Vol. 43 No. 5 SCIENCE IN CHINA (Series A) May 2000

PHYSICS

Microscale mechanics for metal thin film delamination along ceramic substrates

WEI Yueguang (魏悦广)

LNM, Institute of Mechanics, Chinese Academy of Sciences, Beijing 100080, China (email: ywei@lnm.imech.ac.cn)

Received February 12, 1999

Abstract The metal thin film delamination along metal/ceramic interface in the case of large scale yielding is studied by employing the strain gradient plasticity theory and the material microscale effects are considered. Two different fracture process models are used in this study to describe the nonlinear delamination phenomena for metal thin films. A set of experiments have been done on the mechanism of copper films delaminating from silica substrates, based on which the peak interface separation stress and the micro-length scale of material, as well as the dislocation-free zone size are predicted.

Keywords: metal thin film, nonlinear delamination, microscale mechanics.

The thin film delamination is the main failing mechanism for the integrated blocks of the microelectronic devices and the microelectronic packaging, inducing considerable economic losses in industries and manufacturing. Therefore, for a long time it has attracted much attention in both realms of mechanics and material science[1, 2]. Recently, some new and important phenomena have been observed, especially in crack propagation along the interface of a metal/ceramic system. These phenomena cannot be explained by the conventional elastic-plastic theory. For example, the peak separation stress ahead of an interface crack tip and at the micron level has been observed to be many times as high as the yielding stress of metal material in a crack propagation along the metal/ceramic interface[3]. However, the corresponding value is only 3 or 4 times that of the yielding stress according to the classical elastic-plastic theory[4, 5]. In ref. [5], the nonlinear delamination for thin films has been studied in detail by using the conventional elastic-plastic theory. The result shows that the peak separation stress during film delaminating can never surpass 4 times the yielding stress. The new phenomena are a challenge to the effective scope of conventional elastic-plastic theory. Therefore, the research on thin film delaminating is of both economic and scientific significance.

Recently, some new strain gradient plasticity theory has been advanced. This theory takes into consideration the material microscale effects in its complete and rigorous theoretical frame[6]. In the new theory, the strain gradient terms are matched with the classical strain terms by introducing a length scale. The length scale characterizes the strain gradient strength on the material points. Experiments on the micro-bending[7] and the micro-indentation[8] show that the length scale ranges 0.1—5 μm, so it is often called micro-length scale. In ref. [9], the strain gradient incremental theory has been further developed, and used to study the steady-state crack growth in mode I. The result shows that the peak interface separation stress ahead of crack tip can reach a value over ten times the material yielding stress.

In the present study, the metal thin film delamination along the interface of metal/ceramic is analyzed by employing the incremental theory of strain gradient plasticity and considering the microscale effects. Two models of fracture process frequently used in the elastic-plastic fracture researches are adopted: the embedded process zone model (EPZ model)[10] and the dislocation-free zone model (SSV model)[11]. Finally, by fitting the analysis results to the experimental ones on the Cu/SiO_2 bonded system, the peak separation stress near interface crack tip, the micro-length scale, and the dislocation-free zone thickness are predicted.

1 Fundamental descriptions of thin film delamination

For the metal thin film delamination in the metal/ceramic bonded system, the metal thin film and the ceramic substrate can be treated as an elastic-plastic material and elastic material, respectively. The delamination of a thin film (or interface crack growth) is caused by its own residue stress (σ_R). In the case of the steady-state interface crack growth——a frequently discussed and concerned case, an active plastic zone is produced near the crack tip and moves forward with it. An unloading zone is formed and swept by the active plastic zone, as shown in fig. 1(a). In the case of plane strain, the total energy per unit length of crack advance (energy release rate) during the thin film delaminating can be expressed as

$$G_{\text{crit}} = \frac{1 - v^2}{2} \frac{h\sigma_R^2}{E}, \tag{1}$$

where E and v are Young's modulus and Poisson's ratio for the metal thin film, respectively. Generally, in the analysis of the elastic-plastic crack growth, two fracture process models, i.e. EPZ model and SSV model, are used. These models are sketched in fig. 1(b). The energy release rate can be separated into two parts in the analysis by employing these models

$$G_{\text{crit}} = \Gamma_0 + \Gamma_P, \tag{2}$$

where Γ_0 is the interface fracture toughness (or interface crack separation energy) and Γ_p is the sum energy dissipated by the nonlinear deformations, including a part absorbed by active plastic zone and the other part dissipated in unloading zone.

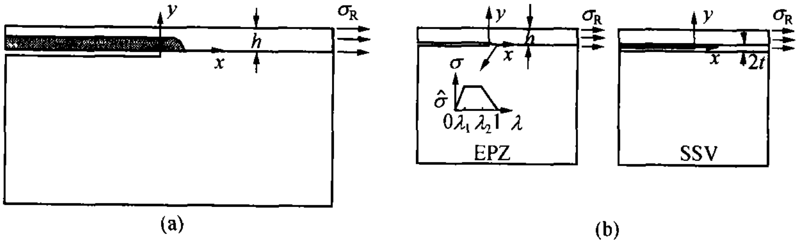

Fig. 1. (a) Delamination of the metal thin film/ceramic bonding system. Active plastic zone and unloading zone; (b) two models (EPZ model and SSV model) commonly used to study elastic-plastic crack growth ($\hat{\sigma}$ is the peak separation stress near the crack tip).

1.1 EPZ model

The relation between the cohesive force (or separation stress) and the crack opening displacement in fracture process zone is sketched in fig. 1(b). A single dimensionless separation measure describing the crack opening displacements in a fracture process zone is defined as

$$\lambda = \sqrt{(\delta_n/\delta_n^c)^2 + (\delta_t/\delta_t^c)^2}, \tag{3}$$

where δ_n and δ_t are normal and tangential relative opening displacements at crack faces, respectively. Let δ_n^c and δ_t^c denote the critical values of correspondent components. Then $\lambda = 1$ corresponds to the critical state of interface crack growth. A potential defined as[12]

$$\Phi(\delta_n, \delta_t) = \delta_n^c \int_0^\lambda \sigma(\lambda')\mathrm{d}\lambda' \tag{4}$$

is used to derive the normal and tangential components of the traction acting on the interface in the fracture process zone. The relations are given by

$$T_n = \frac{\partial \Phi}{\partial \delta_n} = \frac{\sigma(\lambda)}{\lambda}\frac{\delta_n}{\delta_n^c}, T_t = \frac{\partial \Phi}{\partial \delta_t} = \frac{\sigma(\lambda)}{\lambda}\frac{\delta_t}{\delta_t^c}\frac{\delta_n^c}{\delta_t^c}. \tag{5}$$

When $\lambda = 1$, from (4) and fig. 1(b), the fracture toughness is obtained

$$\Gamma_0 = (1/2)\hat{\sigma}\delta_n^c(1 + \lambda_2 - \lambda_1). \tag{6}$$

In finite element calculation, we usually take $\delta_t^c = \delta_n^c$. According to previous researches[12, 5], the model parameters (λ_1, λ_2) are insensitive to the final results. So in this research we take $\lambda_1 = 0.15$ and $\lambda_2 = 0.5$. Therefore, there are two important and independent parameters for EPZ model, $(\Gamma_0, \hat{\sigma})$. Another important parameter δ_n^c in EPZ model can be expressed by the above two independent parameters of (6).

1.2 SSV model

For SSV model, as shown in fig. 1(b), the fracture toughness Γ_0 can be determined by calculating the J-integral. The integration paths are taken within the dislocation-free layer and arround crack tip. There are also two important and independent parameters for SSV model, namely (Γ_0, t).

1.3 Relationship between energy and parameters in thin film delaminating

To reduce the number of parameters involved in the present research, the effect of the modulus mismatch between thin film and substrate is neglected, so we let $E_s = E$ and $v_s = v$. The tensile stress-strain relation is taken as

$$\varepsilon = \sigma/E, \text{ for } \sigma \leqslant \sigma_Y; \ \varepsilon = (\sigma_Y/E)(\sigma/\sigma_Y)^{1/N}, \text{ for } \sigma > \sigma_Y, \tag{7}$$

where N is the strain-hardening exponent and σ_Y the yielding stress. For metal thin film, the residue stress σ_R is equal to the material yielding stress σ_Y[2]. Therefore, the relation between the normalized energy and parameters (including the material parameters, model parameters and micro-length scale parameter) can be written as

$$\begin{aligned}\frac{G_{\text{crit}}}{\Gamma_0} = \frac{1}{6\pi}\frac{h}{R_0} &= f\left(\frac{E}{\sigma_Y}, v, N, \frac{\hat{\sigma}}{\sigma_Y}, \frac{l}{R_0}\right) \quad (\text{for EPZ model}), \\ &= g\left(\frac{E}{\sigma_Y}, v, N, \frac{t}{R_0}, \frac{l}{R_0}\right) \quad (\text{for SSV model}),\end{aligned} \tag{8}$$

where a characteristic length scale

$$R_0 = \frac{E\Gamma_0}{3\pi(1 - v^2)\sigma_Y^2} \tag{9}$$

is defined, and its value is roughly equal to the height of active plastic zone in the small scale yielding case; l is micro-length scale defined in the strain gradient plasticity theory; h is the thickness of thin film. A detailed expression for energy-parameter relation (8) can be obtained through application of the incremental theory of strain gradient plasticity to the thin film delamination analysis.

For a detailed description of the strain gradient plasticity and its increment theory, the reader may refer to refs. [6, 9].

2 Numerical method for steady-state delamination

Consider the case where a semi-infinite length crack lies along the interface between a metal thin film and a ceramic substrate and propagates in steady-state. Because the strain gradient incremental constitutive equations are rate-independent, they can be transformed into a set of partial differential equations in full quantity form within the active plastic zone and a set of the linear stress-strain relations in full quantity form also within the elastic and unloading region[13]. In fact, for crack growth in steady state, any field variables appearing in rate form can be expressed as (taking plastic strain tensor as an example here)

$$\dot{\varepsilon}_{ij}^{p} = -\dot{a}\,\frac{\partial \varepsilon_{ij}^{p}}{\partial x_1}, \tag{10}$$

where $\dot{a}$ is the velocity of crack growth and x_1 is a coordinate embedded on the crack tip and moving forward with it. Substituting all such relations as (10) into the incremental constitutive equations, the partial differential equations with respect to x_1 are obtained, which characterize the stress-strain relation within the active plastic zone and are independent of the crack advance velocity $\dot{a}$.

The finite element method is used to solve the partial differential equations numerically. An effective approach is to design a special element strip in x_1 direction with constant height within the active plastic zone and unloading zone in order to make integration with respect to x_1 in the iterating and solving procedure. In strain gradient plasticity theory, since the second order displacement-derivative terms (or strain gradient terms) are included, generally speaking, the conventional finite element method based on the displacement continuous at nodes is no longer valid, and a compatible pure-derivative element is needed. However, for mode I crack growth in plastic case, a satisfactory result has been reached by adopting the isoparametric displacement element with nine nodes[9]. But in the case of thin film elastic plastic delamination the mode mixing Ψ_{tip} of stresses at crack tip tends to 0° (mode I case) very quickly with an increase in $\hat{\sigma}/\sigma_{\text{Y}}$ (for EPZ model) or R_0/t (for SSV model). Therefore, in the present study the isoparametric displacement element with nine-nodes is employed and the 2×2 Gauss specimen points are selected to integrate equations in the entire material region.

3 Results and discussions

Consider a copper thin film. The related material parameters are taken as $E/\sigma_{\text{Y}} = 300$, $\nu = 0.3$, $N = 0.1$. The variations in normalized energy G_{crit}/Γ_0 with the increase in model parameters $\hat{\sigma}/\sigma_{\text{Y}}$ (for EPZ model) and R_0/t (for SSV model), as well as the micro-length scale l/R_0 are investigated here. G_{crit}/Γ_0 is the ratio of the total energy per unit length of crack advance to the interface fracture toughness during thin film delamination. Through application of the analysis results to the experiment for Cu/SiO$_2$, the model parameters ($\hat{\sigma}$, t, l) are predicted.

The shape and size of the active plastic zone near the interface crack tip are calculated and shown in fig. 2. In these calculations, both EPZ model and SSV model are adopted and quite similar results are obtained, so only the EPZ model results are shown in the figure. The results of a typical strain gradient case ($l/R_0 = 0.3$) and the conventional case ($l/R_0 = 0$) are given for comparison. By fig. 2, the strain gradient effect is insignificant, and the active plastic zone size exceeds half of the thin film layer thickness. Therefore, the metal thin film delamination occurs in a large scale yielding case.

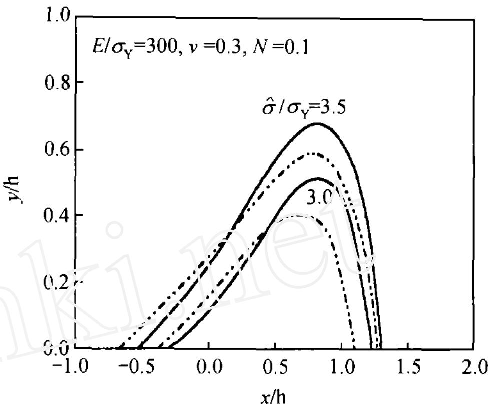

Fig. 2. Active plastic zone near interface crack tip. —, $l/R_0 = 0.0$, ---, $= 0.3$. $\hat{\sigma}/\sigma_Y = 3.5$.

The variations in normalized energy with the increases in the peak separation stress ($\hat{\sigma}/\sigma_Y$) for different values of micro-length scale from EPZ model are given in fig. 3(a). $\hat{\sigma}/\sigma_Y$ rises as micro-length scale increases (i.e. as strain gradient strength increases). For example, when normalized energy is equal to ten and $l/R_0 = 1.0$, $\hat{\sigma}/\sigma_Y$ increases by about 2.5 times that predicted from conventional theory. The larger the peak interface separation stress, the larger the critical energy release rate G_{crit} is obtained and the larger the critical thin film thickness h_{crit} is reached from (8), when residue stress in thin film is kept constant (fig. 3(a)).

The variations in normalized energy with the decrease in the dislocation-free zone thickness (t) for several different values of micro-length scale from SSV model are shown in fig. 3(b). Evidently, the results obtained from both EPZ model and SSV model are very similar to each other through comparing fig. 3(b) with 3(a), and the effects of model parameters R_0/t (for SSV model) and $\hat{\sigma}/\sigma_Y$ (for EPZ model) are also very similar to each other. If replacing R_0/t by $2\hat{\sigma}/\sigma_Y$ in fig. 3(b), the results of fig. 3(a) can be reproduced approximately. Fig. 3(b) shows that with diminishing t, the energy release rate and the interface strength increase. Therefore, the stronger the interface, the smaller the dislocation-free zone size. From (8), the smaller the t

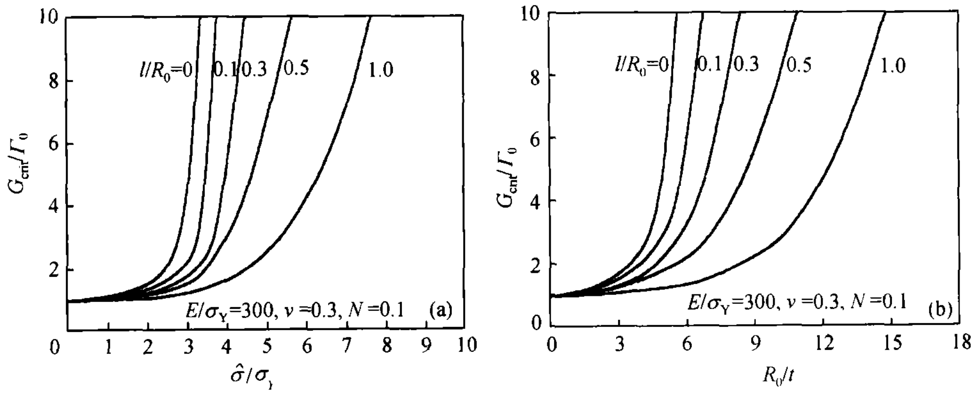

Fig. 3. Parameter relations between energy release rate with peak separation stress ((a) for EPZ model) or with dislocation-free zone thickness ((b) for SSV model).

value, the thicker the critical thin film, when the residue stress in the thin film is kept constant.

4 Experimental result on Cu/SiO$_2$ and prediction of micro-length scale

By combining the parameter results shown in fig. 3(a) and 3(b) with an experimental result which will be introduced in this section, the parameters ($\hat{\sigma}$, t) of the EPZ model and the SSV model, and the micro-length scale l are predicted.

A lot of experimental evidence for the metal thin film/ceramic substrate bonded system indicates that[1, 2, 14] the residue stress (or yielding stress) in thin film is inversely proportional to the thin film thickness. The residue stress of thin film is related with thin film thickness roughly by an inversely exponential relation. The experimental result of Cu/SiO$_2$ system by Bagchi and Evans[14] is used here. The variation in yielding stress of copper thin film with the film thickness is shown in fig. 4(a). In the figure a simulation of the experimental data is also shown by a straight line, which is approximately related with the thin film thickness

$$\sigma_Y = \sigma_Y^0 [1 + \sqrt{h_0/h_{Cu}}], \tag{11}$$

where the simulation parameters (σ_Y^0, h_0) are given in fig. 4(a). Substituting (11) into the first formula of (8) gives

$$\frac{G_{crit}}{\Gamma_0} = f\left(\frac{E}{\sigma_Y}, v, N\frac{\hat{\sigma}}{\sigma_Y^0[1 + \sqrt{h_0/h_{Cu}}]}, \frac{l}{R_0}\right). \tag{12}$$

By (12) and fig. 3(a), the curves are calculated in four cases, as shown in fig. 4(b). In the figure are also given the experimental results of ref. [14], which are obtained using two different interface techniques for the thin film/substrate bonding——the evaporation and diffusion bonding techniques. By the latter method a strong metal/ceramic interface is obtained from experimental data in fig. 4(b).

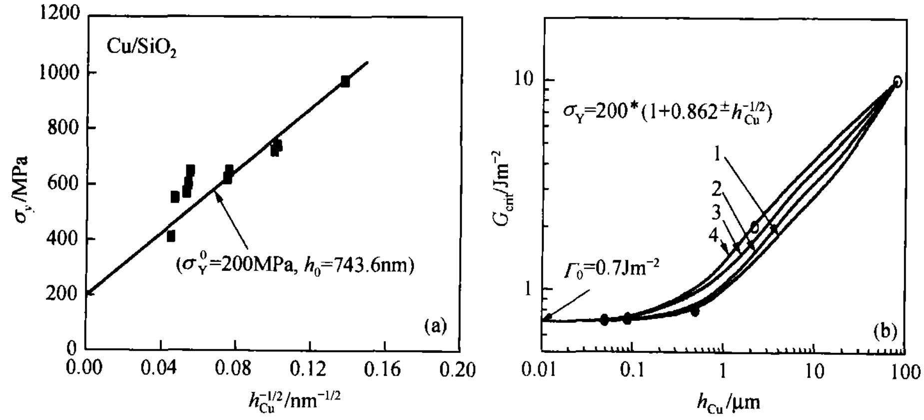

Fig. 4. (a) Variations in yielding stress vs. thin film thickness. Experimental resutls and simulation result; (b) variation of the energy release rate vs. thin film thickness. Comparison of prediction results with experimental results. 1, ($\hat{\sigma}$, l/R_0) = (0.76, 0.0); 2, 1.00, 0.3; 3, 1.24, 0.5; 4, 1.75; 1.0. ●, Evaporation technique[14]; ○, diffusion bonding technique[14].

Via comparison of the predicted curves with the experimental data points in fig. 4(b), we can draw the following conclusions:

1) When critical thin film thickness is small ($h_{\mathrm{Cu}} \leqslant 0.1\ \mu\mathrm{m}$), the energy release rate is insensitive to the thin film thickness and the interface strengths, and the energy release rate is lower and the contributed part from plastic deformation can be neglected.

2) With the increase in the thin film thickness, the relation of the peak interface strength $\hat{\sigma}$ with the micro-length scale l/R_0 is approximately a straight line, as shown in fig. 5. Neglecting the strain gradient effect that corresponds to a weak interface, with the increase in l, the peak separation stress increases. The conclusion is consistent with that from mode I when crack propagates in steady-state[9].

3) When the strain gradient effect is considered, the curve $G_{\mathrm{crit}} \sim \mathrm{Log}_{10} h_{\mathrm{Cu}}$ has a small curvature.

Through comparison, the peak separation stress and the micro-length scale matching the experimental resutls are obtained approximately

$$\hat{\sigma} \approx 1.75\ \mathrm{GPa}, \quad l/R_0 \approx 1.0.$$

Furthermore, taking $E/\sigma_{\mathrm{Y}} = 300$, $\nu = 0.3$, $\sigma_{\mathrm{Y}} = 300$ Mpa ($h_{\mathrm{Cu}} > 2\ \mu\mathrm{m}$), we have

$$l \approx 0.1\ \mu\mathrm{m}.$$

Clearly, the micro-length scale for thin film delamination discussed in the present study is smaller than that obtained by micro-bending[7] and micro-indentation experiments[8].

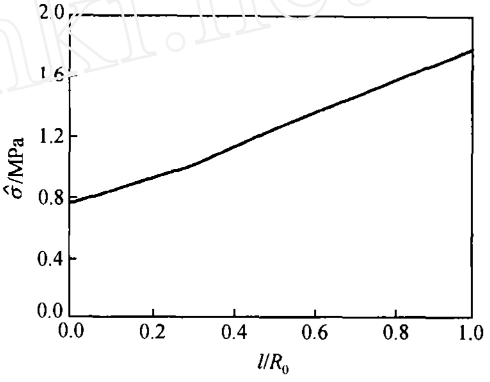

Fig. 5. Relation of peak separation stress with the micro-length scale.

Considering that the parameter relation of $G_{\mathrm{crit}}/\Gamma_0$ with l/R_0 should be independent of the employed models, and comparing fig. 3(a) with 3(b), the dislocation-free zone size can be taken as

$$2t \approx \frac{R_0}{\hat{\sigma}/\sigma_{\mathrm{Y}}} \approx 0.017\ \mu\mathrm{m}.$$

This value is of the same order of magnitude as that predicted by the self-consistent method in ref. [15].

5 Concluding remarks

In this paper, the nonlinear delamination for metal thin film along the interface of metal/ceramic system has been analyzed in detail by using the microscale mechanics approach. By combining the theoretical results with the experimental resutls on Cu/SiO$_2$ system, the peak interface separation stress and the dislocation-free zone thickness, as well as the micro-length scale have been predicted.

The nonlinear delamination for thin film is a typical case where the micro-length scale effect is prevailing. During thin film delaminating, some new and important phenomena caused by the microscale effect have been observed in experiments. Under the action of the mismatch residue stress, metal thin films with small thickness (micro, sub-micro, etc.) are bonded or deposited on the ceramic substrates. The formation of a strong (or a weak) interface of metal/ceramic joint depends on the bonding technique employed. During delamination, the metal thin film undergoes a large scale yielding deformation due to the strong interface separation stress. The conventional elastic-plastic theory is unable to explain satisfactorily such a phenomenon. Recently, Lipkin et al.[3] have studied the interface properties of the metal/ceramic bonded system experimentally. They observed that using different interface bonding techniques, the change in the interface fracture toughness Γ_0 is negligible. However, the increases of energy release rate G_{crit} may differ by as much as 100 times between a weak interface case and a strong interface case. This phenomenon implies that the peak separation stress $\hat{\sigma}$ increases greatly, even 3 or 4 times as much as that predicted by classical theory. Obviously, on the one hand, it is necessary to employ the two interface parameters to describe the nonlinear thin film delamination. On the other hand, from analysis in the last section, the change in interface peak stress $\hat{\sigma}$ depends on the strain gradient strength directly, which can be described by micro-length scale l. Therefore, it is necessary to use the strain gradient plasticity theory in the study of the thin film nonlinear delamination.

Acknowledgements This work was supported by the National Natural Science Foundation of China (Grant No. 19891180-01) and the Fundamental Research Project from the Chinese Academy of Sciences.

References

1. Nix, W. D., Mechanical properties of thin films, Metall. Trans., 1989, 20A: 2217.
2. Vinci, R. P., Zielinski, E. M., Bravman, J. C., Thermal strain and stress in copper thin films, Thin Solid Films, 1995, 262: 142.
3. Lipkin, D. M., Clarke, D. R., Evans, A. G., Effect of interfacial carbon on adhesion and toughness of gold-sapphire interfaces, Acta Mater., 1998, 46(13): 4835.
4. Tvergaard, V., Hutchinson, J. W., The relation between crack growth resistance and fracture process parameters in elastic-plastic solids, J. Mech. Phys. Solids, 1992, 40(6): 1377.
5. Wei, Y., Hutchinson, J. W., Nonlinear delamination mechanics for thin films, J. Mech. Phys. Solids, 1997a, 45(7): 1137.
6. Fleck, N. A., Hutchinson, J. W., Strain gradient plasticity, Advances in Applied Mechanics (eds. Hutchinson, J. W., Wu, T. Y.), 1997, 33: 295.
7. Stolken, J. S., Evans, A. G., A micro-bend test method for measuring the plasticity length scale, Acta Mater., 1998, 46 (14): 5109.
8. Begley, M. R., Hutchinson, J. W., The mechanics of size-dependent indentation, J. Mech. Phys. Solids, 1998, 46(10): 2049.
9. Wei, Y., Hutchinson, J. W., Steady-state crack growth and work of fracture for solids characterized by strain gradient plasticity, J. Mech. Phys. Solids, 1997b, 45(8): 1253.
10. Needleman, A., A continuum model for void nucleation by inclusion debonding, J. Applied Mechanics, 1987, 54: 525.
11. Suo, Z., Shih, C. F., Varias, A. G., A theory for cleavage cracking in the presence of plastic flow, Acta Metall. Mater., 1993, 41(5): 151.
12. Tvergaard, V., Hutchinson, J. W., The influence of plasticity on mixed mode interface toughness, J. Mech. Phys. Solids, 1993, 41(6): 1119.
13. Dean, R. H., Hutchinson, J. W., Quasi-static steady crack growth in small scale yielding, in Fracture Mechanics, 1980, ASTM STP700, 383.
14. Bagchi, A., Evans, A. G., The mechanics and physics of thin film decohesion and its measurement, Interface Sci., 1996, 3: 169.
15. Beltz, G. E., Rice, J. R., Shih, C. F. et al., A self-consistent model for cleavage in the presence of plastic flow, Acta Mater., 1996, 44(10): 3943.