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The Role of Atomic Structures on the Oxygen Corrosion of Polycrystalline Copper Surface

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Abstract

The mechanical property of materials for pressure vessel, like steel, Ti, Cu and their alloys always turns out to be poor in the severely corrosive environment. The knowledge of oxygen corrosion on metal surface at atomic level is still lack. Using reactive molecular dynamic simulation, the oxygen corrosion behavior on polycrystalline copper is studied at the early stage of oxidation. Results indicate a higher reactivity at the grain boundary. The preferential dissociation of oxygen molecules at grain boundary is ascribed to the diffusion-related trapping effect and dissociation barriers. In addition, the difference of oxygen corrosion between grain boundary and grain on copper surface is elucidated in terms of the atomic-structure-related radial distribution functions. This study directly shows us the origin of intergranular oxygen corrosion and provides us useful information for the corrosion prevention, especially in the situation that the atomic structure changes under the thermal or mechanical loadings.

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Keywords: Molecular dynamic simulation; Grain boundary; Oxygen corrosion; Pressure vessel

1. Introduction

The reliability of pressure vessel has been a hot topic due to its fundamental importance in the industrial safety. Almost all the pressure vessels are required to operate under thermal and mechanical loadings. Thus, fatigue or creep induced damage attracts great attention in the assessment of failure of pressure vessels. A large number of investigations have been done corning on the effect of fatigue or creep on the crack initiation and growth [1-3].

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Some existing studies show that crack growth rate is promoted in the severely corrosive environment [4,5]. Such enhancement is attributed to the decrease of mechanical property caused by chemical damage, e.g. oxygen corrosion [6] and hydrogen embrittlement [7]. The corrosive damage always originates from grain boundary on the surface [8]. Then it is followed by intergranular corrosion [9]. This corrosion behavior is very common. However, the origin of intergranular corrosion still remains unclear at the atomic level.

The corrosion behavior is related to the chemical composition of medium or alloy [10], microstructure of alloy [11], etc. In this work, we study the early stage of oxygen corrosion on polycrystalline metal surface using reactive molecular dynamic simulation. Materials for pressure vessel include steel, Ti, Cu and their alloys [12-15], etc. Herein, we choose copper for the simulation. We show the diffusion and dissociation behaviors of oxygen molecules are distinct at grain boundary where the atomic structure is much different from grain on the substrate surface. The study of structure-depended reactivity provides us useful information for the corrosion prevention, especially in the situation that the atomic structure changes under the mechanical loadings.

2. Computational details

A simulation box of polycrystalline copper was constructed with the domain size of 60 × 50 × 28 Å. The thickness of copper substrate was 8 Å, with two 10 Å vacuum slab on each side of the substrate. The polycrystalline copper structure shown in Fig. 1(a) was generated using Voronoi tessellation which has been widely used in computational geometry [16]. The grain boundary is almost marked with a green color. All the simulations were done using the reactive force field (ReaxFF) developed by van Duin et al [17]. The ReaxFF integrated in LAMMPS code was chose due to its capability to describe chemical reactions. And it has been applied successfully in the molecular dynamics simulation of the initial oxidation in the interaction systems such as Cu/O [18], Ni/H2O [19], Fe/O2 [20]. Prior to the start of simulations, the polycrystalline structure was relaxed with 1 picoseconds of equilibrating at 300 K and 3 picoseconds of equilibrating at 800 K. We performed MD simulations of oxidation behavior on copper surface by oxygen molecules with the oxygen exposure time about 200 ps at 650 K. In the whole simulation process, we kept the number of oxygen molecules constant to prevent the high pressure in the vacuum layer. All the simulations were carried out using Canonical (NVT) ensemble, with Nose-Hoover thermostat technique employed for temperature control. The velocities and positions were updated with a time step of 0.1 fs. Periodic boundary conditions were used in x, y directions and fixed boundary conditions were imposed along z direction. In addition, reflecting boundary conditions were added to avoid losing atoms in the upper limit of simulation boxes.

3. Results and discussion

In Fig. 1, snapshots from MD simulations of the oxidation reaction between oxygen and polycrystalline copper are shown. Adsorption, diffusion and dissociation of oxygen molecules on copper substrate from experimental findings [21,22] are observed in our simulations. At ~60 ps, the dissociation process occurs initially at grain boundary with a random distribution of adsorbed oxygen molecules on surface shown in Fig. 1(b). The dissociated oxygen atoms are indicated with white arrows. However, in this period, no sign for dissociation behavior is observed at other sites. Subsequently, continuous incorporation of dissociated oxygen atoms is present at grain boundary, as shown in Fig. 1(c). The accumulation of oxygen atoms at grain boundary is indicated with white circle in Fig. 1(d). It is interesting to note that, at the end of simulations, the distribution of dissociated atoms is not uniform on the substrate surface and approximate 95 percent stays along the grain boundary. The dissociation behavior of oxygen molecules appears much more active at grain boundary. Besides, it is interesting to find that oxygen molecules (marked with white rectangular box) can be easily captured by intergranular atoms as they move across the grain boundary. This indicates a poor diffusibility of oxygen at the grain interface. Thus, the difference in diffusion and dissociation energy of oxygen molecules on substrate surface needs to be addressed.

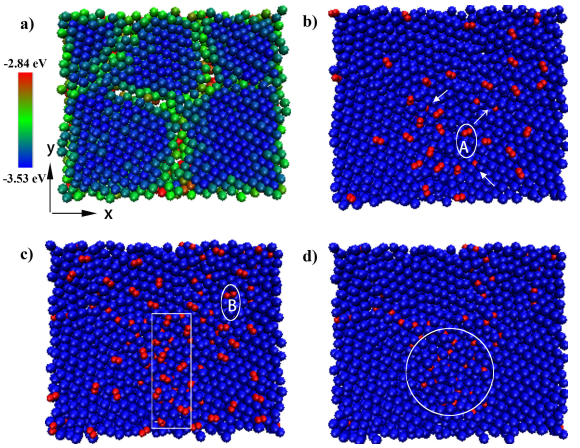

Fig. 1. Snapshots. (a) Identified grain and grain boundary colored by atomic potential energy. (b), (c) Top view of oxidized polycrystalline copper at about 60 ps and 200 ps. (d) Top view of snapshot (c) after removing adsorbed oxygen molecules, and clearly the O2 dissociation occurs along the grain boundary.

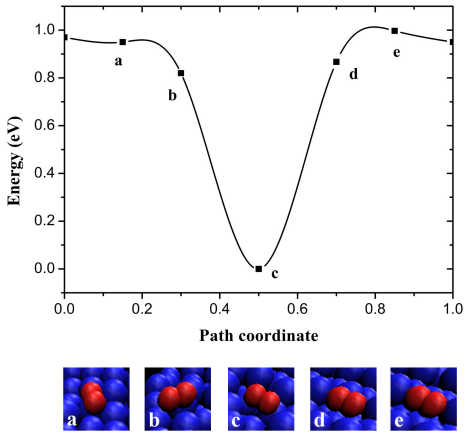

Fig. 2. The diffusion energy of single oxygen molecule across the grain boundary with certain sites labeled as (a)-(e) in bottom panels. A reference value of system energy selected as the oxygen molecule adsorbed at grain boundary (c). O2 is about 3 Å from the grain boundary at (b) and (d); 4-5 Å from the grain boundary at (a) and (e).

Before the determination of diffusion energy, we need to examine the adsorption energy of oxygen molecule on copper substrate. Yuan et al. [23] reported that the O2 adsorption energies for the most stable configurations on Cu clusters are from 0.7 eV to 2 eV calculated by first principles. Zhao et al. [24] calculated that adsorption energies of O2 on single crystal copper are between 0.14 eV and 3.58 eV. In terms of the existing calculation method [23], our calculated O2 adsorption energies on grain surface [Fig. 2(d)] and at grain interface [Fig. 2(c)] are about 2.14 eV and 3.02eV, respectively. Good agreement between our calculation and the existing calculations is achieved accordingly. The diffusion energy is generally determined by the change of system potential energy in the diffusion path [25]. Accordingly, we calculate the system potential energies with a single oxygen molecule adsorbed at or near grain interface along the certain diffusion path. The active energies for diffusion of oxygen molecule across the grain boundary are obtained shown in Fig. 2. The results indicate that the O2 diffusion barriers are < 0.13 eV on

grain surface. However, at the grain interface the energy barrier is relatively very high with about 0.9 eV. In this case, it is inferred that huge potential wells exist along the grain boundary. And the oxygen molecule can hardly escape once it gets into those potential wells. Such trapping effect could enhance the probability of O2 dissociation at grain boundary by preventing oxygen molecules moving outward.

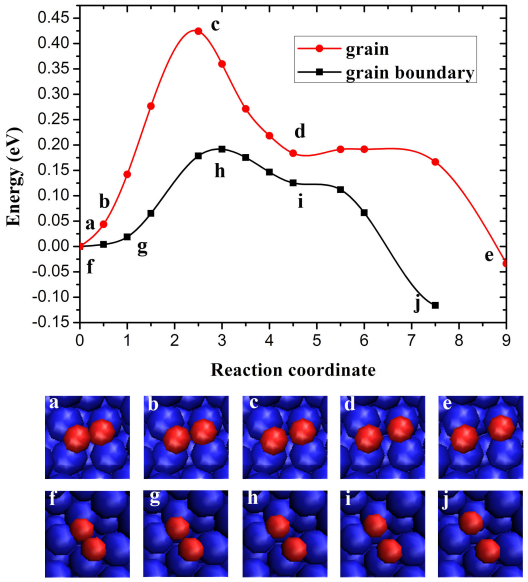

Fig. 3. Energy pathway for a oxygen molecule dissociation on grain surface and at grain boundary. Typical configurations sampled along the pathway [(a)-(e) for grain surface; (f-j) for grain boundary] are shown in bottom panels. The system energy at the initial state [(a) and (f)] is taken as a reference value.

Next, the dissociation process of a single oxygen molecule at grain boundary and on grain surface is examined, labeled with A and B in Fig. 1(b) and Fig. 1(c) respectively. Fig. 3 presents the relevant energy barrier and energy path of a single O2 dissociation. In the initial state [Fig. 3(a), (f)], the O-O bond length is nearly the same and reaches about 1.73 Å in both cases. As the oxygen molecule moves to the intermediate precursor state [Fig. 3(b), (g)], the energy increases slightly. Then, in the transition state [Fig. 3(c), (h)], both the O-O distances are about 2.06 Å. For B, the system energy increases sharply to 0.42 eV which is higher than the initial configuration, indicating a barrier of 0.42 eV for single oxygen molecule dissociation. For A, the dissociation barrier is much smaller with 0.19 eV. The calculated dissociation barriers of oxygen molecule are in good agreement with the existing theoretical studies [24,26](0.1 eV ~ 1 eV). After dissociation, the O-O bond is broken and the system energy decreases gradually. This indicates that oxygen molecule is more likely to dissociate at the grain boundary with lower active energy barrier. Compared with the previously calculated O2 diffusion energy of 0.13 eV on grain surface, the O2 dissociation energy is relatively high. On the contrary, the O2 dissociation is much more favorable with higher diffusion barrier at the grain boundary. Thus, there are only a few oxygen molecules dissociating at sites on grain surface. In addition, we find that the substrate atoms become a little disordered at the site where the dissociation occurs for B on grain surface. It seems that the atom structure has an effect on O2 dissociation.

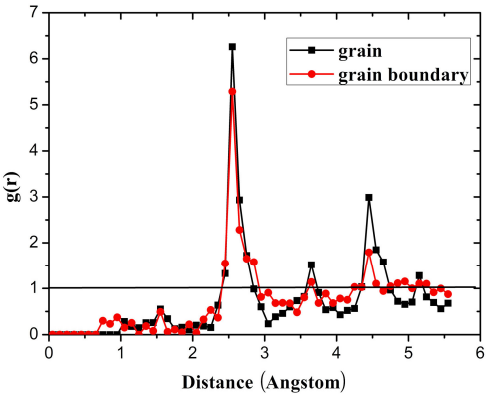

Fig. 4. Radial distribution function obtained for grain interior and grain boundary

It has been widely recognized that the arrangement of atoms is much different at grain boundary. And we further study the atomic structure and energy to uncover distinct diffusion and dissociation behaviors between grain interior and grain boundary. Fig. 4 shows the radial distribution functions computed for those two various structures. The RDF is defined as the possibility of finding an atom at a distance r from another atom compare to a homogeneous distribution. Compared with the case of grain structure, the intensity of main peak is relatively lower, and other peaks tend to disappear. This suggests that the atomic structure is incompact, and highly unordered. Such structure is very common in the real grain boundary induced by the existing of voids, dislocations and other defects at grain boundary [27,28]. In this structure, most intergranular atoms have fewer neighboring atoms around. Owing to fewer binding bonds, the potential energy of intergranular atoms ranged from -3.28 eV to -2.84 eV is higher than the intragranular atom energy ranged between -3.53 eV and -3.19 eV shown in Fig. 1(a). Therefore, the grain boundary with high reactivity provides more active sites for the dissociated adsorption of oxygen molecules. Furthermore, the atomic structure could play an important role in surface reactivity.

4. Conclusion

The nucleation of oxide preferentially occurs at grain boundary has been observed using reactive molecular dynamic simulation. We have also found that the enhanced dissociation of oxygen molecules is contributed to a trapping effect and lower dissociation barriers at the grain boundary. The distinct diffusion and dissociation behaviors are supposed to have a close correlation with various atomic structures and structure-related atom energy between grain and grain boundary. This result presents an atomic insight into the preferential oxygen corrosion at grain boundary. It provides useful information for the corrosion prevention as the atomic structure changes under the mechanical loadings.

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