Phase-dependent mechanical properties of two-dimensional silica films: A molecular dynamics study

Jin Zhang

Shenzhen Graduate School, Harbin Institute of Technology, Shenzhen 518055, China

ARTICLE INFO

ABSTRACT

The recently discovered two-dimensional (2D) silica films can be grown in both crystalline phase and vitreous phase. Using molecular dynamics simulations, we show that these two phases of 2D silica possess vastly different mechanical properties. It is shown that the Young's modulus of crystalline 2D silica is 54% greater than that of its vitreous counterpart. The vitreous 2D silica fails via the ductile fracture with serrated stress-strain curves, while its crystalline counterpart exhibits the abrupt brittle rupture. Both the fracture strength and fracture strain of crystalline 2D silica are significantly larger than those of its vitreous counterpart. Fracture mechanics theory is utilized to give some explanations to the different fracture properties of these two phases. Moreover, 2D silica mixed by crystalline phase and vitreous phase is also studied. Owing to the different mechanical properties of crystalline and vitreous phases, the elastic and fracture properties of this mixed-phase 2D silica are strongly dependent on its crystallinity. Specifically, we find that the Young's modulus of the mixed-phase 2D silica, ranging between the values of its crystalline and vitreous counterparts, increases as the degree of crystallinity increases. Meanwhile, with the increase of the degree of crystallinity the fracture strain of the mixed-phase 2D silica is found to decrease, making its value smaller than that of the purely crystalline and vitreous 2D silica.

1. Introduction

As one of the most abundant substances on earth, silicon dioxide or silica (SiO_2) has received considerable attention due to its wide range of applications in mechanics, optics and electronics [1]. Recently, inspired by the discovery of some two-dimensional (2D) nanomaterials having unique properties significantly different to their bulk counterparts [2–6], many efforts have been made to fabricate single- or few-atom thick silica at the nanoscale. In 2005, Weissenrieder et al. [7] synthesised the first monolayer silica film on a Mo(1 1 2) single crystal. However, this monolayer silica does not have the correct stoichiometry, as it needs to be covalently bonded to Mo substrate, which makes it actually stoichiometry $SiO_{2.5}$. Five years later, a 2D bilayer silica film with the proper SiO_2 stoichiometry was grown on a Ru(0001) single crystal [8]. Since then, similar 2D silica structure was also observed on Pd(0 0 1) [9], Pt(1 1 1) [10], graphene [11] and so on [12,13]. This 2D bilayer silica material exists in both crystalline phase and vitreous phase [14–18], consisting of a continuous random network of corner-sharing SiO_4 tetrahedra. As an excellent insulator with a wide band gap [8,13], 2D silica is highly expected to provide opportunities for developing next-generation nanoelectronics and nanodevices.

Since the discovery of the novel 2D silica structure, a lot of research has been devoted to its material properties. For example, using first-principles calculations, Zhou et al. [19] and Gao et al. [20,21] have studied the structural and electronic properties of crystalline 2D silica. Their results reveal that these 2D silica structures have high stability and good flexibility comparable to the graphene. Moreover, in their theoretical studies 2D silica was also found to be an excellent insulator with a wide band gap. Meanwhile, the mechanical properties of 2D silica also have attracted great interest from the community due to the observation that the electronic properties of 2D silica can be efficiently tuned by applying the mechanical strain on it [20,22]. In the first-principles calculations conducted by Zhou et al. [19], Gao et al. [20,21] and Romdhane et al. [23], the in-plane stiffness and the bending stiffness of 2D silica were predicted to be extremely high when comparing to other 2D nanomaterials. Using *ab initio* simulations and first-principles calculations, Gao et al. [22] and Han and Hu [24] have theoretically predicted a negative Poisson's ratio in the 2D silica with certain crystal structure. Very recently, using molecular dynamics (MD) simulations we [25] have reported a novel phase transformation in 2D silica under uniaxial compression, which can greatly affect the mechanical properties of 2D sil-

E-mail address: jinzhang@hit.edu.cn

ica. The efforts made so far have significantly expanded the knowledge of the mechanical properties of 2D silica. It is noted that aforementioned studies were all focused on the crystalline 2D silica. The mechanical properties of vitreous 2D silica, however, still remain unexplored. Considering the fact that 2D bilayer silica films can exist in both crystalline phase and vitreous phase [14–18], a comprehensive understanding of the mechanical properties for both crystalline and vitreous phases of 2D silica is crucial for the success of future applications of 2D silica.

Motivated by these ideas, this paper aims to conduct a comprehensive comparison study of the mechanical properties of crystalline and vitreous 2D silica films. The systematic study on this issue is of practical importance for the deep understanding of the mechanics of 2D silica but still remains unreported to date. In the present study, MD simulations are performed to measure Young's modulus, fracture strength and fracture strain of crystalline and vitreous 2D silica in the tensile test. The results show that crystalline 2D silica and vitreous 2D silica have very different elastic and fracture properties. In addition, efforts are also made to study the dependence of the mechanical properties of the mixed-phase 2D silica on its crystallinity.

2. Simulation method

The 2D bilayer silica films grown on metal or graphene substrate can possess both crystalline and vitreous phases [14–18]. Specifically, the crystalline 2D silica films are composed of corner-sharing SiO_4 tetrahedra, forming a hexagonal, honeycomb arrangement (see Fig. 1a), while their vitreous counterparts consist of a continuous random network of corner-sharing SiO_4 tetrahedra, resulting in arbitrary arrangement of various ring sizes ranging from four- to nine-membered rings (see Fig. 1b). In the present study, the crystalline 2D silica structure was created using the lattice constant calculated based on the first-principles [20], while the structure of the vitreous 2D silica was developed based on the coordinates measured from an atomically resolved scanning tunnelling microscopy (STM) image in Ref. [14]. After obtaining the molecular model of the crystalline and vitreous 2D silica films, we performed the tensile tests for these 2D silica structures to investigate their mechanical properties, which were implemented by MD simulations. In MD simulations, the interactions between Si—Si, Si—O and O—O were described by an effective interatomic potential developed by Vashishta et al. [26]. In this potential, the total interaction U combines a two-body term U_2 and a three-body term U_3, i.e.,

$$U = \sum_{i,j} U_2(r_{ij}) + \sum_{i,j,k} U_3(r_{ij}, r_{ik}), \qquad (1)$$

where r is the distance between atoms and i, j and k represent different atoms in the system. The expression of U_2 and U_3 in Eq. (1) is presented in the Supplementary Information. The values of the parameters in the potential shown in Eq. (1) were taken from Refs. [27,28], which are listed in Table S1 in the Supplementary Information. It is noted here that the interatomic interaction of Eq. (1) was initially developed for bulk and nanoscaled quartz (SiO_2) structures and recently has been proven to be reliable in characterizing the mechanical properties of 2D silica structures studied here [25].

In the present study, MD simulations were performed by employing large-scale atomic/molecular massively parallel simulator (LAMMPS) [29] with no periodic boundary condition applied in all directions. Our simulation was completed in the following procedure. First, the initial configuration of the 2D silica film was relaxed to its energy-minimized configuration by using the conjugate gradient method. Second, the initial locally energy-minimized structure of 2D silica was relaxed under room temperature (300 K) within the NVT ensemble (constant number of particles, volume and temperature) for 20 ps to reach its equilibrium state. To obtain the equilibrium thermodynamic state at this constant temperature, the Nosé-Hoover temperature thermostat was utilized [30]. In addition, the velocity Verlet algorithm with the time step of 0.5 fs was utilized to integrate the Hamiltonian equations of motion determined by Newton's second law. Third, a displacement control methodology was adopted to apply external tension to 2D silica. In doing this, atoms within a distance of 5 Å to one end of 2D silica were frozen, while atoms within the same distance to the opposite one end were moved by a relatively small incremental displacement, which is illustrated in Fig. S1 in the Supplementary Information. In this process, we chose a relatively low strain rate of 0.001
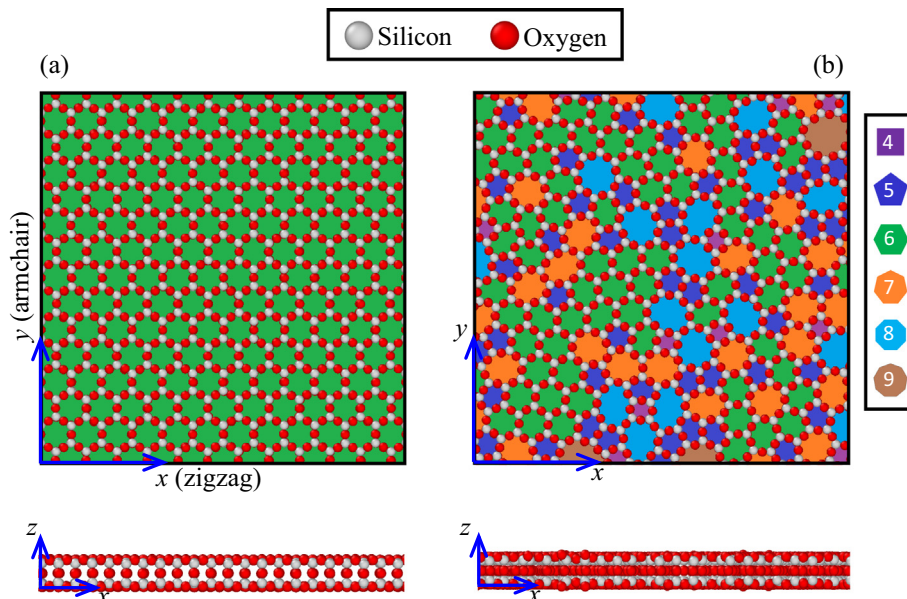

Fig. 1. Top (upper panel) and side (lower panel) views of (a) the crystalline 2D silica and (b) the vitreous 2D silica. Silicon and oxygen atoms are identified with grey and red balls, respectively; ring sizes are colour coded. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)