Ceramics International 30 (2004) 441–446

CERAMICS INTERNATIONAL

www.elsevier.com/locate/ceramint

Effect of geometrical factors on the mechanical properties of Si_3N_4/BN multilayer ceramics

Qingfeng Zan*, Chang-an Wang, Yong Huang, Shike Zhao, Cuiwei Li

The State Key Laboratory of New Ceramics and Fine Processing, Department of Materials Science and Engineering, Tsinghua University, Beijing 100084, People's Republic of China

Received 5 December 2002; received in revised form 14 April 2003; accepted 15 May 2003

Abstract

The relationships between mechanical properties of Si_3N_4/BN multilayer materials and the geometrical factors, number of layers (N) and layer thickness ratio (λ_h), have been studied. At a given number of layers, with the increase of layer thickness ratio, the toughness and bending strength both increase, and when it continues to increase, the two curves both level off. Keeping the layer thickness ratio of about 10 and increasing the number of layers, the bending strength decreases slightly, and a maximum toughness was obtained at $N \approx 30$. Furthermore, a mechanical model was established and used to analyze the above experimental results. According to the experimental and calculated results, suitable geometrical parameters were advised in the design of Si_3N_4/BN multilayer materials as $\lambda_h \approx 10$ and $N \approx 30$.

Keywords: Si_3N_4/BN; Multilayer; Geometrical factor; Number of layers; Layer thickness ratio

1. Introduction

Since Clegg et al. [1] first fabricated SiC/C multilayer composites in 1990, multilayer ceramics have received much attention because of their improved properties. The improved properties can be achieved by designing weak interfaces for crack deflection [2–5], controlling the frontal shape of the transformation zones in ZrO_2 ceramics with barrier layers, and forming residual compression in surface layers [6–8]. As one of the most promising systems, Si_3N_4/BN multilayer ceramics have been studied intensively in the past tens of years [2–6], their bending strength and work of fracture can reach 500 MPa and 5000 J/m^2 respectively. In Si_3N_4/BN multilayer ceramics, there exists a weak interface between Si_3N_4 and BN layers, which was formed by the incorporation of weak BN interlayer. The weak interface can deflect the crack propagating perpendicularly to the plane of laminates repeatedly during fracture, thus leading to extremely high work of fracture.

It was proposed that the mechanical properties of Si_3N_4/BN multilayer ceramics were strongly dependent on geometrical factors (such as layer thickness and thickness ratio) [6]. Unfortunately, it has not been documented particularly so far, although a lot of literature has been accumulated on Si_3N_4/BN multilayer ceramics.

In this work, a series of experiments were designed and conducted to investigate the influences of geometrical factors on the mechanical properties of the Si_3N_4/BN multilayer ceramics. A mechanical model based on the flexure beam was also established to analyze the relationships between theoretical predictions and the experimental results.

2. Experiment procedure

The raw powders of Si_3N_4 (Founder High Tech. Ceramic Co. China) with 8 wt.% Y_2O_3 (99.9%), 2.5 wt.% Al_2O_3 (99.9%), and 1.5 wt.% MgO (99.9%) were ball-milled for 24 h in an alcohol medium. Then the sluny was dried and sieved in a 60-mesh screen. Selecting PVA as a binder, the mixed powders were ball-milled for 24 h to prepare the slurry, and then the slurry

* Corresponding author. Tel.: +86-10-6278-5488; fax: +86-10-6277-2857.

E-mail address: zanqf98@mails.tsinghua.edu.cn (Q. Zan).

0272-8842/$30.00
doi:10.1016/S0272-8842(03)00129-9

was cast into tapes on a polyester film and formed green tapes with different thickness. The obtained tapes were dried in air and punched into a rectangle (32×38 mm). Then the punched tapes were coated with slurry of BN (commercial powder), whose thickness was controlled by the slurry concentration and the immerging time in the slurry. The coated green tapes were dried and stacked into the graphite die. After the removal of the binder, the green body was hot pressed at 1820 °C for 1.5 h in a flow on N_2 atmosphere.

The sintered specimens were sliced into test bars with the dimensions of 4×3×36 mm for bending strength and 4×6×30 mm for fracture toughness and work of fracture. A three-point bending test for bending strength was carried out at room temperature with a span of 30 mm and crosshead rate of 0.5 mm/min. The work of fracture was defined and calculated as the area under the load–displacement curve divided by two-fold of the cross-section area of the bend bar, which was measured by the single-edge-notch-beam method (SENB) at room temperature with a span of 24 mm and crosshead rate of 0.05 mm/min. Five specimens were tested for each test to get an average value. All the tests was performed on a A-2000 Shimadzu universal materials testing machine. The loads were applied perpendicular to the plane of the layers. The microstructure of the materials was observed by scanning electron microscope (SEM) of CSM950.

3. Results

3.1. Fracture behavior of the Si_3N_4/BN multilayer ceramics

Fig. 1 (L–D curves) shows two typical load–displacement curves of Si_3N_4/BN multilayer composite and conventional monolithic Si_3N_4, respectively. It can be seen that the Si_3N_4/BN multilayer composite exhibits a non-brittle failure while the conventional monolithic Si_3N_4 fractures catastrophically. For the Si_3N_4/BN multilayer composite, after the first load drop, the load bearing ability of the testing bar still retains over 50% of the peak load. Until totally fractured, the Si_3N_4/BN multilayer composite gives a prolonged deflection besides the elastic deformation. This shows that the multilayer ceramic exhibits a different fracture behavior from that of the monolithic ceramic. Hence it may be inferred that the mechanical properties of ceramic materials can be substantially improved by a special structural design similar to that conducted on biomaterials as already indicated in the literature [2,5].

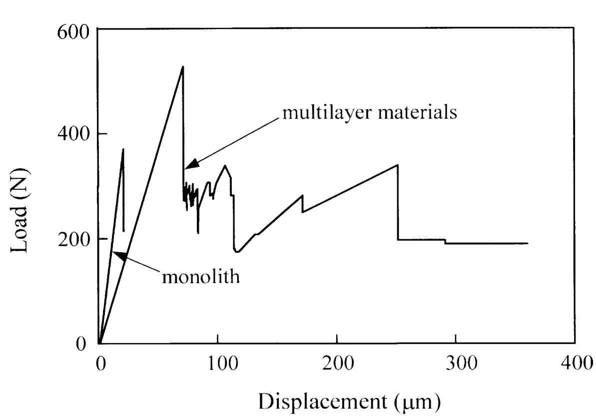

Fig. 1. Typical load–displacement curves of Si_3N_4/BN multilayer materials and conventional monolithic Si_3N_4.

Fig. 2 shows the fracture behaviors of the Si_3N_4/BN multilayer ceramics. It can be observed from Fig. 2(a) that Si_3N_4 layers are about 80–100 μm thick and BN interlayers only 10–20 μm thick. The multilayer structure mainly contributes to high toughness of the Si_3N_4/BN multilayer composites. The main toughening mechanism of the multilayer composites is that the interlayer deflects the crack repeatedly [as shown in Fig. 2(a)], and there are other toughening mechanisms in the ceramics, such as crack bridging [Fig. 2(b)], friction and sliding [Fig. 2(c)], and matrix-layer pull-out [Fig. 2(d)] and so on. These toughening mechanisms from the interlayers are considered to mainly contribute to high fracture toughness and work of fracture of the multilayer composites because a large amount of fracture energy is absorbed during fracture.

3.2. Effect of thickness ratio (λ_h)

At a given number of layers, the change of the thickness ratio between matrix-layers and interlayers in a standard specimen, 3 mm thick, will change the absolute thickness of matrix-layers and interlayers respectively. Therefore, thickness ratio ($\lambda_h = h_2/h$l, where h is the layer thickness, the subscript 1 and 2 denoted the interlayer and matrix-layer, respectively) must be a sensitive geometrical parameter influencing the mechanical properties of Si_3N_4/BN multilayer ceramics. Fig. 3 shows the curves of the bending strength and work of fracture of the Si_3N_4/BN multilayer materials with various λ_h. With the increasing of the thickness ratio, the work of fracture increases firstly, and then levels off after about $\lambda_h = 4$–5. The similar curve of bending strength vs. layer thickness ratio is investigated from Fig. 3.

This can be easily understood from the structure of the Si_3N_4/BN multilayer ceramics. With the increase of λ_h, the matrix-layers become thicker, and the interlayers become thinner, correspondingly. In the Si_3N_4/BN multilayer ceramics, bending strength is mainly determined by matrix-layers, and work of fracture depends on the crack deflection by the interlayer. Hence, if the λ_h is very small (about $\lambda_h = 2$ in Fig. 3), the thick and loose BN applies a large space and small resistance of transverse

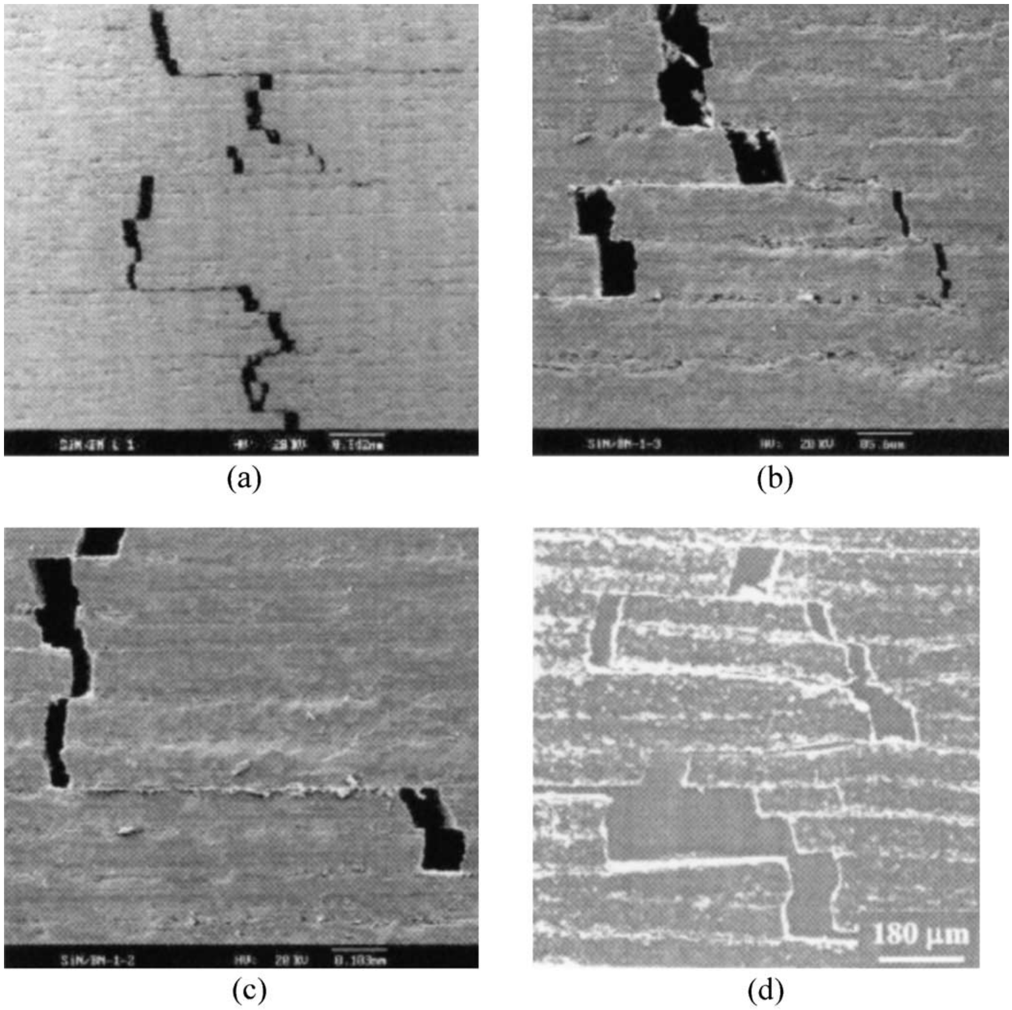

Fig. 2. SEM photographs of crack propagating paths in Si_3N_4/BN multilayer ceramics: (a) crack deflection; (b) crack bridging; (c) friction and sliding; (d) matrix-layer pull-out.

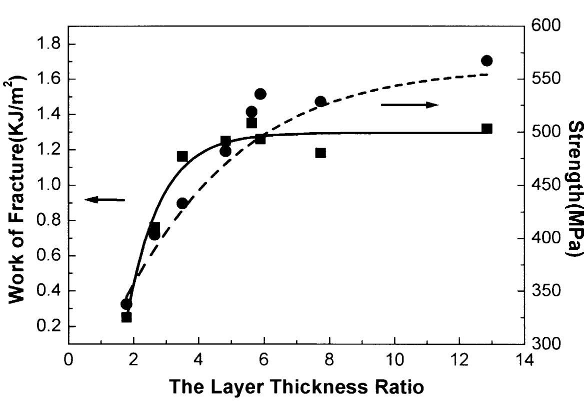

Fig. 3. Bending strength and work of fracture with various layer thickness ratios (- - - bending strength; —— work of fracture).

crack propagation, hence it will lower both the bending strength and work of fracture. On the other hand, if the λ_h is too large (i.e. the matrix layer is too thick and interlayer too thin), it is favorable to the strength obviously. But, because of the interlayer being too thin, the preparation procedure becomes difficult, which would lead to a defect in the interlayer increasing largely and lowering the strength of the multilayer material; on the other hand the too thin interlayer weakens the ability of deflecting crack, when the multilayer material is loaded, crack is difficult to deflect and propagate in the interlayer, in other words, the material is easy to fracture catastrophically and low work of fracture is obtained. These are the reasons that the two curves tend to level off when the layer thickness ratio is large enough.

Considering both bending strength and work of fracture, a suitable value of layer thickness ratio is suggested as about 10 in the multilayer structure design.

3.3. *Effect of layer number (*N*)*

In the case of approximately fixed thickness ratio, for example, about 10 for thickness ratio designed in the present work, the effect of matrix-layer thickness on mechanical properties of the laminated Si_3N_4/BN ceramics can be interpreted in terms of the effect of layer number (N) of a specimen with fixed thickness ($\sim$3 mm).

According to Fig. 4, it is found that the bending strength decreases with the layer number increasing. But the extent of decrease is very small (no more than 100 MPa), and with the layer number exceeding more than about 30, the curve tends to level off. For work of fracture of the multilayer materials, a maximum is observed at $N\approx 30$.

Similar to the analysis of the effect of thickness ratio on mechanical properties, the results above are also easily understood. Because the thickness of specimen is fixed, the increase of layer number means the thickness decrease of both matrix-layer and interlayer. Obviously, this will reduce the strength of the materials. It is well known that work of fracture is determined by both distance and times of crack deflection [3]. With an increase of layer number, the thickness of interlayers is deduced, and the following crack deflection distance is reduced. At the same time, the crack deflection times are increased obviously because of the interlayer number is increasing. These two factors influence the work of fracture together, and will lead to an optimal value in various numbers of layers, which was determined to be about 30 in this experiment.

4. Analysis and Discussions

4.1. Mechanical model of the Si_3N_4/BN multilayer materials in bending

In order to simplify the calculation process, a four-point bending test is used to analyze the stress of the multilayer ceramics, which is illustrated in Fig. 5. The specimen possessed the following parameters: layer thickness (h_1, h_2), Young's modulus (E_1, E_2), and Poisson's ratio (υ_1, υ_2), and the total thickness of a couple of matrix-layer and interlayer $h=h_1+h_2$. Total thickness,

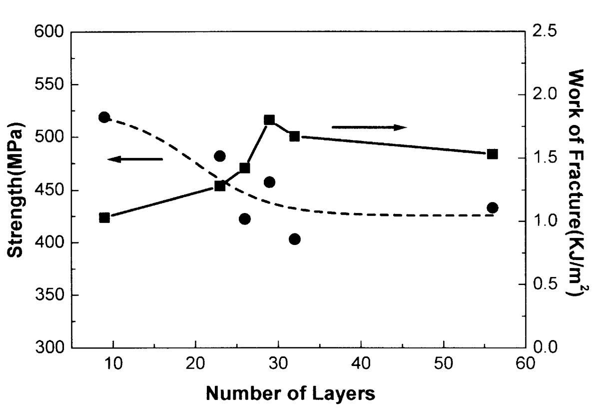

Fig. 4. Bending strength and work of fracture with a various number of layers (- - - bending strength; —— work of fracture).

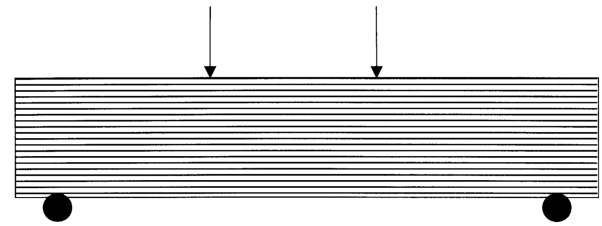

Fig. 5. The illustration of the flexure test.

width and span of the specimen are H, B and 2L in four-point bending, respectively. And some hypotheses are described as follows: (a) $H/2L \leqslant 0.2$; (b) The ratio of flexibility [$w(x)$] and the curvature radius [$\rho(x)$] is far less than 1, i.e. $w(x)/\rho(x) < < 1$; (c) $h_1 < < h_2$.

The number of the matrix-layer (m) is odd, i.e. $m=2n-1$, then $H=mh-h_1$. Hence, the maximum tensile stress and the maximum shear stress can be expressed as:

$$\sigma_{\max} = \frac{E_2 M}{2\Sigma}(mh - h_1) \tag{1}$$

$$\tau_{\max} = \frac{Q}{B\Sigma} S_{Z,\max} \tag{2}$$

where M is the bending moment in the segment with the stress of pure bending, Σ is the equivalent bending rigidity, Q is the shear stress, and $S_{z},_{\max}$ is the maximum equivalent static moment:

$$M = \int_{H/2}^{H/2} \frac{WBy^2}{\rho} \mathrm{d}y \tag{3}$$

$$\Sigma = \int_{H/2}^{H/2} BE(y)y^2 \mathrm{d}y \tag{4}$$

$$S_{Z,\max} = \int_{A*} E(y)y\mathrm{d}A$$

$$= \frac{E_2 B}{8}\left(H^2 - h_2^2\right) - \frac{E_2 - E_1}{8} Bhh_1\left(m^2 - 1\right) \tag{5}$$

4.2. Bending strength analysis

In the flexural test with the crosshead speed of 0.5 mm/min, the specimen will fracture by way of shear lapse or bending lapse due to the maximum shear stress ($\tau_{\max}$) or the maximum tensile stress ($\sigma_{\max}$), respectively. The latter is more familiar to the Si_3N_4/BN multilayer ceramics, when the expression below is satisfied:

$$\frac{\sigma_{\max}}{\tau_{\max}} \geqslant \frac{\sigma_b}{\tau_i} \tag{6}$$

where σ_{max} is the strength of the matrix-layer, τ_{max} is the shear strength of the interlayer.

For this material, according to the Eq. (1) and the fracture condition of $\sigma_{max} = \sigma_b$, the expression of the bending moment is:

$$M_b = \frac{2\Sigma\sigma_b}{e_2 H} \tag{7}$$

For monolithic Si_3N_4 ceramic:

$$\sigma^0_{max} = \frac{6M}{BH^2} = \sigma_b \qquad M^0_b = \frac{BH^2\sigma_b}{6} \tag{8}$$

Now the decreasing coefficient (κ) of the bending strength is defined by:

$$\kappa = \frac{M_b}{M^0_b} = 1 - \frac{\left(1 - \frac{1}{\lambda_e}\right)(m-1)\left[\lambda_h^2 + m(m-2)\right]\lambda_h}{(m-\lambda_h)^3} \tag{9}$$

where λ_e is ratio of Young's modulus ($\lambda_e E_2/E_1$), and its value is larger than 10 in this material.

From the expression [Eq. (9)], the strength curve with various thickness ratio (λ_h) and layer number (N) is obtained and illustrated in Figs. 6 and 7. The trends of the two curves are similar to experimental results (Figs. 3 and 4).

4.3. Toughness analysis

According to the propagation conditions of longitudinal and interfacial crack, a hypothesis of crack propagation process was made as follows [7,8]:

1. Displacement loading mode was used. The sequence of events is that a crack propagates instantaneously through the topmost layer (in the through-thickness direction), and then is deflected into interlayer and propagates along the interlayer between the fractured matrix-layer and the one below. Such through-thickness and interfacial cracking then takes place alternately until the beam is completely broken.
2. The through-thickness cracks occur in the center of the beam and the interfacial cracks propagate symmetrically from the center.
3. It is assumed that the portion of a layer between the through-thickness crack and the tip of an interfacial crack (a "debonded ligament") can carry no load and that such ligaments do not interfere with each other.
4. The critical strain energy release rate, Γ_m, of through-thickness crack propagation is mainly determined by the matrix-layer. Because of the presence of weak bonding interlayer, the total critical strain energy release rate, Γ_v, can be expressed as $\Gamma_v\Gamma_m h_2/h+\Gamma_i h_1/h$ according to the mixed rule. In general, Φ_i appears to be dependent on the balance of opening to shearing modes. This is commonly characterized by the so-called phase angle of loading, ψ, defined as the angle having a tangent equal to the ratio of the shearing to opening stress intensity factors. During delamination under bending, the crack tip loading is strongly mixed mode and the value of ψ is thought to remain approximately constant in the range of 40–60°.

Considering the hypotheses and using the model above, we can plot the curve of load–displacement in the case of given structural and geometrical factors of the multilayer materials. According to the area of load–displacement curve, the work of fracture can be calculated (the details of algorithm and calculation process about the model were discussed in Ref. [9]). The work of fracture of the multilayer material with a different number of layers (N) and thickness ratio of layers ($h_2/$

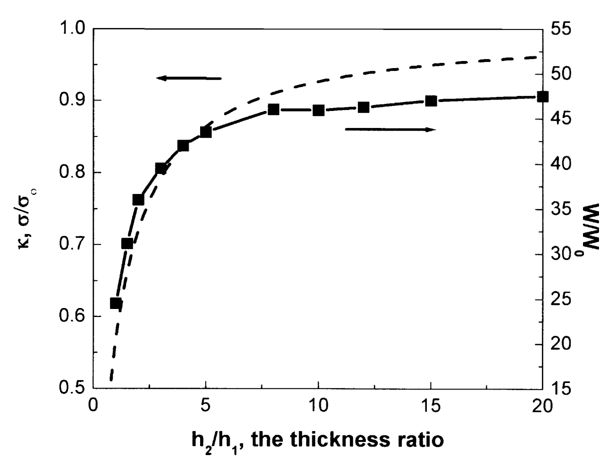

Fig. 6. Calculated result about the mechanical properties with various layer thickness ratios (- - - $\kappa,\sigma/\sigma_0$; —— W/W_0).

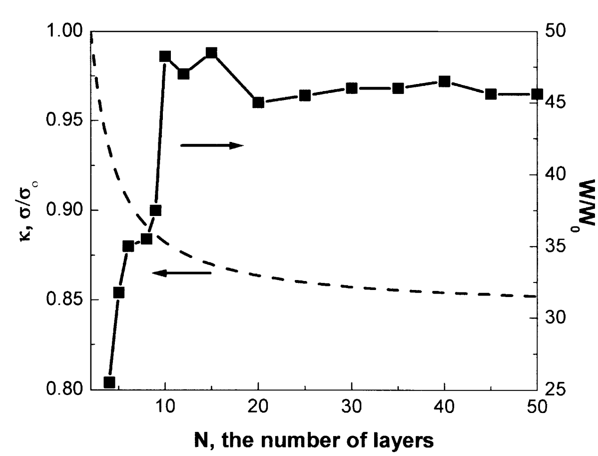

Fig. 7. Calculated results about the mechanical properties with various numbers of layers (- - - $\kappa,\sigma/\sigma_0$; —— W/W_0).

h_1) are plotted in Figs. 6 and 7, respectively, where W and W_0 are the work of fracture of multilayered Si_3N_4/BN and monolithic $S1_3N_4$ ceramics respectively.

Comparing Figs. 6 and 7 to Figs. 3 and 4, the similar curve trend was found, but different critical points and absolute value of mechanical properties. That is because of the following two reasons. On the one hand, in the model calculation only the absorbed energy by the crack deflection is calculated. However, in fact many other toughening mechanisms in Si_3N_4/BN multilayer materials were identified, such as bridging, frictional sliding, whisker toughening, and so on [2,3,5]. A mass of energy is absorbed during the fracture process by these mechanisms. On the other hand, some defects will be unavoidably introduced into the multilayer materials in fabrication processing; therefore the mechanical properties of the multilayer materials are lower than those calculated results by the model.

According to the above four figures, synthetically considering bending strength and work of fracture, optimal geometrical factors are selected as $\lambda_h \approx 10$ and $N \approx 30$ (corresponding to a thickness of about 100 μm for Si_3N_4 layer and 10 μm for BN layer) for work of fracture reaching optimum and bending strength remaining relative high level.

5. Conclusions

(1) At a given number of layers, with the increase of layer thickness ratio in a range, the toughness and bending strength both increase, and when it continues increasing, the two curves both level off.

(2) Keeping the layer thickness ratio of about 10 and increasing the number of layers, the bending strength decreases slightly, and a maximal toughness was obtained at $N \approx 30$.

(3) According to the mechanical model arid experimental results, it is suggested that the number of layers and ratio of layer thickness are 30 and 10 in structural design.

References

[1] W.J. Clegg, K. Kendall, N.McN. Alford, T.W. Button, J.D. Birchall, A simple way to make tough ceramics, Nature (London) 347 (1990) 455–457.

[2] H. Liu, S.M. Hsu, Fracture behavior of multilayer silicon nitride/boron nitride ceramics, J. Am. Ceram. Soc. 79 (1996) 2452–2457.

[3] D. Kovar, M.D. Thouless, J.W. Halloran, Crack deflection and propagation in layered silicon nitride/boron nitride ceramics, J. Am. Ceram. Soc. 81 (1998) 1004–1012.

[4] Y. Huang, H. Guo, Z.P. Xie, The fine micro-structure of interface layer for laminated Si_3N_4 ceramics, J. Mater. Sci. Lett. 17 (1998) 569–571.

[5] C.A. Wang, Y. Huang, Q.F. Zan, H. Guo, S.Y. Cai, Biomimetic structure design—a possible approach to change the brittleness of ceramics in nature, Mat. Sci. Eng. C: Biomimetic and Supramolecular Systems 11 (2000) 9–12.

[6] S.Y. Cai, PhD Dissertation, Tsinghua University, 1998.

[7] C.A. Folsom, F.W. Zok, F.F. Lange, Flexural properties of brittle multilayer materials: I, modelling, J. Am. Ceram. Soc. 77 (1994) 689–696.

[8] A.J. Phillipps, W.J. Clegg, T.W. Clyne, Fracture behavior of ceramic laminates in bending—I. modeling of crack propagation, Acta Metall. Mater. 41 (1993) 805–817.

[9] X.H. Guo, Q.H. Cai, C.A. Wang, Y. Huang, A mechanics analysis of strengthening toughening design of multilayer structure ceramics, Acta Mech. Solid Sina 21 (2000) 313–324.