ORAL/POSTER REFERENCE: ICF100514OR

FIRST-PRINCIPLE AND CLASSICAL MOLECULAR DYNAMICS STUDY ON TENSILE AND SHEAR STRENGTH OF SILICON NITRIDE

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ABSTRACT

The intrinsic tensile and shear strength of β- type silicon nitride were determined, using a first-principle molecular dynamics(FPMD) technique based on density functional theory. The results of this simulation were compared with the results obtained from a classical MD simulation, using a 3-body interatomic potential for silicon nitride. The agreement was satisfactory. The classical MD technique, based on the 3-body interatomic potential, was used to perform several simulations of crack propagation, slip and dislocation motion in α- and β- type silicon nitride. From the simulations an estimate of the critical stress intensity factor, the slip planes and directions, and the atomic structure of the dislocation were determined.

KEYWORDS

silicon nitride, tensile strength, shear strength, crack, stress intensity factor, slip plane, dislocation, classical molecular dynamics, first-principle molecular dynamics

INTRODUCTION

Currently, silicon nitride(Si_3N_4) is a very important material because of the inherent superior mechanical properties of the material. In general, Si_3N_4 is classed as polycrystalline, which means that the material properties of Si_3N_4 are determined by studying the single crystal and grain boundaries of Si_3N_4. However, existing experimental studies have used the polycrystalline material for experimental work since large single crystals are difficult to make. Hence, numerical methods are very important for studying the properties of the single crystal. In this study, the mechanical properties of Si_3N_4 are determined by specifically modeling the mechanical behavior of the single crystal.

Ching *et al.*[1] and Belkada *et al.*[2] have calculated the elastic constants, lattice constants and atomic structure of Si_3N_4 single crystals using first principle(FP) calculations based on density functional theory. Wendel and Goddard[3] did the same by using the method of linear combination of atomic orbitals (LCAO). Vashishta *et al.*[4] reported large-scale simulations of a sintering process and the mechanical behavior of a nanocrystal and amorphous Si_3N_4 using the classical MD method, introducing a 3-body interatomic potential for the materials in question.

In this paper, the intrinsic tensile and shear strength of β-Si_3N_4 are obtained using the FPMD simulation technique based on density functional theory. In addition, the slip planes and crack propagation of α- and β-Si_3N_4 are investigated using the classical MD simulation method, with the 3-body interatomic potential proposed by Vashishta *et al.*[5].

INTRINSIC TENSILE AND SHEAR STRENGTH OF β-Si_3N_4

In the FP calculation, a plane wave representation was used to obtain a solution of the Kohn-Sham equations. The local density approximation is adopted to describe the exchange-correlation energy as a function of the electron density. In addition, the pseudopotential approximation is used to estimate ion - electron interaction. The pseudopotentials are generated from the scheme of Troullier-Martins[6]. The pseudopotentials are given in the form used by Kleinman-Bylander[7] and the occupation of states is determined by the Fermi distribution function. Furthermore, the stress tensor is calculated from the explicit symmetric formulas proposed by Lee *et al.*[8].

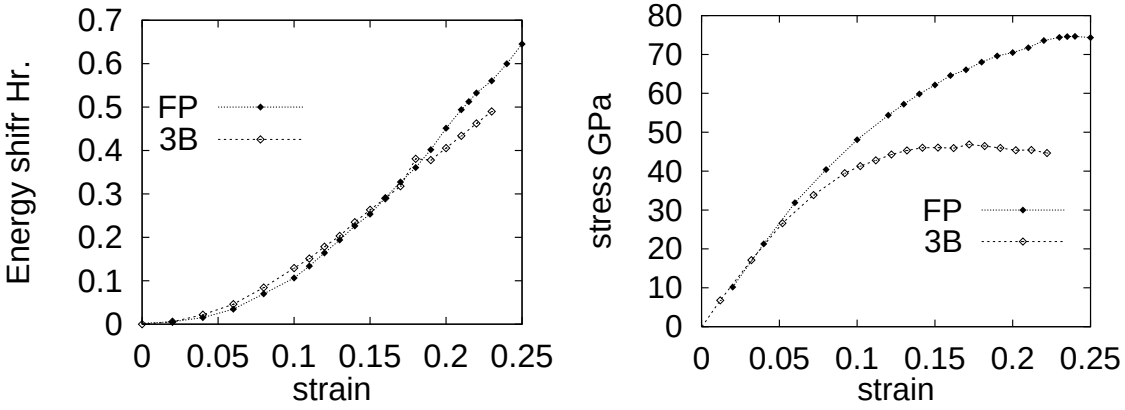
The conjugate gradient algorithm proposed by Bylander, Kleinman and Lee[9] is employed to achieve a self-consistent electronic ground state. Each relaxed atomic configuration is obtained by the simulated annealing process.

Tensile Strength

The simulation performed to estimate the tensile strength was carried out using a 28 atom periodically repeated supercell($7.95 \times 13.155 \times 2.902 \times 10^{-10}$m) with a plane wave cutoff energy of 2.6×10^{-16}J (60 Ry). Five k-points were used for the Brillouin zone sampling. Uniaxial tensile deformations were simulated by increasing the uniaxial tensile strains along the z-direction, while the other supercell dimensions were fixed. The atomic configuration was relaxed at each strain level and this configuration was then used as the initial configuration for the next stain level. The tensile axis is defined to be parallel to the c-axis direction of the β-Si_3N_4 which is predicted to be the strongest direction. Calculated energy-strain and stress-strain curves are shown in Figure 1. The ideal tensile strength in this direction is estimated as 74 GPa. To examine the validity of the 3-body potential for the investigation of tensile properties, the result obtained using the 3-body potential was compared with this first-principle calculation(also shown in Figure 1). The energy values obtained from the 3-body potential are in good agreement with the values obtained from the FP calculation. However, calculated stress at large strain levels and the tensile strength are smaller.

Shear Strength

The supercell used in the calculation of the shear strength is shown in Figure 2. The x,y and z directions are defined as the <1000>, <01$\bar{1}$0> and <0001> directions of the β- crystal, respectively. This model contains 84 atoms, with the dimensions of the model



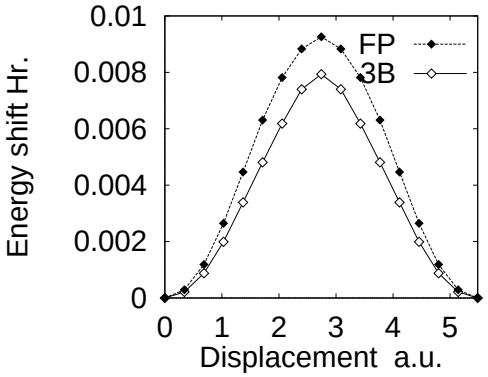
Energy-stain curve
(1Hr.=4.36×10^{-18} J)

Stress-strain curve

Figure 1: Energy-strain and stress-strain curve of β-Si_3N_4

shown in Figure 2. Shear deformation is simulated by means of a sliding procedure of the rigid center region as shown in Figure 2. The slip direction and plane orientation correspond to the primary slip system of β-Si_3N_4 proposed from experimental results. This was also confirmed using a MD simulation which will be discussed later.

For this simulation process, the relaxation of the atomic structure was not performed. The plane wave cut-off energy of 1.6×10^{-16}J (36 Ry) and four k-points are used for the FP calculation. The energy shift from a perfect crystal are estimated from the FP calculation and the 3-body potential (Figure 2). The results obtained from the 3-body potential are in good agreement with that of the first principle method. The ideal shear strength was estimated from the atomic force as approximately 22 GPa using the first principle method, and approximately 28 GPa using the 3-body potential. It is concluded that the 3-body interatomic potential is valid for the modeling of the shearing process.



Model super cell

Energy-displacement curve
(1Hr.=4.36×10^{-18} J)
(1 a.u.=0.529×10^{-10} m)

Figure 2: Model super cell and energy curve of first principle shearing simulation

Four types of shear deformation are modeled, to examine the properties of the slip system, which are, α-$\{1\bar{1}01\}<11\bar{2}0>$ (Case 1), α-$\{10\bar{1}0\}<0001>$ (Case 2), β-$\{10\bar{1}0\}<0001>$ (Case 3), and β-$\{10\bar{1}0\}<1\bar{2}10>$ (Case 4). Cases 1 and 2 correspond to the possible primary slip systems which were reported by Suematsu *et al.*[10], and Niihara and Hirai [11], respectively. Case 3 corresponds to the primary slip system which was reported by Evans *et al.*[12], Kawahara *et al.*[13] and Milhet *et al.*[14]. Finally Case 4 corresponds to the secondary slip system reported by Kawahara *et al.*[13].

Snapshots of the atomic arrangement in the model under shear deformation are shown in Figure 3. The sliding directions are from left to right of the figures. The sliding speed was 10 m/s. The black and gray circles represent nitrogen and silicon atoms, respectively. Only those atoms which have a coordination number that differs from that of the perfect crystal are shown.

The shear stress was calculated as the sum of all the atomic forces of the fixed atoms along the shear direction divided by the initial area of the slip plane. The shear strength is estimated from the maximum shear stresses as 25 GPa(Case 1), 20 GPa(Case 2), 18.5 GPa(Case 3), and 17 GPa(Case 4).

In Case 1, no plastic deformation in the sliding direction was observed (Figure 3). However, Suematsu *et al.* [10] asserted that this direction is one of the possible slip systems from Knoop hardness testing. This discrepancy may be due to an effect of hydrostatic stress produced by Knoop hardness testing. However, slip deformation in Case 2, Case 3, and Case 4 was observed. In particular in Case 3 slip progresses by dislocation movement. The width of the dislocation is estimated to be approximately 8Å.

Case 1 Case2

Case 3 Case4

Figure 3: Atomic arrangements under shear deformation

CLASSICAL MD CRACK PROPAGATION IN α AND β-Si_3N_4

The MD crack propagation analysis was performed to determine the critical stress intensity factor. Cubic cells in which the a and c axes of α- and β-Si_3N_4 are along the x and z directions, respectively, were used. The cells contain approximately 10,000 atoms and the dimensions for the x, y and z axis were approximately 9 nm, 9 nm and 1.2 nm, respectively. Initially, a crack was inserted and the crack front was placed along the z axes. The initial atomic displacements were taken from the mode I linear elastic solutions corresponding to a stress intensity factor K_I. Following this the atomic configurations was relaxed for 1.2 ps, except for the outermost atoms within 0.6 nm of the cell boundaries. If the crack tip did not move after the relaxation process is complete, the critical stress intensity factor K_{Ic} can be estimated from K_I. Four cases were calculated, i.e. for the crack tip in the prism and basal plane of each α- and β- crystal. The results are listed in Table 1. Reimanis *et al.*[15] estimated the K_{Ic} for α- crystal as 1.5$\sim$2.0 MPa$\sqrt{\text{m}}$ from the Knoop tests and for β- as 1.3$\sim$1.4 MPa$\sqrt{\text{m}}$ from the theoretically calculated bond energy. The results presented in this paper are in good agreement with the estimations of Reimanis *et al.* However, it is possible that our estimated of K_{Ic} is somewhat smaller than the true K_{Ic} value because of the under estimation of stress and ideal strength by the 3-body potential.

Table 1: Critical stress intensity factors of silicon nitride

crystal type	crack plane	crack front	K_{Ic} MPa$\sqrt{\text{m}}$
α	(0001)	$[01\bar{1}0]$	1.5
α	$(01\bar{1}0)$	[0001]	1.3
β	(0001)	$[01\bar{1}0]$	1.7
β	$(01\bar{1}0)$	[0001]	1.4

CONCLUSION

The intrinsic tensile and shear strength of β-Si_3N_4 was found to be 74GPa and 28GPa, respectively. It is found that the 3-body potential used to calculate results for Si_3N_4 are of an acceptable accuracy. Furthermore, the primary slip system of α- and β- Si_3N_4 was determined as $\{10\bar{1}0\}<0001>$. The critical stress intensity factors for several mode I cracks were also calculated and are in good agreement with the data reported by Reimanis *et al.*

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