

## Molecular Dynamics Modeling of Mechanical Properties of Nanocrystalline SiC

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Molecular dynamics simulations using the Tersoff bond-order potential are employed to study the effects of temperature and grain size on mechanical properties of nanocrystalline silicon carbide. In this work, the simulated nanocrystalline SiC samples have a mean grain size varying from 2.5 to 5 nm and contain about  $10^5$  atoms in the model system. Tension tests with periodic boundary conditions and engineering strain rate of  $10^{-4}$  ps $^{-1}$  are simulated, which result in the stress-strain curves of the single- and nanocrystalline SiC in terms of the average virial stress and true strain. The elastic moduli of the single- and nanocrystalline silicon carbide are determined from fitting the stress-strain curves. In this work, the Young's modulus of nanocrystalline SiC is compared with those of the monocrystalline SiC for different temperatures in the range from 300 K to 3000 K. The numerical results show that the temperature has an obvious effect on Young's modulus, which is attributed to the large volume fraction of grain boundaries in nanocrystalline samples. With increasing temperature, the nanocrystalline SiC shows a brittle-to-ductile transition at temperatures above 600 K. In addition, the reduction in Young's modulus of the nanocrystalline SiC with increasing temperature exhibits a nonlinear trend. It is found that the plasticity of the nanocrystalline SiC samples sharply increases at temperatures above 2000 K. This effect was explained by a decrease in the melting point of the nanocrystalline materials in comparison to monocrystalline solids. The grain size dependence of elastic modulus of nanocrystalline SiC only becomes distinct at high temperatures and at a grain size greater than about 3 nm, while at room temperature elastic properties are almost invariant with the change of grain size. We expect that the quantifications of temperature and grain size dependence of mechanical properties will have implications in the development of nanocrystalline silicon carbide nanostructured materials for high performance structural applications.

**Keywords:** Silicon carbide, Nanocrystal, Strain, Molecular dynamics.

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### 1. INTRODUCTION

Recently, nanostructured materials are the basis for a whole range of industries, including microelectronics, optoelectronics, power engineering, military industry, etc. On the other hand, due to their unique electronic, optical, photo- and thermoelectric, magnetic, mechanical properties [1-5], nanostructured materials have also attracted considerable attention for modern material science.

Nowadays, nanomaterials that are used in extreme conditions at high temperatures, pressures and mechanical loads have become important. These materials include the structures based on silicon carbide. Silicon carbide has a set of unique physical and chemical properties, such as high hardness and mechanical stability at high temperatures, excellent thermal conductivity and low coefficient of thermal expansion, high resistance to corrosion and oxidation, wide bandgap and others [6]. SiC based materials have been widely used for a variety of applications in the automotive and aerospace industries, in high power electronic devices and bulletproof vests [7, 8]. Silicon carbide nanostructured ceramics and nanocomposites attract the greatest attention due to their unusual mechanical characteristics [9, 10]. In particular, it has been found experimentally that the hardness for nanosized SiC films with a grain size of 5-20 nm is higher than that for monocrystalline silicon carbide [11]. Using computer simulations it has been shown that nanoparticles of silicon carbide as inclusions in composite materials can significantly increase mechanical

strength [12]. However, the study of the SiC based materials is far from complete, and the physical mechanisms of changing their mechanical properties in a nanostructured state are not well understood.

An important tool for understanding the physical properties of nanomaterials at the atomic level is the computer modeling methods, among which an important role in modern physics and materials science plays the molecular dynamics. In particular, by using the molecular dynamics, the deformation process of SiC nanoparticles was investigated and the possibility of structural transformation in the process of their loading was shown [13]. The nonlinear dependence of elastic modules on the volume fraction of SiC in nanocomposites was revealed [14]. The mechanical properties of nanoscale silicon carbide with different microstructures were analyzed [15].

In this work, the molecular dynamics method has been used to study the mechanical properties of nanocrystalline silicon carbide (nc-SiC). The process of uniaxial tension of nanocrystalline silicon carbide with different sizes of nanocrystals is simulated in a wide temperature range. From the calculated stress-strain curves, the Young's modulus of the material is determined and its temperature and size dependencies are shown/plotted.

### 2. SIMULATION MODEL AND METHODS

Three-dimensional (3D) nanocrystalline SiC samples (Fig. 1) have been constructed by a computer code Atomsk [16] using the Voronoi tessellation method. Each 3D nanocrystalline sample is cubic in shape and consists

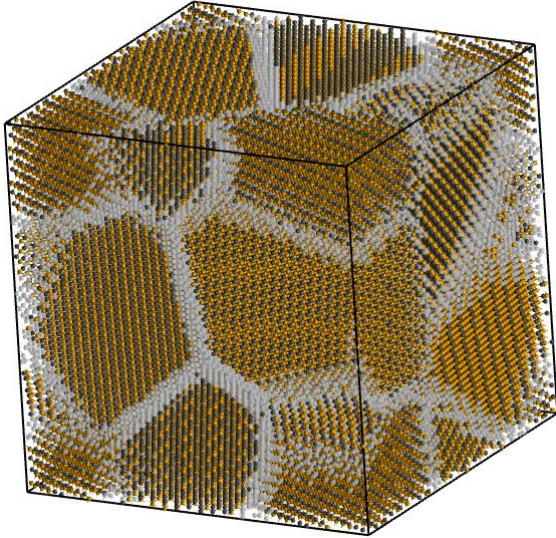
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of a certain number of grains. The mean grain size  $d$  is found to vary from 2.2 to 5 nm. In this study, molecular dynamics simulations of mechanical properties of the nanocrystalline SiC are performed using the Largescale Atomic/Molecular Massively Parallel Simulator (LAMMPS) open source code [17]. Periodic boundary conditions were applied in three dimensions to mimic the situation that is deep within the bulk of a larger sample. A timestep of 1 fs is used with the velocity-Verlet integration algorithm in order to provide a good compromise between efficiency and accuracy.

In this paper, the interactions between the Si-Si, C-C and Si-C atoms were simulated using Tersoff potential [18], which can accurately reproduce the lattice and elastic constants of SiC and is generally used in the molecular simulation community. The Tersoff potential computes a 3-body potential  $U$  for the energy of a system of SiC atoms as

$$U = \frac{1}{2} \sum_{j \neq i} f_C(r_{ij}) [f_R(r_{ij}) + b_{ij} f_A(r_{ij})], \quad (1)$$

where  $r_{ij}$  is a distance between the atoms  $i$  and  $j$ ,  $f_C$  is a smooth cutoff function,  $f_R$  is a repulsive two-body term, while  $f_A$  is an attractive three-body term managed by the  $b_{ij}$  bond function. The term  $b_{ij}$  is the strength of each bond, it depends on the local environment and is lowered when the number of neighbors is relatively high.



**Fig. 1** – An example of the nanocrystalline SiC structure. Atoms are colored according to a crystalline structure. Intra-granular phase, which is pristine diamond lattice, is represented by yellow (Si) and gray (C) atoms, whilst grain boundaries are mainly shown in white

As the first step of the molecular dynamics simulations, the atomistic model nc-SiC constructed above is equilibrated by minimizing the total energy of the SiC system, allowing unfavorable configurations in the grain boundaries to relax. After equilibration process, nanocrystalline samples are relaxed to equilibrium configurations at a given pressure of 0 bar by using a Nose-Hoover type equation of motion sampled from isothermal-isobaric (NPT) ensemble. Simulation times of 0.5 ns are found to be adequate for such relaxations.

To study the size and temperature dependence of the mechanical properties of nanocrystalline SiC, a series of tension simulations were performed. Uniaxial tensile deformation along z-axis was simulated at a strain rate of  $10^{-4}$  ps $^{-1}$  until the strain increases up to 20 %. Mechanical properties, such as Young's modulus, were calculated from engineering stress-strain curves. The average stresses in the atomistic nc-SiC systems were calculated using the virial theorem as follows:

$$\sigma_{ij} = - \left( \frac{\sum_{k=1}^N m_k v_{ki} v_{kj}}{V} + \frac{\sum_{k=1}^N r_{ki} f_{kj}}{V} \right), \quad (2)$$

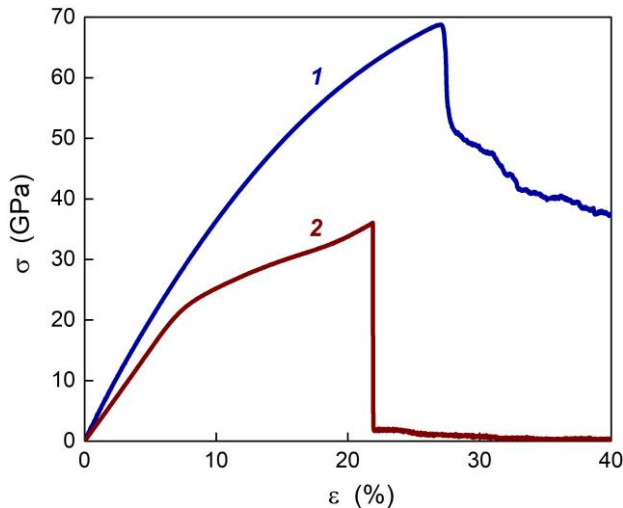
where  $N$  is the total number of atoms in structure,  $m_k$  is the mass of atom  $k$ ,  $v_{ki}$  is the  $i$ -th component of velocity vector of the atom  $k$ ,  $r_{ki}$  is the  $i$ -th component of position vectors of the atom  $k$ ,  $f_{ki}$  is the  $i$ -th component of interatomic force exerted on atom  $k$ , and  $V$  is the total volume.

### 3. RESULTS AND DISCUSSION

Reliable force fields are very important for molecular dynamics simulations of mechanical properties of nanocrystalline solids. Currently, there are many types of force fields available, which have been parameterized to describe a variety of multicomponent systems, including silicon carbide. In this paper, two potentials are considered to compare their performance for reproduction of mechanical properties of SiC. The first of them is the most widely used to study physical properties of solids with MD simulations Tersoff potential [18], and the second one is the Vashishta potential, recently parameterized for SiC system [19]. In order to begin examining these potentials we present a comparison of the mechanical properties predictions of Tersoff and Vashishta potentials for monocrystalline silicon carbide because their parameters are well known in the literature [20].

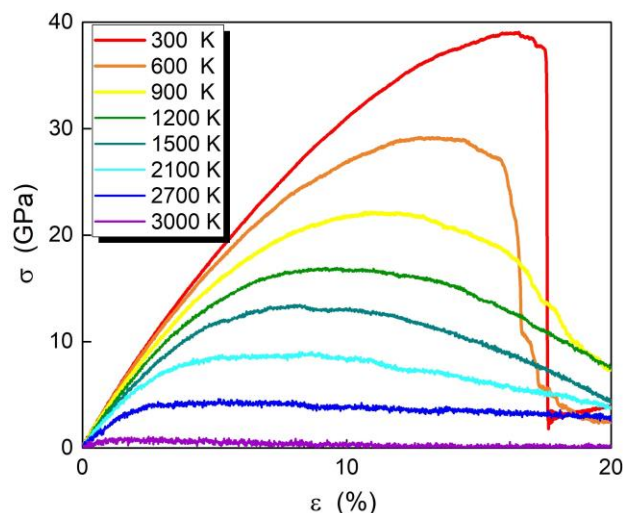
Fig. 2 depicts the typical strain-stress curves of a monocrystalline SiC in uniaxial tensile tests for the two considered potentials at 300 K. At the initial stage, the value of each stress-strain curve increases linearly but with different slopes. This behavior corresponds to the purely elastic region, when the mechanical stress  $\sigma$  and strain  $\varepsilon$  are in accordance with Hooke's law  $\sigma = E \varepsilon$ , where  $E$  is the Young's modulus. After elastic region both the Tersoff and Vashishta stress-strain curves display a plastic-elastic response up to a maximum stress which is called tensile strength, followed by an abrupt drop of stress, resembling the brittle fracture.

We estimate the Young's modulus  $E$  from the slope of a stress-strain  $\sigma(\varepsilon)$  curves, presented in Fig. 2, in the elastic deformation range. The value of  $E$  resulting from Tersoff calculations for monocrystalline SiC was found to be about  $E = 430$  GPa. On the other hand, the Vashishta potential gives the value  $E = 303$  GPa. Comparing these values with the experimental Young's modulus of SiC, which is found to be in the range of 392-694 GPa [20], we conclude that the Tersoff potential gives better agreement. Therefore, further research in our work was carried out only with the use of the Tersoff empirical potential.



**Fig. 2** – Computed stress-strain curves for uniaxial tension of single-crystal SiC for Tersoff (curve 1) and Vashishta (curve 2) potentials

Fig. 3 shows the calculated stress-strain dependencies  $\sigma(\epsilon)$  for the uniaxial tensile process of nanocrystalline silicon carbide with a mean grain size  $d \approx 2.5$  nm at various temperatures. It is noticeable that an increase in temperature leads to a decrease in the slope of linear region of  $\sigma(\epsilon)$  curves, indicating a decrease in the Young's modulus of the material. In addition, an increase in temperature leads to a decrease in the strength of the structure and to the transformation of the general form of the stress-strain curve. The stress-strain curve at a temperature of 300 K has a sharp decline at the tensile strength point, which is a brittle failure. With the temperature increase to above 600 K, the stress shows a smooth curve before failure, which means a brittle-to-ductile transition. The plasticity of the nc-SiC sharply increases at temperatures above 2000 K. It should be noted that in the same temperature range of 300-3000 K the  $\sigma(\epsilon)$  curve for monocrystalline SiC has a constant shape, which is characteristic for brittle failure.

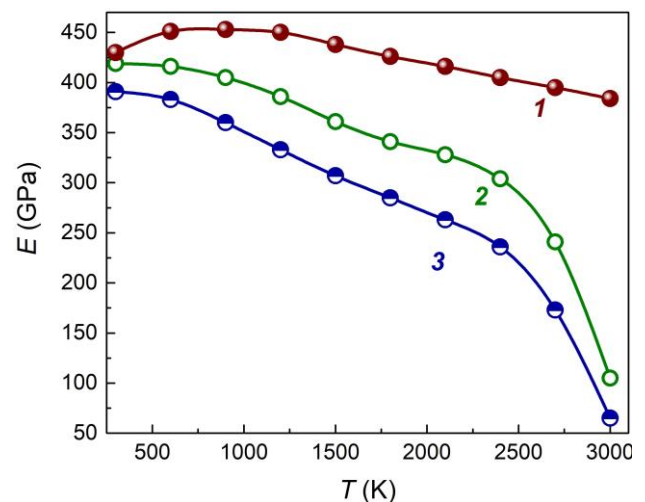


**Fig. 3** – Calculated stress-strain curves for nc-SiC with mean grain size of 2.5 nm at different temperatures from 300 K to 3000 K

From the obtained stress-strain curves (Fig. 3), we have calculated the Young's modulus  $E$  of the material in the temperature range of 300-3000 K. The corresponding dependencies  $E(T)$  are shown in Fig. 4 for mono- and nanocrystalline silicon carbide. The Young's modulus of monocrystalline SiC decreases linearly as the temperature increases from 300 to 3000 K due to the softness of the material. It is seen that when the temperature increases tenfold (from 300 K to 3000 K), the Young's modulus  $E$  of SiC varies from approximately 450 to 400 GPa, i.e., the relative change  $\Delta E$  is about 11 %. For nc-SiC, the dependence  $E(T)$  becomes substantially nonlinear with increasing temperature, and the decrease in the Young's modulus with temperature is much faster. In the temperature range from 300 K to 3000 K, the relative change in the Young's modulus is  $\Delta E \approx 83$  % for nc-SiC with a mean grain size  $d \approx 2.5$  nm and  $\Delta E \approx 75$  % for the structure with  $d \approx 5$  nm.

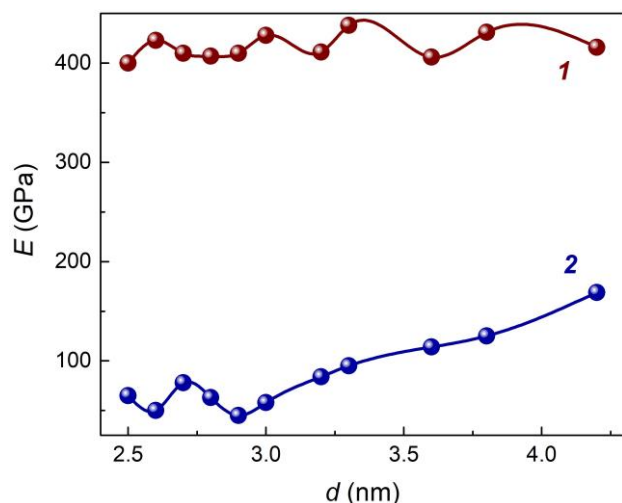
An increase in the plasticity of nc-SiC with temperature increase can be explained by the presence in the structure of a significant volume fraction of disordered atoms on the grain boundaries. These atoms have higher energy in comparison with atoms in the volume of nanocrystals, making it easier to form torn bonds when heated nanocrystalline sample, which causes plasticization of the material. The change in the slope of the  $E(T)$  curve in the temperature range above 2000 K is associated with a decrease in the melting point of the nanocrystalline materials. It is known that for a monocrystalline SiC  $T_m = 3103$  K [20], while for nc-SiC the value of  $T_m$  varies inversely with respect to the mean grain size. As a result, significant plasticization of nanocrystalline material will occur at lower temperatures than for a monocrystalline SiC.

In the next step, we investigated the effect of the grain size of nanocrystals on the value of Young's modulus of nc-SiC. For this purpose, we generated 11 model cells of the same volume  $10 \times 10 \times 10$  nm<sup>3</sup> with different number of nanocrystalline grains in the range of sizes  $d = 2.5$ -5 nm. For each structure, we calculated the  $\sigma(\epsilon)$  curves at two boundary temperatures of 300 K and 3000 K. From the obtained stress-strain curves, the



**Fig. 4** – Temperature dependence of the Young's modulus of bulk SiC (curve 1) and nanocrystalline SiC with mean grain size  $d \approx 5$  nm (curve 2) and  $d \approx 2.5$  nm (curve 3)





**Fig. 5** – Size dependence of the Young's modulus for nanocrystalline SiC samples at temperatures  $T = 300$  K (curve 1) and  $T = 3000$  K (curve 2)

Young's modulus  $E$  for structures with different grain sizes  $d$  was calculated. The corresponding dependencies  $E(d)$  are shown in Fig. 5.

As seen, at room temperature the Young's modulus of the nc-SiC does not show a clear dependence on  $d$  for the studied range of mean grain sizes of nanocrystals (curve 1 in Fig. 5). The value of Young's modulus oscillates within 400-440 GPa and is close to the corresponding Young's modulus of monocrystalline SiC. At

high temperatures (curve 2 in Fig. 5), the Young's modulus of the nc-SiC has a slightly different grain size dependence. In particular, in structures with a grain size less than 3 nm, as well as at room temperature, an oscillating dependence  $E(d)$  is observed. However, with a further increase in the grain size, there is a clear tendency to increase the Young's modulus. The increase in the Young's modulus with increasing  $d$  is due to the fact that an increase in the mean grain size in the structure results in a decrease in the volume fraction of disordered atoms localized on the boundary of the grains. As a result, the plasticity of the material decreases with a simultaneous increase in  $E$ .

#### 4. CONCLUSIONS

In conclusion, in this paper, the stress-strain curves for uniaxial tension of nanocrystalline silicon carbide in the temperature range of 300-3000 K were calculated. It is shown that with increasing temperature there is a brittle-to-ductile transition. It was found that in the temperature range over 2000 K, the Young's modulus of the nc-SiC significantly decreases, resulting in softening the material. It is shown that at room temperature the Young's modulus of the nanocrystalline SiC almost does not change with an increase in the grain size from 2.5 to 5 nm, whereas at  $T = 3000$  K a monotonous increase in the magnitude  $E$  occurs when the grain size is larger than 3 nm.

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### Модельовання механічних властивостей нанокристалічного SiC методом молекулярної динаміки

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Методом молекулярної динаміки з використанням потенціалу міжатомної взаємодії Tersoff досліджено вплив температури і розміру зерен на механічні властивості нанокристалічного карбиду кремнію. Середні розміри зерен модельованих зразків нанокристалічного SiC становили 2.5-5 нм, а кількість атомів у структурі досягала близько  $10^5$ . Для досліджуваних структур розраховувались криві напруження-деформація в результаті моделювання одновісного розтягу зразків зі швидкістю деформації  $10^{-4}$  пс $^{-1}$ . Шляхом лінійної апроксимації початкової ділянки кривих напруження-деформація було розраховано пружні модулі для моно- і нанокристалічного карбиду кремнію. Проведено порівняння величини модуля Юнга для моно- і нанокристалічного SiC в інтервалі температур 300-3000 К.

Результати розрахунку показують, що модуль Юнга нанокристалічних зразків суттєво змінюється з ростом температури, що пояснено суттєвою часткою атомів на границях зерен в нанокристалічному SiC. Встановлено, що з ростом температури вище 600 K відбувається перехід від крихкого до в'язкого руйнування нанокристалічних зразків. Крім того, показано, що зменшення модуля Юнга нанокристалічного карбиду кремнію з ростом температури має нелінійний характер. Виявлено, що при підвищенні температури понад 2000 K, пластичність нанокристалічного SiC різко збільшується. Вказаний ефект пов'язується зі зменшенням температури плавлення нанокристалічних зразків у порівнянні з монокристалом SiC. Показано, що вплив розміру зерен на модуль пружності нанокристалічного SiC має місце лише при високих температурах і розмірах зерен вище 3 нм, тоді як при кімнатній температурі пружні властивості не залежать від розмірів нанокристалів. Передбачається, що отримані результати з температурними та розмірними залежностями механічних характеристик нанокристалічного карбиду кремнію можуть бути використані при розробці наноструктурованих матеріалів на його основі для високоефективних конструкційних застосувань.

**Ключові слова:** Карбід кремнію, Нанокристал, Деформація, Молекулярна динаміка.