Journal of Alloys and Compounds 757 (2018) 228–232

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Fractal analysis on the cluster network in metallic liquid and glass

M.H. Yang, J.H. Li*, B.X. Liu

Key Laboratory of Advanced Materials (MOE), School of Materials Science and Engineering, Tsinghua University, Beijing 100084, China

ARTICLE INFO

Article history:
Received 24 March 2018
Received in revised form
28 April 2018
Accepted 6 May 2018
Available online 9 May 2018

Keywords:
Amorphous materials
Atomic scale structure
Quenching
Computer simulations

ABSTRACT

To explore the atomic-level structure of metallic glasses, liquid melt quenched Tantalum was obtained by computer simulations, which mimics the experimental approach to the vitrification of metallic liquids by achieving an unprecedentedly high liquid-quenching rate. Through the local atomic structure analysis of the quenched Tantalum, it is shown that the atomic clusters assemble in space to further form skeletal networks by sharing various linking patterns, *i.e.* vertex, edge, face or volume linkages. By employing the established box-counting method, the cluster network is revealed to exhibit a fractal characteristic. The fractal dimension of the atomic cluster network is computed to be 2.3–2.5. Compared to other indirect fractal analysis method, the present work first provides a straightforward evidence of fractal structure, which promotes the understanding to the atomic configuration of metallic glasses.

1. Introduction

Differing from the crystalline metals, which show sharp Bragg peaks by diffraction techniques because of the long-range periodically arrangements of atoms, metallic glasses (MGs) exhibit an apparently disordered structure that is metastable in nature, and present a maze-like pattern at the atom-level scale [1–3]. Although tremendous efforts have been devoted to reveal the arrangements of atoms in MGs, the short-range order (SRO) and medium-range order (MRO) structures in disordered materials will inevitably break down over the long scale because of the spatial incompatibility [2,4,5]. Therefore, both the SRO and MRO structures could not fully describe and characterize the atomic configuration of MGs. Coincidentally, the fractal concept indicates the underlying order in inherently random and chaotic arrangements, and may provide a picture of the hierarchical structures in MGs, as well as many dynamical properties, *e.g.* aging [6], plasticity [7] and glass transition [8]. Due to the characteristics described by self-similarity, fractal dimension and scale invariance [9–11], the fractal geometry has become a powerful tool to study the structural properties in a great number of condensed-matter systems [12,13].

The proposition of fractal packing in MGs is enlightening, and considerable research work has been suggested to study the atomic scale structure. Zeng et al. [14] have revealed that the dimensionality of metallic glasses is about 2.5 by employing high pressure to probe the density of different MGs. Interestingly, Ma et al. [10] have performed the neutron and X-ray diffraction (XRD) experiments to study the self-similar packing at the MRO scale, and obtained a universal fractal dimension of 2.3 for a variety of bulk MGs. Chen et al. [2] have observed a fractal short-range around 2.5, whereas a homogeneous long-range with dimensionality of 3. However, a few researchers hold the opposite viewpoint on the fractal packing. Cheng et al. [7] have declared that the evidence showing the existence of fractal structure in MGs were not observed in their recent studies. Although these experimental fractal analysis of MGs structure bring new insights into the structural mechanism of MGs system, the atomic structural origin of the fractal dimension still remains a mystery.

Considering the heated debate over fractal analysis of MGs, the previous fractal dimensionalities are derived from the changes in the radial distribution functions (RDFs) and structural factors induced by pressure and compositional variations, rather than directly computed by the fractal definition from the atomic configuration of MGs. Besides, the previously proposed fractal analysis can't provide the atomic structural origin of fractal dimension, which may be unsound and misleading because of the heterogeneous nature of MGs structure and many other variables, *e.g.* topological structure, scattering factors, thermal history, chemical interaction and its nonaffine deformation [15–17].

It is well known that the basic structural unit in amorphous materials is the atom cluster [18–20]. The present authors believe that these clusters aggregate to form complicated congeries in space by sharing various linking patterns, *i.e.* vertex, edge, face and

* Corresponding author.
E-mail address: lijiahao@mail.tsinghua.edu.cn (J.H. Li).

https://doi.org/10.1016/j.jallcom.2018.05.069
0925-8388/

volume. The congeries pile up further in a complex manner to form a network, *i.e.* the skeleton of clusters that exhibit fractal characteristics. Here, we try to directly investigate the fractal characteristics of the skeleton of clusters by the method of box counting [9,21]. In order to eliminate the interference of other trivial factors and simplify the analysis and characterization, we devote to explore the atomic configuration of liquid melt quenched Tantalum because it has analogous characteristic with common metallic glasses in structure.

2. Methods and simulations

Basing on the newly constructed potential [25], Molecular Dynamics (MD) simulations were performed to obtain the liquid melt quenched Tantalum, by using large-scale atomic/molecular massively parallel simulator (LAMMPS) packages [26]. In the simulation, the initial *bcc* Ta model containing 2.0 million atoms was constructed on the three-dimensional cubic box with dimensions of 33.5 nm (x) × 33.5 nm (y) × 33.5 nm (z). Periodic boundary conditions were adopted in three Cartesian directions. MD simulation was performed in the framework of NPT ensemble with a time step of 5 femtoseconds. The Nose-Hoover thermostat and barostat were implemented to control the temperature and pressure, respectively. The initial Ta alloy was firstly heated from 300 K to 5000 K at a rate of 10^{13} K/s and then equilibrated at 5000 K for 100 ps. The equilibrium temperature was high enough to enable the system to fully relax. Afterwards, the equilibrium system was quenched from 5000 K to 300 K at the rate of 10^{14} K/s, and then the system relaxed again at 300 K for 100 ps to achieve a meta-stable state, where the related dynamic variables present no secular variation. The cooling effect mimics the experimental approach to the vitrification of Ta metallic liquids and glass [24].

The radial distribution functions $g(r)$ and structural factor $S(q)$ of the liquid melt quenched Tantalum was computed as presented in Fig. 1. The melt point T_m of Ta was estimated to be around 3500 K from the significant energy jump, close to the experimental observation [27]. Besides, the energy change with temperature in the cooling process denotes the glass transition temperature T_g as 1700 K, which is also near the experimental result [24]. By comparison, the present calculated $g(r)$ in Fig. 1(a) matches well with *ab initio* calculation [22] and Jiang et al.'s work [23]. In addition, the main peak positions of the $S(q)$ in Fig. 1(b) were measured to be 2.65 $Å^{-1}$, 4.45 $Å^{-1}$ and 5.24 $Å^{-1}$, corresponding to $q_2/q_1 = 1.68$ and $q_3/q_1 = 1.98$. These results are consistent with Zhong et al.'s simulated work by using different atomic potential, as well as the experimental structural factor through ultrafast quenching liquid method at the same cooling rate of 10^{14} K/s [24].

Based on the unsupervised machine-learning algorithm, *hierarchical clustering analysis* was performed to identify and characterize the space distribution of the data point set [28,29]. It should be noted that the *cluster analysis* is also referred to the segmentation analysis or taxonomy analysis, *i.e.* partitioning all of the data points into segmentation or taxonomy. Specifically, *Hierarchical Clustering* groups the data point set over various scales by creating a multilevel hierarchy of the dendrogram. The Statistic and Machine Toolbox^{TM} function *clusterdata* performs all the necessary steps and consists of all the necessary steps, *i.e. pdist*, *linkage*, and *cluster* functions. Firstly, the similarity or dissimilarity between every pair of data points in the group was determined by using the Euclidean distance. Secondly, the data points were grouped into a binary, hierarchical dendrogram. As data points are paired into binary clusters, the newly formed clusters are grouped into larger clusters until a hierarchical dendrogram is determined. Thirdly, the cluster function, $T = cluster(X, \text{cutoff}, c)$ was used to create clusters by detecting natural groupings in the hierarchical dendrogram. Here c is the inconsistency coefficient ($0 < c < 2$) and a threshold for cutting the hierarchical dendrogram X. The inconsistency coefficient describes each link in the dendrogram by comparing its height with the average height of other links of the hierarchy in the same level. The higher the inconsistency coefficient c, the less similar the data points connected by the link. In present work, an optimized inconsistency coefficient 1.0 was used to ensure the distance between the data points being joined is approximately the same as the distances between the data points they contain, exhibiting the links have a high level of consistency.

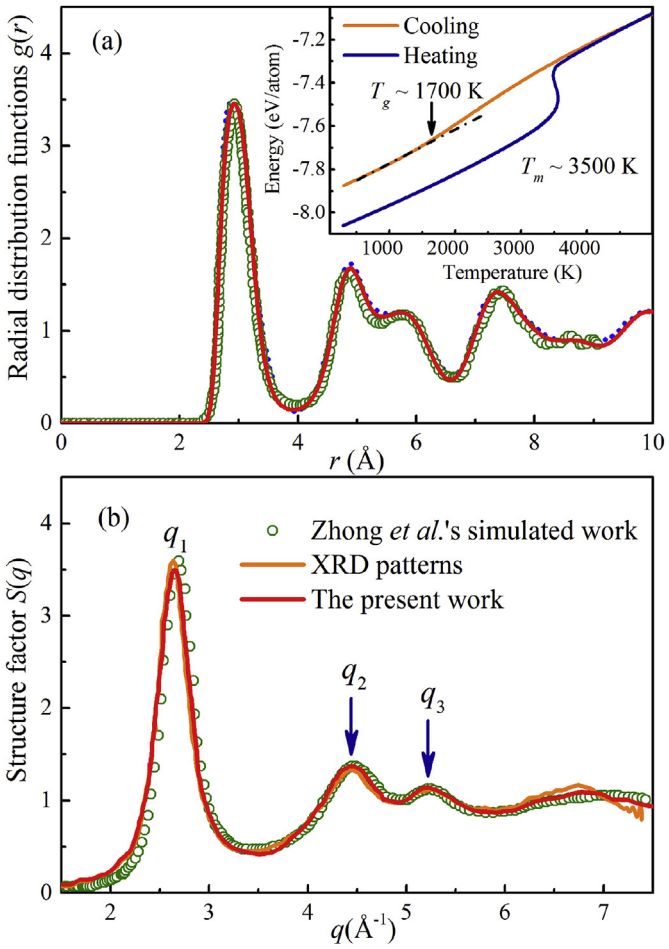

Fig. 1. (a) The radial distribution functions $g(r)$ of liquid melt quenched Tantalum (red line) obtained in the present work comparing with *ab initio* calculation (circles) [22] and simulated results of Jiang et al.'s work (blue dots) [23]. The inset shows the correlation of energy with temperature of the system upon heating up and cooling down. (b) The structure factor $S(q)$ of Ta MGs (red line) is similar to the ultrafast liquid quenching data (orange line) and Zhong et al.'s simulated results (circles) [24]. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

3. Results and discussion

To analyze the local atomic structure, the spatial topology of atomic arrangements has been investigated through Voronoi tessellations. From Fig. 2(a), one can see that the dominant coordination numbers (CNs) in the quenched Tantalum distribute over a wide range, and centralize from 12 to 14. The fractions of these atom clusters, accounting for more than 70% of all clusters in the quenched Tantalum, are presented in Fig. 2(b). It is found that the fractions of the predominant clusters <0,1,10,2>, <0,3,6,4>,