

Molecular dynamics — simulations of the fracture toughness of sapphire

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Abstract

A new method for the estimation of the fracture toughness K_{IC} by using the Molecular Dynamics method is presented. The Al_2O_3 single crystals were deformed under constant displacement conditions with an initial crack of length 1 nm. The strain was applied in the a axis, which was set perpendicular to the crack plane, so that the mode I crack deformation was achieved. The total energy, which is a result of MD simulations, was analyzed in the elastic, plastic and surface energy part. From the structural plots the crack propagation was measured and the analysis using the Griffith criterion was applied. From the critical condition the fracture toughness was estimated. The experimental values for the fracture toughness in the four crack orientations (plane and direction) (11.0)[001], (11.2)[12-1], (00.1)[100], and (11.0)[010] were 2.84, 2.71, 5.62, 2.67 $MPa\ m^{-1/2}$, respectively, and the calculations are in good agreement. Also, a heterogeneous alumina–zirconia nanocomposite material was analyzed by this method. Although the particle had only a diameter of 1 nm, the toughness increased because the interface was found to be quite strong. © 2000 Published by Elsevier Science Ltd.

Keywords: Alumina; Molecular dynamics; Composite materials; Fracture toughness; Crack propagation; Gibbs free energy; Surface energy

1. Introduction

Molecular Dynamics is a well established materials development process. At an atomic scale all processes, which have been observed by experiment, can be studied more accurately and modeled in detail. Successful applications have been the calculation of atomic structures of grain boundaries and interfaces and properties [1–4]. The computer program ‘MOLDY’ [5,6] also considers the long-range Coulomb interaction and is especially suitable for ceramic materials; especially for ZrO_2 [2,7,8] and Al_2O_3 [9], good two-body pair potentials are available. The crack propagation on an atomic scale is an unknown phenomena, which can not be studied by experiment, but which has been already studied by

a combined MD-/FEM-Method (Finite Element Method) [10,11]. Computers are now capable of handling an increased number of atoms in a supercell. The output of the molecular dynamics calculations can be analyzed in different ways and it is the aim of this paper to evaluate the method of calculating the fracture toughness of ceramics from the output of the MD calculations. The Griffith criterion is based on the analysis of the energy release rate during the crack propagating as a function of the crack length.

The improvement of fracture toughness is one of the important challenges for developing new structural ceramic materials. Recently, functional gradient materials (FGM) have been introduced, which give the perspective of making a smooth bonding between metal and ceramic materials. New composite materials consisting of an ultrafine distribution of alumina and zirconia particles [12] promise better thermal-shock properties and higher fracture toughness than monolithic alumina [13]. The experimental search for suitable material

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Table 1
MD-parameter according to the Buckingham potential in Eq. (1) for alumina and cubic-zirconia

	q_i, q_j (e)	$f_o(b_i + b_j)$ (kcal mol ⁻¹)	$a_i + a_j$ (0.1 nm)	$1/(b_i + b_j)$ (10/nm)	$c_i^* c_j$ [(0.1 nm) 6/kcal mol ⁻¹]	D_{ij} (kcal mol ⁻¹)	α_{ij}	r_{ij}
Al-O	1.8000	0.2903	3.0493	3.4447	0.0000	21.6893	2.0000	1.8000
O-O	-1.2000	0.3604	3.7766	2.7747	0.1696	0.0000	0.0000	0.0000
Zr-O	4.0000	46.269	0.0000	2.657	46.122	0.0000	0.0000	0.0000
O-O	-2.0000	987.134	0.0000	6.711	2582.832	0.0000	0.0000	0.0000

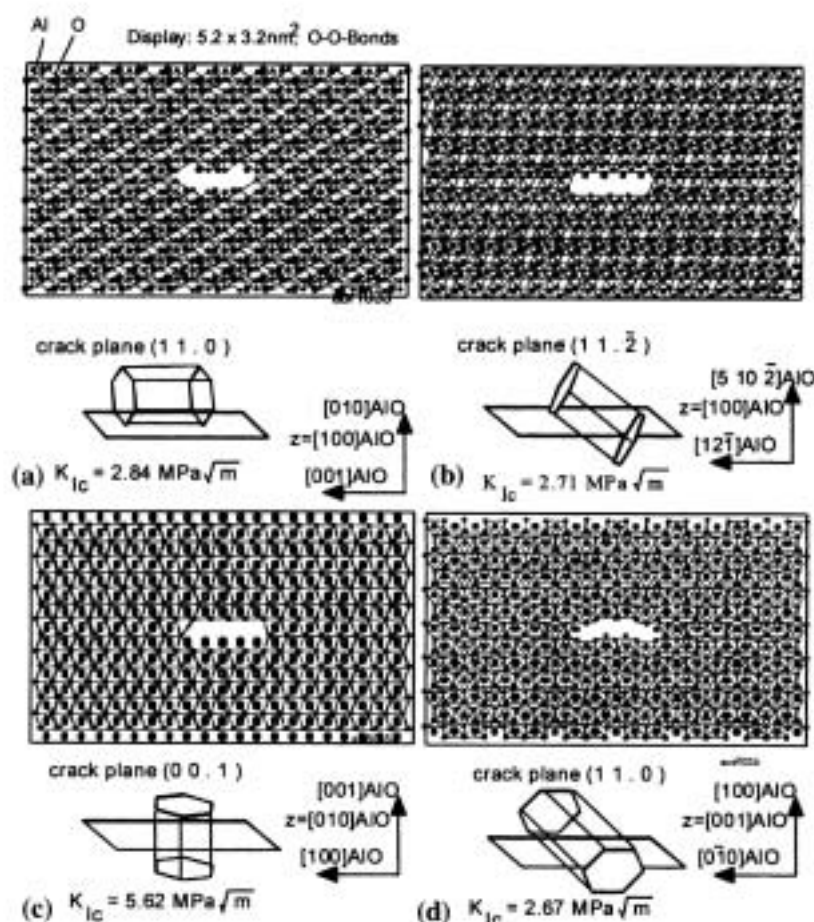


Fig. 1. Initial supercell for four different orientations of single crystals: (a) (11.0)[001]; (b) (11.-2)[12-1]; (c) (00.1)[100]; and (d) (11.0)[010].

combinations and microstructural parameters, like particle size, orientation relationship and volume fraction is quite time consuming, therefore, computer simulation will become an important tool for this purpose in the future. It is the goal of further engineering, on an atomic scale, to use these new dimensions of material development for practical applications.

This study had two goals: (1) to test the ability of the MD method for calculating the fracture toughness and to evaluate the analyzing method; and (2) to understand the crack toughening mechanism of alumina-zirconia composite materials on an atomistic scale.

2. Method

The molecular dynamic program 'MOLDY' [5,6] used in this study allows the use of two-body central-force potential parameters including the long-range Coulomb interaction. The Buckingham potential in the following form was applied

$$E_R = \frac{z_i z_j e^2}{r} + f_o (b_i - b_j) \exp\left(\frac{a_i + a_j - r}{b_i - b_j}\right) - \frac{c_i c_j}{r^6}$$

$$+ D_{ij} (e^{-2\alpha_{ij}(r-r_0)}) - 2D_{ij} (e^{-\alpha_{ij}(r-r_0)}) \quad (1)$$

with $f_o = 6.9511 \times 10^{-11}$ N. The adjustable parameters were taken from previous works [4,7,8] and are listed in Table 1. The parameters for alumina are excellent concerning the accuracy and stability of the crystal lattice, those for cubic zirconia [7,8] work suitably well, however, the lattice constants are slightly too large.

The following schedule for calculating the crack propagation was studied. A crack of length 1 nm and one atomic layer was introduced in supercells with three different sizes: $3.2964 \times 5.2148 \times 0.4759$ nm³ with 950 atoms; $3.2964 \times 5.2148 \times 1.9032$ nm³ 4000 atoms; and $10.13 \times 10.42 \times 1.903$ nm³ with 23 000 atoms. Four different orientations for the supercell were chosen as shown in Fig. 1, so that the crack plane was set parallel to a $(10\bar{1}0)$ plane, the crack direction is $[0001]$. The loading was performed by constant strain in the $[10\bar{1}0]$ direction, so that the crack propagates in mode I. This value is near the lower limit for all possible crystallographic directions. The simulations were continued by increasing gradually the uniaxial strain in the $[10\bar{1}0]$ direction: after each 2000 time steps the strain was increased by 0.625%. Each time step has a length of 0.01 ps = 10^{-14} s. The strain was increased until the crack has propagated through the whole supercell. The

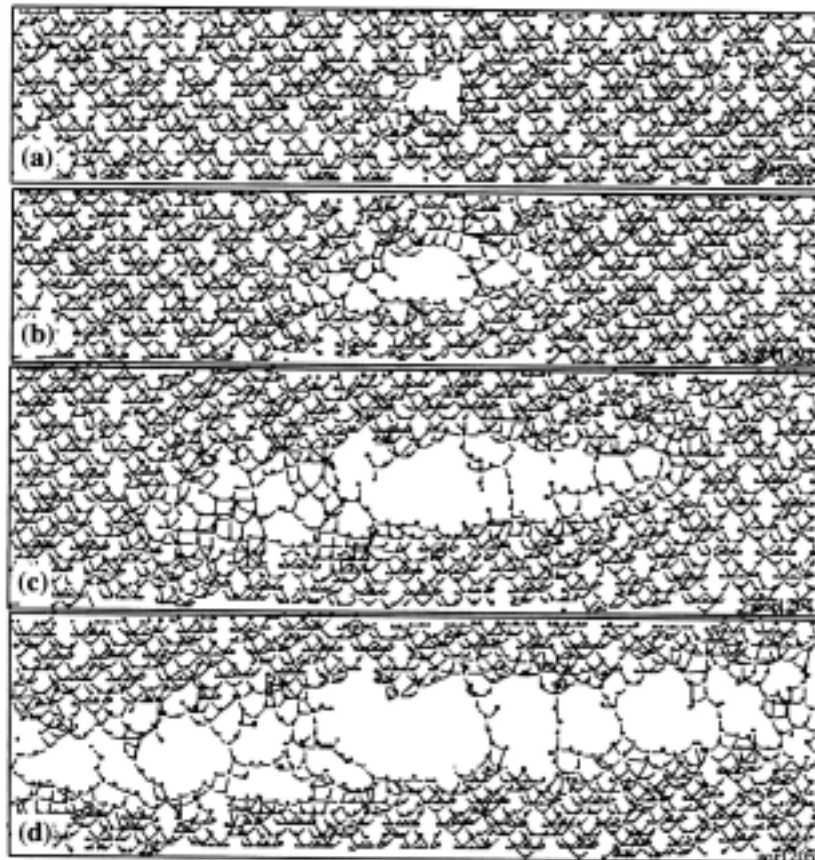


Fig. 2. Structural plot of the extension of a (11.0)[001] crack when the strain is increased, $\epsilon =$ (a) 1.05; (b) 1.07; (c) 1.08; and (d) 1.09.

crack length was measured from plots of the structure after each simulation run (Fig. 2).

For estimating the fracture toughness the analysis according to the flow chart in Fig. 3 was applied: The internal energy U was plotted as a function of the time (Fig. 3a), then the values were normalized by considering the values of single crystal calculations. From the plot of the lattice constants vs. time (Fig. 3b) the strain was calculated. The crack length (Fig. 3c) as a function of the strain also shows almost linear behavior. From other calculations the surface energy γ was estimated and the plot as a function of the crack length c is linear. For the following analysis the Griffith formula and the definition of the fracture toughness was used:

$$U = \pi \cdot \sigma^2 \cdot d \cdot l / E' \cdot c^2 + 2 \cdot c \cdot d \cdot \gamma \quad (2)$$

and

$$K_{IC} = \sigma_c \cdot \sqrt{c_c} = \sqrt{\frac{2 \cdot E' \cdot \gamma_c}{\pi}} \quad (3)$$

Finally the total energy per mole was plotted as a function of the crack length (Fig. 3d). The maximum was fitted according to the Griffith criterion by adjusting the stress σ and the critical crack length c_c . The plastic deformation, which is not included in the Grif-

fith theory, remains as a constant part after the crack has propagated through the specimen (Fig. 3d). This analysis depends on the fit parameter and the influence was checked: When the stress is varied from 10.5 MPa (Fig. 4a), 9 MPa (Fig. 4b), to 8 MPa (Fig. 4c), the fracture toughness changes from 2.82 to 3.78 MPa $m^{-1/2}$. This shows the limits of the accuracy of this method.

For the composite material a 0.9-nm large spherical cubic zirconia particle was embedded in an alumina matrix with the following orientation relationship (100)ZrO₂//(10 $\bar{1}$ 0)Al₂O₃, [001]ZrO₂//[0001]Al₂O₃. This was performed by deleting all atoms which are situated inside the sphere of the alumina matrix. In the second cell of the same size consisting of the zirconia phase only, the atoms inside the sphere were taken and finally both cells were combined. The size of the supercell is 3.2964 \times 5.2148 \times 1.9032 nm³ with 1391 Al, 2059 O-atoms belonging to alumina and 91 Zr, 216 O atoms belonging to the zirconia particle. The parameters at the heterogeneous interface were chosen in the following way: the interaction between the oxygen cations of the alumina and the zirconia phases were assumed as in the zirconia phase. The anion-cation interactions were chosen in the same way as the particular phases. The initial crack was placed near the particle off the center in order to simulate a kind of arbitrary case.

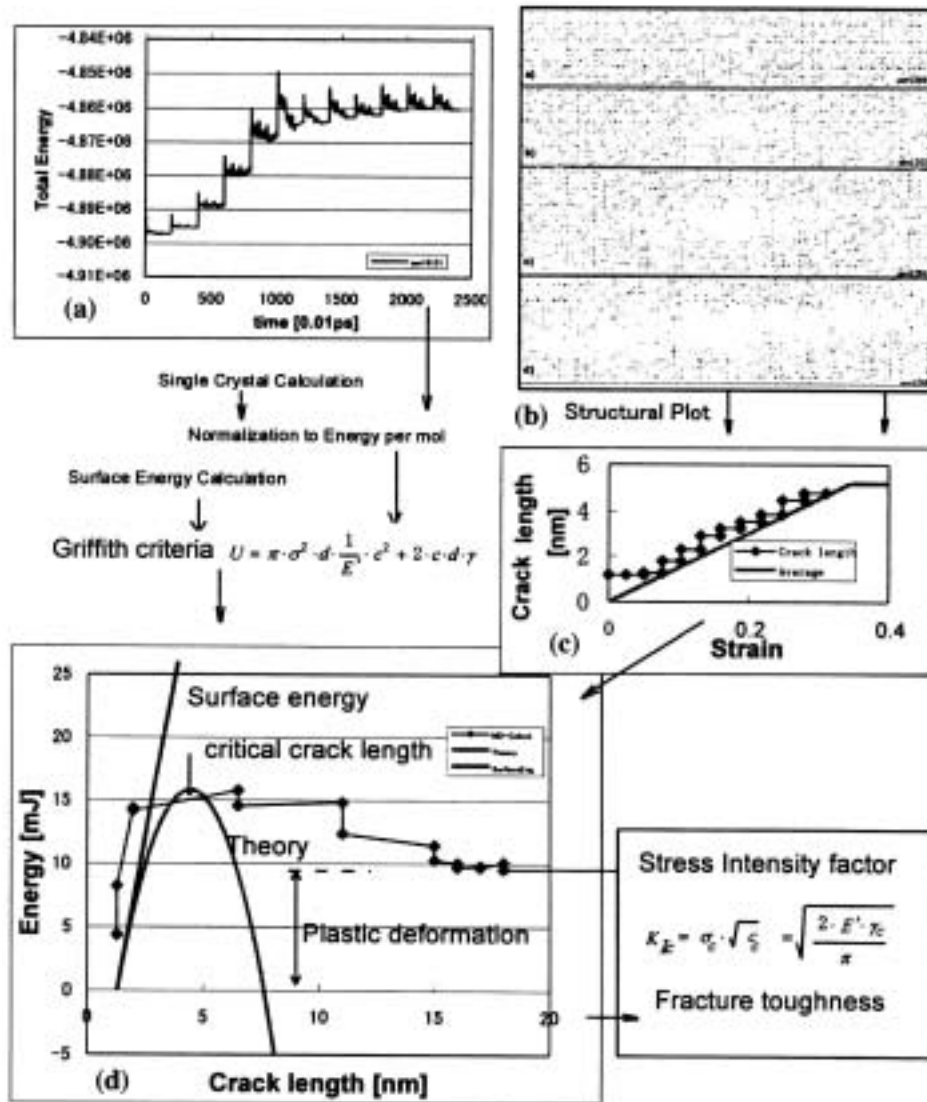


Fig. 3. Scheme for analyzing the data, from top to the bottom: total energy as a function of time, deformation rate, crack length vs. strain, plot for the Griffith criteria, energy as a function of crack length.

3. Results

The crack propagation in alumina single crystals shows, in general, the same behavior, which is also observed in experiments. At a strain level of $\varepsilon = 1.05$ the crack first enlarges its crack opening displacement, then at 1.1 it creates a damaged zone in front of the crack tip where atomic bonds are broken before the crack tip, then it starts to propagate. Finally, before the whole specimen breaks, a lot of crack bridging between the two crack flanks is observed. These chains of single atoms are also observed in in situ HRTEM experiments, where two parts are pulled apart. In this experiment they are stable only for a few seconds, while the time window of the MD simulation is even much smaller — in the range of picoseconds. Hence, the MD calculations are realistic, but it needs a longer time and a higher strain to relax these bonds. For this orientation the experimentally determined K_{IC} value for single

crystals is $2.84 \text{ MPa}/\sqrt{m}$ [13]. The energy as a function of time is shown in Fig. 5a, together with the plots of the other orientations (Fig. 5b–d) corresponding to the geometry plots in Fig. 2. The orientation with the highest fracture toughness (Fig. 5c) has the steepest increase in the energy function. When calculations using supercells with larger dimensions in crack direction are compared to results on smaller ones, it was found that for small supercells a higher strain was needed to propagate the crack.

The alumina-zirconia composite material with the initial crack is displayed in Fig. 6a after a strain of $\varepsilon = 1.09$ and in Fig. 6b after a strain of 1.20. The supercell size of the display in Fig. 6a is 3.2964×5.2148 , and in Fig. 6b is $4 \times 8 \text{ nm}^2$. The atoms in the zirconia particle have the same cubic crystalline structure as in a large crystal, but as soon as stress is applied their atomic bonding gets more and more disturbed and transforms into an amorphous phase. It cannot be

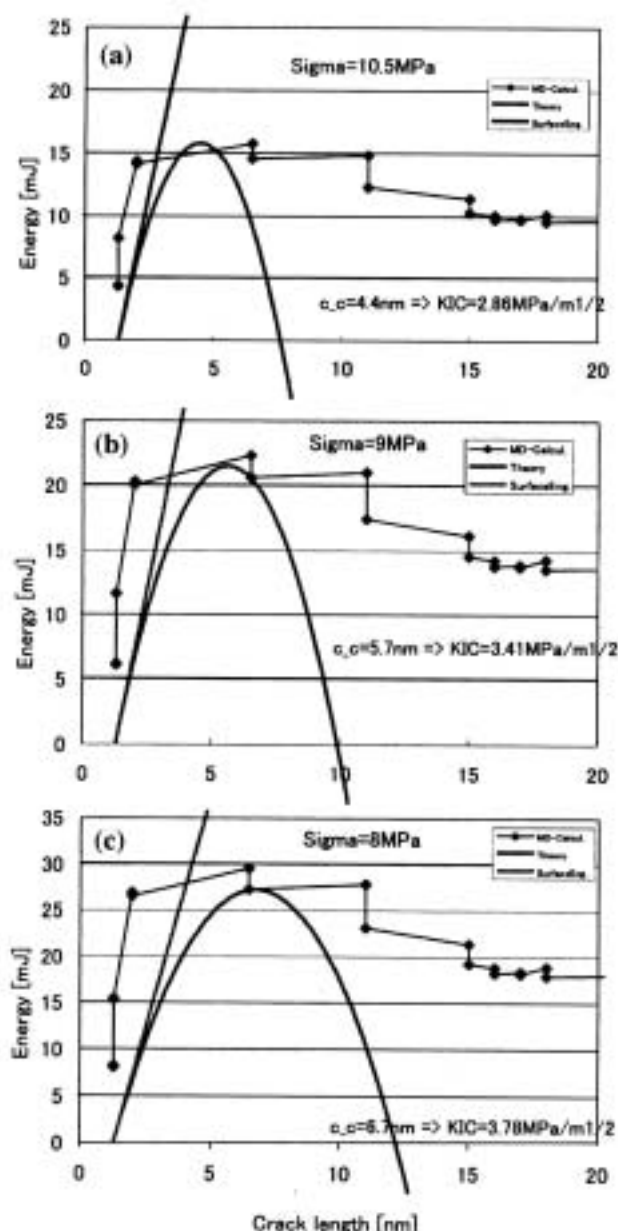


Fig. 4. Dependence of the estimated fracture toughness on the value of the stress.

overlooked that for this purpose, the potential is still not sufficient. By increasing the strain the crack tip approaches the particle and is stopped there. At the interface a lot of energy of the crack is absorbed for straining the particle. The interface between alumina and zirconia was found to be rather strong even when the strain reached a value of more than 1.30. At that level the bonding between the alumina atoms in the matrix, as well as in a previous single crystal study, had already failed. This result is consistent with experimental observation, where it was found that ultrafine zirconia particles lead to a toughening mechanism [12]. The final crack penetrates through the particle, not through the interface.

4. Discussion

The analysis of the output of MD calculations in order to calculate the fracture toughness has been presented. The higher the number of atoms in the supercell, the more accurate the analysis. Of course the crack dimensions should be small compared to the size of the supercell in order to fulfill the assumptions of the Griffith criterion. The accuracy also increases, when the deformation rate is reduced. The direct deformation of the supercell is straightforward compared to the FEM-MD coupling method [10,11], it avoids the problems with coupling and the internal energy can be calculated and used for this analysis. The FEM method was introduced in order to consider the long-range stress field in front of crack tips. However, the computer facilities have increased in the last years, so that even systems with a large number of atoms can be calculated precisely in a reasonable length of time. The molecular dynamics simulations have the advantage that the energy data can be directly compared, and the contributions of each part, like surface energy, elastic and plastic energy, can be separated.

This calculation method will be useful in the future to predict the properties of new composite materials. The properties of the interface can be calculated and the influence of the microstructural parameters like volume fraction, size, orientation, thermal mismatch can be studied.

5. Conclusion

The molecular dynamics simulations of the crack propagation could be successfully applied to calculate the fracture toughness. It allows the study of crack propagation on an atomic scale and the values deduced from the Griffith analysis are in good agreement with the experimental values. The limits of this method and the influence on the fit parameters have been discussed. The method can also be applied for nanocomposite material consisting of zirconia and alumina, in which a toughening due to the nanosize particle was found.

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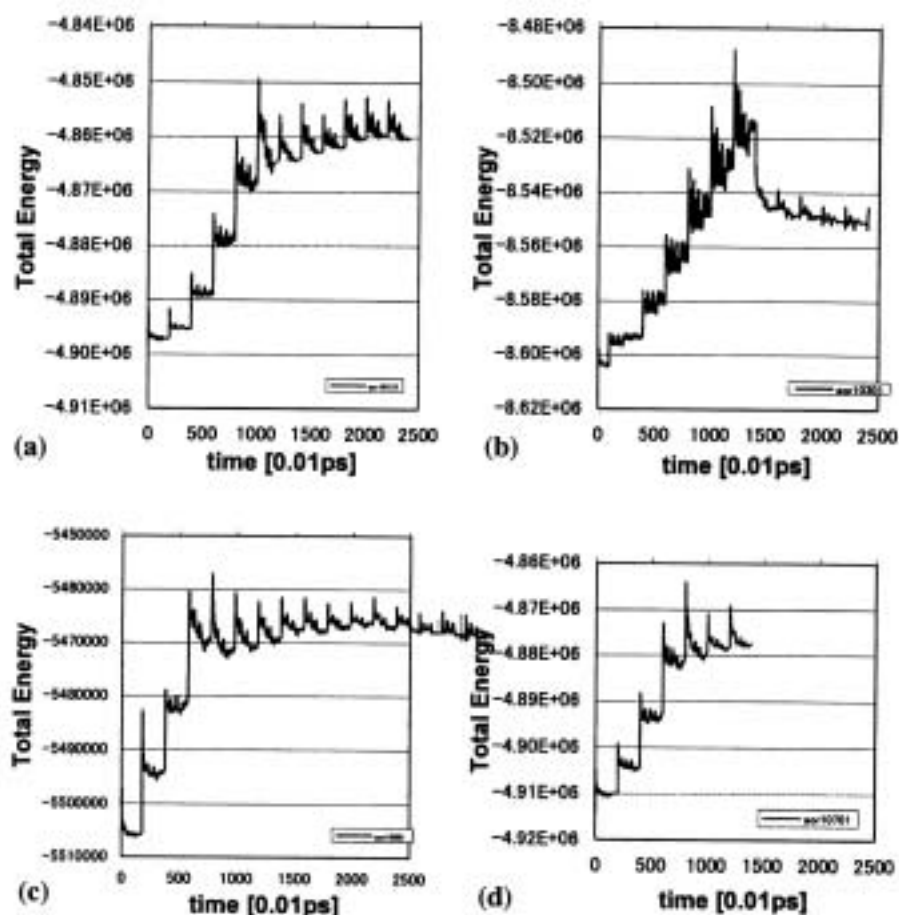


Fig. 5. Total energy as a function of the time for the four different orientations of the crack according to Fig. 1.

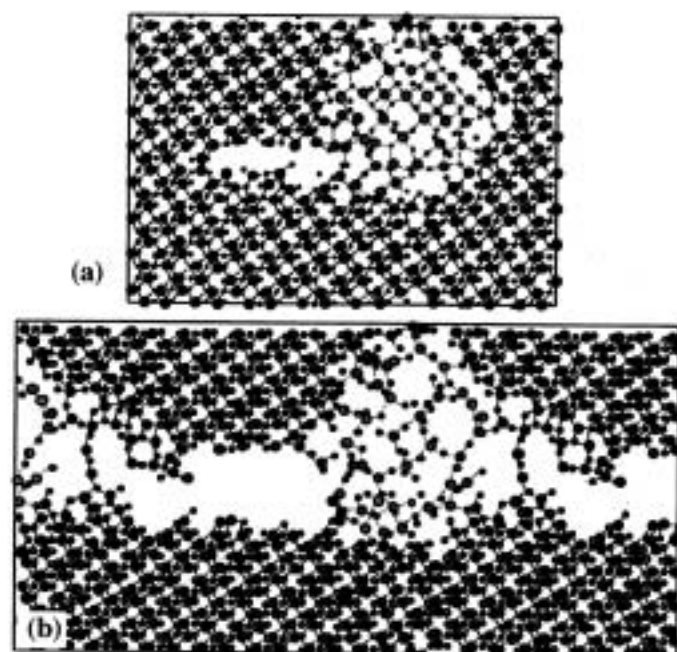


Fig. 6. Atomic structure of a crack in a composite material of alumina with a 0.9-nm zirconia particle: (a) at strain $\epsilon = 1.09$; (size of display $3.296 \times 5.215 \text{ nm}^2$ which is supercell size); and (b) at strain $\epsilon = 1.20$ (size of display $4 \times 8 \text{ nm}^2$).

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