Transactions of the Materials Research Society of Japan 24 [2] 257-260 (1999)

Molecular Dynamics Simulation of Crack Propagation in β-Silicon Nitride

Naoto HIROSAKI, Shigenobu OGATA*, and Hiroshi KITAGAWA*

National Institute for Research in Inorganic Materials,
1-1, Namiki, Tsukuba-shi, Ibaraki, 305-0044, Japan
Fax: +81-298-52-7449, e-mail:hirosaki@nirim.go.jp
** Department of Adaptive Machine Systems, Osaka University,*
2-1, Yamadaoka, Suita-shi, Osaka, 565-0871, Japan
Fax: +81-6-879-7246, e-mail:ogata@ams.eng.osaka-u.ac.jp, kitagawa@ams.eng.osaka-u.ac.jp

Abstract

Crack propagation in a β-silicon nitride crystal is investigated by molecular dynamics simulations using a three-body potential. A crack propagates when K_I is greater than 0.7 MPa$\sqrt{\text{m}}$, but shrinks when K_I is smaller than 0.65 MPa$\sqrt{\text{m}}$, indicating that the K_{IC} value is about 0.7 MPa$\sqrt{\text{m}}$. This value agrees well with that calculated using Griffith's theory. The stress distribution near the crack tip is calculated from the MD results, assuming that the stress is the average of the atomic stresses. The calculated stress distribution is in good agreement with the linear elastic solution.

Key Words: β-Si_3N_4, molecular dynamic, simulation, stress intensity factor, stress, linear elastic solution

1. Introduction

Silicon nitride ceramics have been investigated for their potential application as structural materials because of their excellent mechanical properties at room and elevated temperatures. However, this material has the disadvantage of being brittle and having low reliability. Polycrystalline silicon nitride ceramics are composed of β-Si_3N_4 grains and oxynitride grain boundary phases. The mechanical properties of silicon nitride ceramics are governed by the fracture of Si_3N_4 grains and of the grain boundary phases. Many studies have been carried out on fracture in polycrystalline silicon nitride ceramics, in which the fracture of the grain boundary phases is important. Few experimental studies of fracture in monocrystalline Si_3N_4 have been reported because it is difficult to obtain large single crystals. In this study, MD simulations are performed to examine crack propagation in a β-Si_3N_4 crystal.

2. Model and potential

In this report, we use the interatomic potential (V) proposed by Vashishta *et al.*[1], which includes both 2-body ($V_{ij}^{(2)}$) and 3-body ($V_{jik}^{(3)}$) interactions:

$$V = V_{ij}^{(2)}(r_{ij}) + V_{jik}^{(3)}(r_{ij,ik}) \tag{1}$$

where $V_{ij}^{(2)}(r_{ij})$ and $V_{jik}^{(3)}(r_{ij,ik})$ are:

$$\begin{aligned} V_{ij}^{(2)}(r_{ij}) &= A_{ij}\left(\frac{\sigma_i+\sigma_j}{r_{ij}}\right)^{\eta_{ij}} \\ &\quad + \frac{Z_iZ_j}{r_{ij}}\exp\left(-\frac{r_{ij}}{r_{s1}}\right) \\ &\quad - \frac{\alpha_iZ_j^2+\alpha_jZ_i^2}{2r_{ij}^4}\exp\left(-\frac{r_{ij}}{r_{s4}}\right) \end{aligned} \tag{2}$$

and

$$\begin{aligned} V_{jik}^{(3)}(r_{ij,ik}) &= B_{jik}\exp\left(\frac{l}{r_{ij}-r_c}+\frac{l}{r_{ij}-r_c}\right) \\ &\quad \times(\cos\theta_{jik}-\cos\theta_{jik}^0) \end{aligned} \tag{3}$$

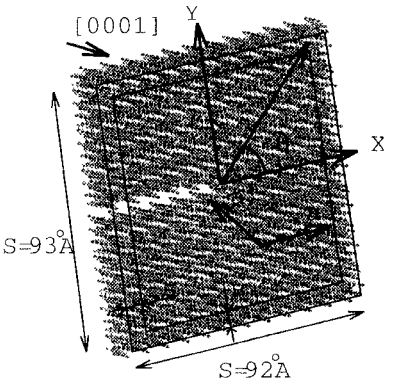


Figure 1. Schematic model of Mode I fracture.

where:

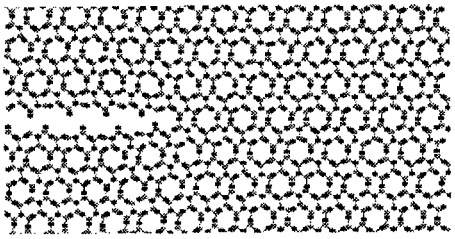
$$\cos\theta_{jik} = \frac{\vec{r}_{ik}\cdot\vec{r}_{ij}}{r_{ik}r_{ij}}. \tag{4}$$

The 2-body potential function consists of steric repulsion, Coulomb interaction, and charge dipole interaction caused by the large electronic polarizability of N^{3-}. The 3-body function consists of bond bending terms to take into account the covalent nature of the bonds. This potential has been applied to various MD calculations and has given excellent results for the equilibrium lattice parameter, elastic constants, phonon density of states, and specific heat when compared with the experimental data.[2),3)]

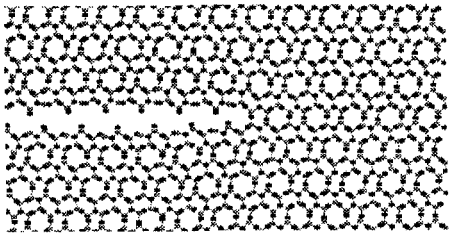
3. Crack Propagation of mode I

In the MD simulation, we used a cubic cell in which the a and c axes of β-Si_3N_4 were placed along the x and z directions, respectively, as illustrated in Figure 1.

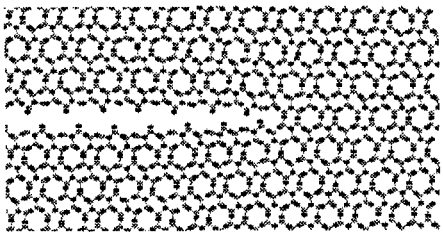
The basic cell contains 9408 atoms (Si:4032, N:5376). The atoms near the cell boundaries in the x



(a) K_I=0.65 MPa$\sqrt{\text{m}}$



(b) K_I=0.70 MPa$\sqrt{\text{m}}$



(c) K_I=0.80 MPa$\sqrt{\text{m}}$

Figure 2. Cracks in crystals with different values of K_I.

and y directions were confined within the cell, whereas those in the z direction were subjected to periodic boundary conditions. A crack of length $S/2$, or half the cell width, was inserted with the crack tip lying along the line x=0, y=0, i.e. the z axis. The atom displacements on introducing the crack were calculated using linear elastic theory assuming macroscopic isotropy. All the atoms were then relaxed at 100 K except for those within a width of $s/S = 0.06$ from the boundaries, which were held fixed.

Figure 2 shows the MD results after relaxation for 1.2 ps when the value of K_I was varied from 0.65 to 0.8 MPa $\sqrt{\text{m}}$. Although the crack length decreased in the case of $K_I = 0.65$ MPa $\sqrt{\text{m}}$, it began to grow for $K_I = 0.7$ MPa $\sqrt{\text{m}}$; the crack length increased with the K_I value. Assuming that the smallest K_I value at which a crack begins to propagate is the critical stress intensity factor, from the MD simulations, K_{IC}^{sim}, is estimated to be about 0.7 MPa$\sqrt{\text{m}}$.

Macroscopic fracture toughness was calculated using linear-elastic fracture mechanics to compare with the K_{IC}^{sim} value. The displacements for opening mode

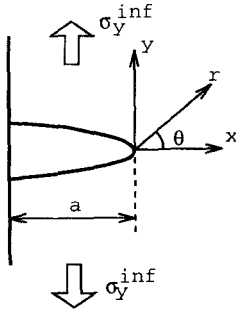


Figure 3. Crack tip

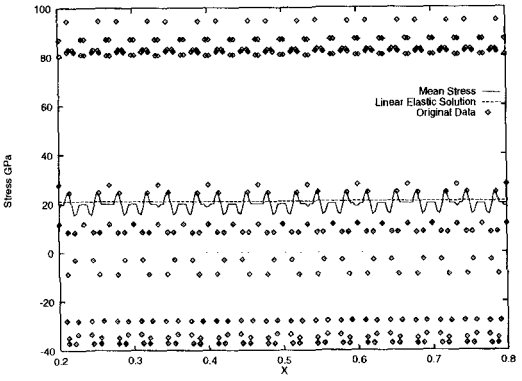


Figure 4. Stress distributions according to tensile model and MD calculation.

I are:

$$\begin{aligned} u &= \frac{K_I}{2G}\sqrt{\frac{r}{2\pi}}\cos\frac{\theta}{2}\left(K-1+2\sin^2\frac{\theta}{2}\right) \\ v &= \frac{K_I}{2G}\sqrt{\frac{r}{2\pi}}\sin\frac{\theta}{2}\left(K-1+2\cos^2\frac{\theta}{2}\right) \end{aligned} \tag{5}$$

where u and v are the displacements in the x and y directions, respectively, G is shear modulus, and K is bulk modulus. The Voigt model, where stress is constant, gives:

$$K = 3 - 4\nu \tag{6}$$

$$G = \frac{1}{30}(3C_{mpmp} - C_{mmpp}) \tag{7}$$

where:

$$\begin{aligned} C_{mpmp} &= (C_{11} + C_{22} + C_{33}) \\ &\quad +2(C_{23} + C_{31} + C_{12}) \\ C_{mmpp} &= (C_{11} + C_{22} + C_{33}) \\ &\quad +2(C_{44} + C_{55} + C_{66}). \end{aligned} \tag{8}$$

These equations indicate that K_I is related to the displacement near the crack tip through macroscopic values, namely the elastic constants and Poisson's ratio, ν. Table I shows values for these constants calculated by differentiating the interatomic potential with

respect to strain. The *ab-initio* values calculated by Ching *et al.*[4] are also shown for comparison with our calculation.

On the $y = 0$ plane in Figure 3, at crack tip:

$$\sigma_{xx} \simeq \sigma_{yy} \simeq \frac{\sigma_y^{inf}\sqrt{a}}{\sqrt{2r}} \tag{9}$$

When $\theta = 0$:

$$\sigma_{xx} = \frac{K_I}{\sqrt{2\pi r}}. \tag{10}$$

Substituting (9) into (10) gives us

$$K_I = \sigma_y^{inf}\sqrt{\pi a}. \tag{11}$$

Next, K_{IC} is estimated using Griffith's energy balance:

$$\begin{aligned}\sigma_c &= \sqrt{\frac{2E\gamma}{\pi a}} \\ &= \sqrt{\frac{4G(1+\nu)\gamma}{\pi a}}\end{aligned} \tag{12}$$

where γ is fracture energy, and σ_c is the critical stress at which crack begins to propagate. Hence:

$$K_{IC} = 2\sqrt{G(1+\nu)\gamma}. \tag{13}$$

Using the elastic constants shown in Table 1 and γ = 0.92 J/m^2 calculated from our MD simulation, the Griffith theory value of K_{IC} is calculated to be 0.89 MPa$\sqrt{\text{m}}$. This value is in good agreement with K_{IC}^{sim}, but somewhat lower than the experimental K_{IC} value, 2 - 3 MPa$\sqrt{\text{m}}$[5]. The difference is possibly due to the microcracks or multiple cracks contained near the crack tip in the real material, as these should increase the fracture toughness.

4. Stress near the crack tip

The stress distribution in the simulation cell was calculated using the MD result of the stress at each atom. The total energy in the cell is given by:

$$E = \sum_{i<j}^{N} V_{ij}^{(2)}(r_{ij}) + \sum_{i,j<k}^{N} V_{jik}^{(3)}(r_{ij}, r_{ik}) \tag{14}$$

and the stress in the cell is then:

$$\sigma_{\alpha\beta} = \frac{1}{V} \cdot \left(\frac{\partial E}{\partial \epsilon_{\alpha\beta}} + \sum_i m_i v_\alpha^i v_\beta^i \right) \tag{15}$$

Table I. Elastic constants of β-Si$_3$N$_4$. G, ν were calculated according to the Voigt model.

	3-body potential	Ching *et al.*[4]
C_{11}(GPa)	591	409
C_{12}(GPa)	182	271
C_{13}(GPa)	162	201
C_{33}(GPa)	690	604
C_{44}(GPa)	377	108
G(GPa)	161	210
ν	0.33	0.27

where:

$$\epsilon_{\alpha\beta} = \frac{1}{2}\left(\frac{\partial u_\alpha}{\partial x_\beta} + \frac{\partial u_\beta}{\partial x_\alpha} \right). \tag{16}$$

The energy of atom i is defined as

$$E_i = \frac{1}{2}\sum_j V_{ij}^{(2)}(r_{ij}) + \frac{1}{2}\sum_{j,k} V_{jik}^{(3)}(r_{ij}, r_{ik}) \tag{17}$$

where:

$$\sum_i E_i = E. \tag{18}$$

The stress at atom i is defined as:

$$\sigma_{\alpha\beta}^i = \frac{1}{V_i} \cdot \left(\frac{\partial E_i}{\partial \epsilon_{\alpha\beta}} + m_i v_\alpha^i v_\beta^i \right) \tag{19}$$

where V_i is the volume of atom i defined as:

$$V_i = \frac{V_0}{N} \tag{20}$$

where V_0 is the equilibrium volume of the cell and N is the number of atoms in the cell. We then defined the stress in the cell as the average of $\sigma_{\alpha\beta}^i$ over all i, $\bar{\sigma}_{\alpha\beta}$.

The stress distribution in the cell was calculated using $\bar{\sigma}_{\alpha\beta}$. First, β-Si$_3N_4$ without a crack was loaded with a tensile stress in the y direction to investigate the validity of using $\bar{\sigma}_{\alpha\beta}$. Figure 4 shows the stress on each atom, $\bar{\sigma}_{\alpha\beta}$ (σ_{22}), and the stress calculated using elastic constants. Although the atomic stresses are scattered, the average value is almost constant, in good agreement with the calculation using elastic constants, suggesting that calculating the stress distribution from the MD results is a valid method. Next, the stress distributions for models containing a crack were calculated. Figure 5 shows the σ_{11}, σ_{12}, and σ_{22} values near the crack tip. The MD values are in good agreement with the results of linear elastic analysis except for near the crack tip.

5. Conclusion

MD simulations of single crystals of β-Si$_3$N$_4$ reveal that a crack propagates, when $K_I \geq 0.7$ MPa$\sqrt{\text{m}}$, in good agreement with the value calculated on the linear elastic condition. Stress near the crack tip, assuming that the stress is the average of the atomic stress, is also in good agreement with the linear elastic solution. These results indicate that our MD simulation describes the fracture behavior of β-Si$_3$N$_4$ well.

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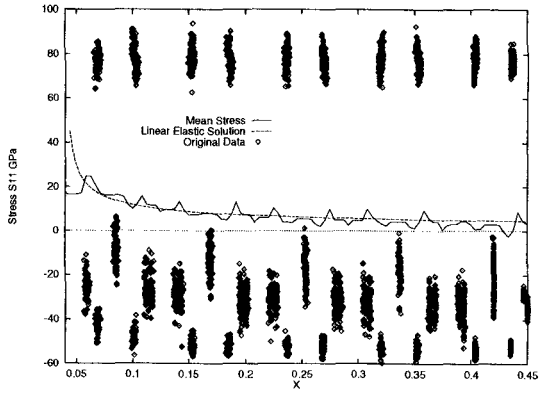
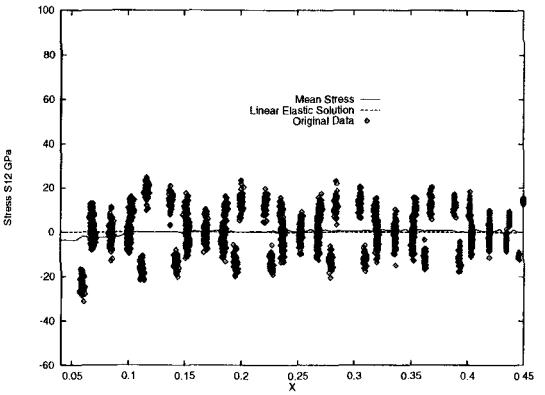
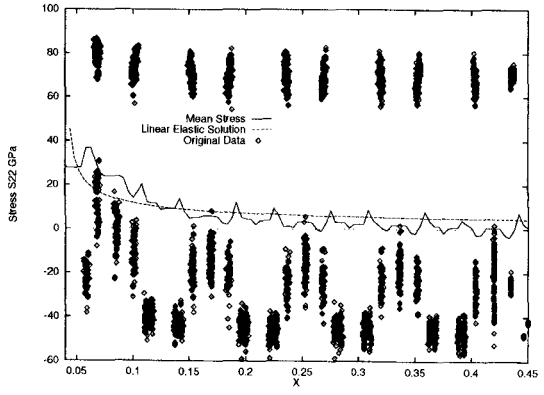


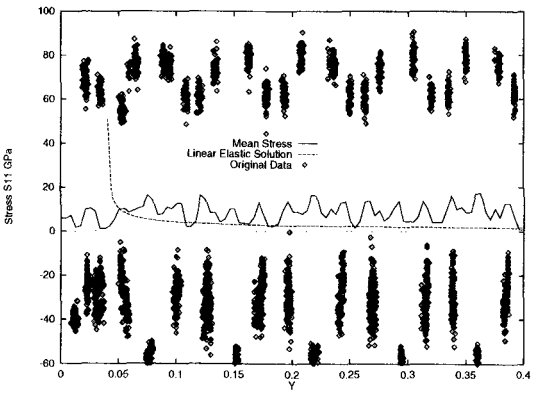
(a) σ_{11} along x axis



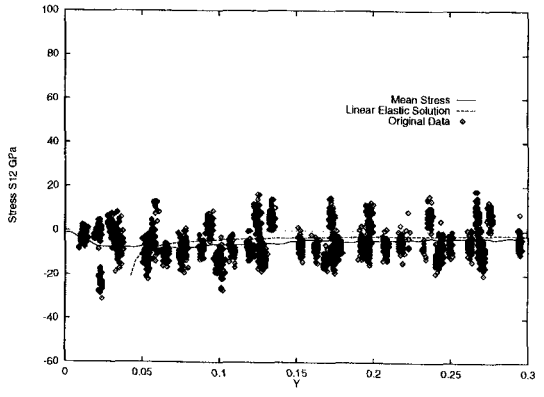
(b) σ_{12} along x axis



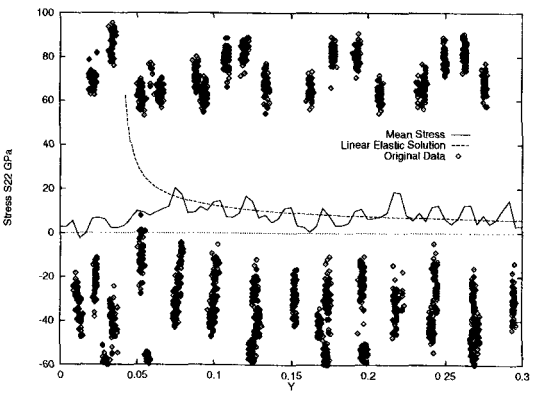
(c) σ_{22} along x axis



(d) σ_{11} along y axis



(e) σ_{12} along y axis



(f) σ_{22} along y axis

Figure 5. Stress distribution near the crack tip

(Received December 11, 1998; accepted February 28, 1999)