B atoms. For example,

$$\phi_{B-B}(r) = \phi\left(\frac{R_A}{R_B}r\right) \tag{1}$$

$$\phi_{A-B}(r) = \phi\left(\frac{2}{(1+R_B/R_A)}r\right) \tag{2}$$

where r is the distance, R_A and R_B are the atomic radii of A atom and B atom, respectively.

At the first step, all atoms are randomly positioned in the cubic cell with the size of about 35 Å. Then, the system is fully heated for 235 ps, relaxed for 1000 ps and cooled down for 4700 ps with periodic boundary condition and 10^{12} K/s cooling rate. During the process, Parinello–Rahman method is applied to keep constant pressure [9]. Finally, we obtain the models with amorphous structure.

3. Weighted Voronoi analysis [6,10]

In the ordinary Voronoi tessellation technique derived from a collection of points rather than the spheres with size, the entire space is partitioned among a collection of equal-sized atoms. The volume of polyhedron assigned to each atom consists of tetrahedra derived by Voronoi tessellation technique. Each face of Voronoi polyhedra is derived from the planes bisecting and perpendicular to the interatomic vectors. Then, the edges of polyhedra are formed by the intersection of these planes. Hence, in the ordinary Voronoi tessellation technique, the locations of all the neighboring atoms are the only necessary information to define the polyhedron. This procedure can be easily done with equally sized atoms. However, as atoms are not of the same size, such as Cu (radius = 1.28 Å) and Zr (radius = 1.60 Å) atoms, bisection cannot depict the real face like in Fig. 1. The volume of the smaller atom is overestimated and the number of faces for the given atom is miscalculated. This unfair partition misleads the calculation of atomic volumes and their structures. In summary, modifications of the Voronoi tessellation technique are necessary to account for the volume and structure of the amorphous structures with more than two compositions.

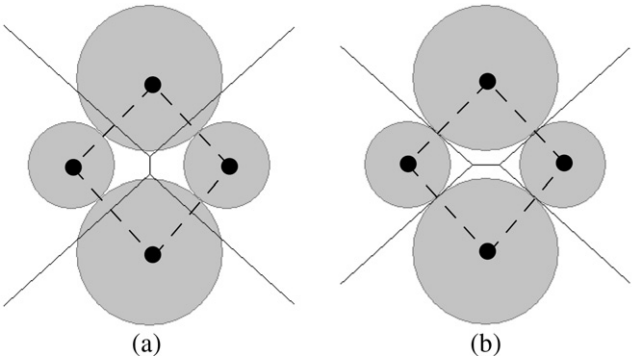

Fig. 1. Two-dimensional schematics of (a) the ordinary Voronoi tessellation technique and (b) the weighted Voronoi tessellation technique. On the ordinary Voronoi tessellation, the size of smaller atoms is overestimated. The atomic structure, i.e., the number of face of each atom also depends on the atomic size.

Researchers in biology field also have had the similar difficulties. The mean density of protein should be close to that of organic crystals showing only local density deviation. To achieve better comparisons of the function-relevant parts of the molecules, emphasis is placed upon accurate volume calculation. And it is also related to the thermostability of cavity-filling mutants [11]. To consider the effect of atomic size, Richards suggests a kind of weighted Voronoi tessellation technique that the distance from the atom to the face of polyhedron is proportional to the atomic radii [6]. Fig. 2 shows the schematic of the Richards's way to define the dividing plane.

Firstly, the dividing plane is positioned between atoms in proportional to the given atomic radii. Then, the dividing plane is placed farther from large atom according to the following:

$$S = \frac{R}{R+r}D \tag{3}$$

where S is the distance of the plane from the large atom, D is the distance between two atoms, R and r are radii of the large atom and small atom, respectively. Fig. 2 shows the schematic of these parameters. Now, it is impossible to decide the vertices of polyhedra simply as the ordinary Voronoi tessellation technique, since the vertices are no longer at center of sphere

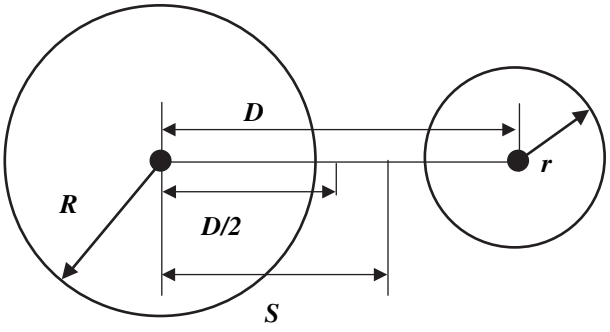

Fig. 2. The schematic of Richards's proportional partitioning method.