COMPOSITES SCIENCE AND TECHNOLOGY

Composites Science and Technology 59 (1999) 283–289

Microstructural engineering of ceramic-matrix layered composites: Effect of grain-size dispersion on single-phase ceramic strength

M. Lugovy [a,*], N. Orlovskaya [a], K. Berroth [b], J. Kuebler [b]

[a] *Institute for Problems of Materials Science, National Academy of Sciences of Ukraine, 3 Krzhizhanovsky str., 252680 Kiev, Ukraine*
[b] *Swiss Federal Laboratories for Materials Testing and Research, EMPA, Duebendorf, Ueberlandstrasse 129, CH-8600, Switzerland*

Received 17 June 1997; received in revised form 4 March 1998; accepted 12 March 1998

Abstract

A failure model is considered which can be applied to n-phase brittle materials (in particular to ceramics). The authors attempt to solve a physical problem of the description of failure of a micro-inhomogeneous solid as stochastic process of the cracking of separate structural elements. The particular model is executed for n = 1 (case of single-phase ceramics) and is applied for the description of the mechanical behaviour of single-phase ceramic layers with various statistical distributions of the grains sizes. An effective continuum and a statistical description of the failure process is used. There are four stages in the process. The first stage is loading without microcracking and the second stage is a stable non-localized microcracking before maximum stress. The third stage is a stable localized microcracking after stress maximum and the fourth stage is an unstable (catastrophic) fracture. The trends of the theoretical calculations agree with data obtained from acoustic emission studies.

Keywords: A. Ceramics; B. Fracture; C. Computational simulation; C. Probabilistic methods; D. Acoustic emission

1. Introduction

Ceramic materials show many outstanding physical and chemical properties which make them interesting materials for different engineering purposes. However, their more intensive technical application is restricted by their brittleness.

The strategy of ceramic strengthening is usually associated with the design of ceramic composites that can enable the operation of different mechanisms of fracture energy absorption and/or dissipation. In addition to these mechanisms, other properties can also be modified by suitable microstructural design of composites, including those which cannot be combined in a single-phase material. In functional gradient materials even more complex microstructural architectures have to be developed. Different structural elements, such as particles, whiskers, filaments, platelets or laminae, and others have been combined to tailor properties in accordance with different service requirements.

Composites are complicated systems which are characterized by a host of interdependent parameters. The theoretical prediction of mechanical behaviour provides information related to the failure of such materials. The strength of ceramic-matrix layered composites depend on the strength of the ceramic-matrix layer.

The goal of the work proposed is to study the interrelationships between structure, mechanical characteristics and fracture behaviour of the individual layers of complex particulate-layered composites. It is necessary for an optimal design and processing parameters for fabrication of the composites.

For this purpose a failure model is considered which can be applied to *n*-phase brittle materials (in particular to ceramics). The authors attempt to solve a physical problem of the description of failure of a micro-inhomogeneous solid as a stochastic process of cracking of the separate structural elements. This particular model is executed for $n = 1$ (case of single-phase ceramics), and is applied for the description of the mechanical behaviour of a single-phase ceramic layer with various statistical distributions of grains sizes. An effective continuum and a statistical description of the failure process is used.

* Corresponding author. E-mail: lugovoj@ipms.kiev.ua.

0266-3538/99/$—see front matter
PII: S0266-3538(98)00070-0

2. Microstructural fracture model of ceramic-matrix layer

We define the structural element of a composite material as a homogeneous volume of material with given properties. The elements have interfaces i.e. the surfaces where the material properties experience sudden changes. Characteristics of the individual structural elements, the form of statistical distribution, and loading parameters are used as the initial data for calculating the volume fraction of the fractured structural element. An arbitrary structural element is adopted for the effective continuum. Anticipation of volume of the fractured structural elements and probability of an individual failed element are calculated with due consideration being given to the local failure criterion. The probability of an individually failed element is assumed to be proportional to the relative number of the fractured structural elements. The local failure criterion is selected to coincide with the energetic approach of crack nucleation [1,2].

2.1. Initial data for the model

The important parameter determining redistribution of stresses between structural elements is the modulus of elasticity of the material, E. The main loading parameter of the model is the density of elastic strain energy in the material, $q = \frac{1}{2}\sigma \cdot e$, where $\sigma = E(e)e$ (the elastic modulus, E, depends on strain, e, in the general case). Note that σ and e are the intensity of the applied stress and the intensity of deformation.

Characteristics of the individual structural elements also affecting stress redistribution are the moduli of elasticity, E_j, and the volume fraction, f_j, of each component of composite. The strength of the individual structural element is determined with the energy of formation of new surface unit square γ_j of a given component (this is the surface energy in the case of tension).

There are the effective residual stresses in structural elements of each component in multi-phase ceramics. During cooling, the difference in deformation due to the different thermal expansion coefficients of the components is accommodated by creep, as long as the temperature is sufficiently high. Below a certain temperature, this will be termed the 'joining' temperature, the different components become bonded together and internal stresses appear. In each phase, the total deformation after sintering, is the sum of an elastic component and of a thermal component. In the case of perfectly rigid bonding between the phases, the total deformation will be the same for all the phases, then:

$$e_j = \frac{\sigma_{rj}}{E_j} + \alpha_{Tj}\Delta T = \text{const}, \tag{1}$$

where σ_{rj} is the residual stress in j^{th} phase, α_{Tj} is the j-phase thermal expansion coefficient, ΔT is the difference in temperature between the actual temperature and the joining temperature.

The force balance requires (in normal stresses):

$$\sum \sigma_{rj} f_j = 0. \tag{2}$$

From Eqs. (1) and (2) for two-phase material, we obtain:

$$\sigma_{r1} = \frac{E_1 E_2 f_2 (\alpha_{T2} - \alpha_{T1})\Delta T}{E_2 f_2 + E_1 f_1} \tag{3}$$

and

$$\sigma_{r2} = \frac{E_2 E_1 f_1 (\alpha_{T2} - \alpha_{T1})\Delta T}{E_2 f_2 + E_1 f_1}. \tag{4}$$

For multi-phase material we have:

$$\sigma_{rj} = E_j \Delta T \frac{\sum_i E_i f_i (\alpha_{Ti} - \alpha_{Tj})}{\sum_i E_i f_i}. \tag{5}$$

It is necessary to determine the statistical distribution of structural element size for the model calculations. This distribution depends strongly on several factors that are affected by the fabrication process. Generally the logarithmic normal distribution of structural element sizes is used. The density of such distribution is

$$f(l_j) = \frac{1}{l_j D_j \sqrt{2\pi}} \exp\left(-\frac{(\ln\ l_j - M_j)^2}{2D_j^2} \right), \tag{6}$$

where l_j is the structural element size, M_j is the expectation of ln l_{mj}, l_{mj} is the average size of j-phase element, D_j is the dispersion of distribution. We use the distribution parameter $l_{\max\ j}/l_{m1}$ and condition $f(l_{\max\ j}) = 0.0001$ where $l_{\max\ j}$ is the effective maximum size of structural element of j-phase. If it is known the parameters M_j and D_j can be obtained from Eq. (6).

The normalized variables is used in the work. The normalized density of strain energy is $q^* = q l_{m1}/\gamma_1$, where l_{m1} is the average size of element of phase called first, γ_1 is the surface energy of its phase. The normalized modulus of elasticity of each component of composite is $E_j^* = E_j/E$. The normalized residual stress in the j phase is $\sigma_{rj}^* = \sigma_{rj}\sqrt{l_{m1}/E\gamma_1}$. The sizes of structural elements are normalized by l_{m1}. All model stresses are normalized, $\sigma^* = \sigma\sqrt{l_{m1}/E\gamma_1}$. All model deformations are normalized, $e* = e\sqrt{El_{ml}/\gamma_1}$.

2.2. Basic equations of model

The probability of arbitrary structural element failure for the j-phase can be calculated if the statistical distribution density $f_{ij}(s_{ij})$ of the structural parameters s_{ij} are known. These parameters are mutually independent, i.e.

$$P_j^{(\mathrm{f})} = \frac{n_j^{(\mathrm{f})}}{n_j} = \int\limits_{(q_j^c<\alpha_j q)} f_{1j}(s_{1j})\ldots f_{kj}(s_{kj})\mathrm{d}s_{1j}\ldots \mathrm{d}s_{kj}, \quad (7)$$

where k is the total number of s_{ij} parameters, $n_j^{(\mathrm{f})}$ is the number of the fractured elements of the j phase, n_j is the number of j-phase structural elements, $q_j^c < \alpha_j q$ is one of the possible forms of the local criterion of failure (criterion of the individual grain fracture), q_j^c is the critical density of strain energy depending on the structural parameters s_{ij}, α_j characterizes the redistribution of the strain energy density between the j-phase element and the effective continuum. Generally the coefficient α_j is related to the redistribution of stresses. Assume that α_j is approximately equal to the ratio of elastic moduli of the j phase and continuum [3]. The fractured structural elements can be represented by pores as a first approximation. In this case, if the strain of a j phase element and the strain of the continuum are same the modulus of effective continuum can be described by the expression $E(1-f_\mathrm{f})$ according to a rule of mixtures assuming that porosity can be replaced by the volume fraction of the fractured structural elements, f_f. Hence, $\alpha_j = E_j/E(1-f_\mathrm{f})$ that corresponds to the additive nature of energy.

The probability of failure of the structural element with size $l_j < l$ is given by

$$P_j^{(\mathrm{fl})} = \int\limits_{\begin{cases} q_j^c < \alpha_{jq} \\ l_j < l \end{cases}} f_{1j}(s_{1j})\ldots f_{kj}(s_{kj})\mathrm{d}s_{1j}\ldots \mathrm{d}s_{kj}. \quad (8)$$

It is evident that $P_j^{(\mathrm{fl})} = n_j^{(\mathrm{fl})}/n_j$ where $n_j^{(\mathrm{fl})}$ is the number of j-phase failed structural elements with size $l_j < l$. Then the probability of event that the fractured element of the j phase has the size $l_j < l$ is:

$$P_j^{(l)} = \frac{n_j^{(\mathrm{fl})}}{n_j^{(\mathrm{f})}} = \frac{n_j^{(\mathrm{fl})}}{n_j}\frac{n_j}{n_j^{(\mathrm{f})}} = \frac{P_j^{(\mathrm{fl})}}{P_j^{(\mathrm{f})}}.$$

The expectation of the volume of the fractured structural element for the j phase is defined as [4]:

$$\xi_j = k_j \int\limits_0^\infty l^3 \frac{\partial P_j^{(l)}}{\partial l}\,\mathrm{d}l, \quad (9a)$$

where k_j is the coefficient depended on the shape of the j-phase structural elements. The expectation of the j-phase element volume is

$$MV_j = k_j \int\limits_0^\infty s_{1j}^3 f_{1j}(s_{1j})\,\mathrm{d}s_{1j}, \quad (9b)$$

where $s_{1j} = l_j$ [4].

The parameter q_j^c for a brittle material can be obtained from an energy balance consideration given by

$$q_j^c = \frac{K_j\gamma_j}{l_j} \pm \frac{\sigma_{rj}^2}{2E_j}, \quad (10)$$

where K_j is the geometrical coefficient of structural elements ($K_j=3$ for spherical elements and transgranular cracks). The second term in Eq. (10) is the elastic energy of residual stresses. It is positive for external tensile load and the compressive internal stress and negative if the internal stress is positive.

The volume fraction of the fractured structural elements is given by

$$f_\mathrm{f} = \sum_j \frac{\xi_j f_j P_j^{(\mathrm{f})}}{MV_j}. \quad (11)$$

If geometrical size of a structural element is the random parameter and other structural parameters are determinate, Eq. (11) can then be transformed to the following form:

$$f_\mathrm{f} = \sum_j f_j \frac{\int_{l_j^*}^\infty l_j^3 f_j(l_j)\,\mathrm{d}l_j}{\int_0^\infty l_j^3 f_j(l_j)\,\mathrm{d}l_j}, \quad (12)$$

where l_j is the size of the structural element, $f_j(l_j)$ is the statistical distribution density of l_j and l_j^* is the minimal size of the fractured element. The minimal size value of the fractured element of the j phase l_j^* can be obtained from the equation $\alpha_j q = q_j^c$ and Eq. (10) when $l_j = l_j^*$.

2.3. Critical parameters of fracture

The dependence of the normalized strain energy density q^* on the state parameter f_f for single-phase brittle material with $l_{\max}/l_m = 3$ is shown in Fig. 1 (for single-phase material $l_{\max 1} = l_{\max}$, $l_1 = l$, $l_{m1} = l_m$). The calculation was performed for a polycrystalline material when l_{m1} is the average grain size. The curve related to an internal stress free material. The interfaces between the structural elements are assumed to be unfractured.

The curve in Fig. 1 determines the potential barrier in the f_f – state space for the transition of the material from non-broken state to the completely broken material. If the fractured structural elements exist initially in the material, the f_f – parameter would not contain volume fraction of the failed element. The maximum of the curve correspond to ultimate states of the material under load. The maximum value of the strain energy density is the critical strain energy density Q_c. The normalized value is $Q_\mathrm{c}^* = \frac{Q_\mathrm{c} l_m}{\gamma_1}$. Note that f_c is the volume fraction of the failed elements in the ultimate state. The stable states exist at $f_\mathrm{f} < f_\mathrm{c}$ whereas the unstable states correspond to $f_\mathrm{f} > f_\mathrm{c}$. There is the strain energy density

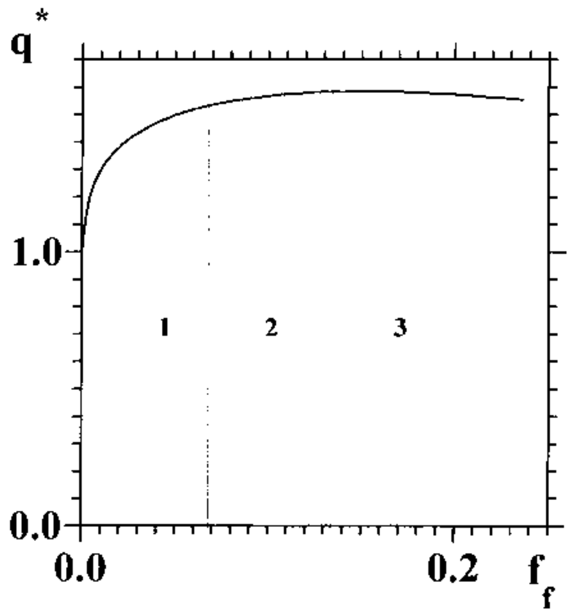

Fig. 1. The dependence of relative strain energy density on the volume fraction of fractured structural elements for a single phase material with $l_{max}/l_m = 3$: (1) stable non-localized microcracking stage; (2) stable localized microcracking stage; (3) unstable (catastrophic) fracture stage.

at initiation of microcracking q_0. The normalized value is $q_0^* = q_0 l_m/\gamma_1$. The spontaneous cracking of the structural elements occurs when $q_0 = 0$.

The dependence of the normalized stress on the normalized strain for a single phase material with $l_{max}/l_m = 3$ is shown in Fig. 2. There are four stages of process: the first is loading without microcracking; the second is a stable non-localized microcracking before stress maximum. The third stage is a stable localized microcracking after maximum stress, and the fourth stage is unstable (catastrophic) fracture. The durations of the third and fourth stages are very short time as a result of fracture localization. Hence, these stages are invisible in a practice. In Fig. 1 the stages of stable non-localized microcracking and stable localized microcracking are before the strain energy density maximum. The stage of unstable (catastrophic) fracture is after the maximum in Fig. 1.

The calculations are compared with acoustic-emission experimental data for a brittle ceramic material under load. The acoustic emission method determines the onset of element failure. In the general case, initiation does not correspond to the complete failure of the sample (Fig. 3 [5]). There is an interval of stable microcracking predicted by the present model. The local cracking before the failure at mechanical loading of ceramics really exists.

The critical strain-energy density, Q_c, is the specific volume value. The strain-energy release rate, G_c, is the specific surface value. The strain-energy release rate, G_c, is connected with the critical strain-energy density, Q_c, by the following ratio:

$$G_c = Q_c l_c,$$

where l_c is the size of an element of structure, which is fractured at the moment of transition from the local cracking to the catastrophic failure of a material. Here l_c is the characteristic linear scale on which occurs the process of failure.

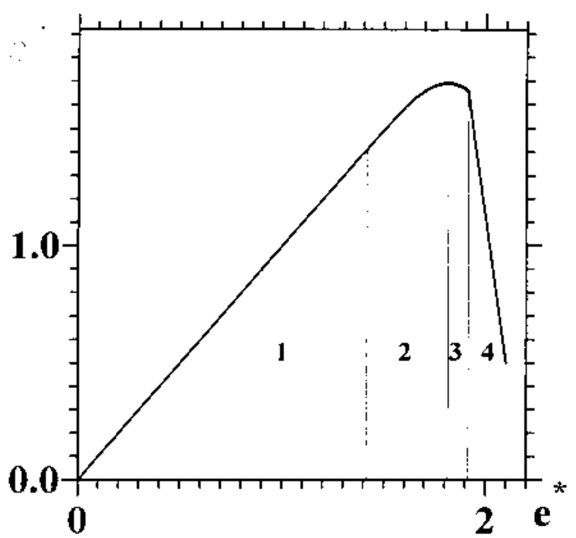

Fig. 2. The dependence of the normalized stress on the normalized strain for a single-phase material with $l_{max}/l_m = 3$: (1) loading without microcracking; (2) stable non-localized microcracking stage; (3) stable localized microcracking stage; (4) unstable (catastrophic) fracture stage.

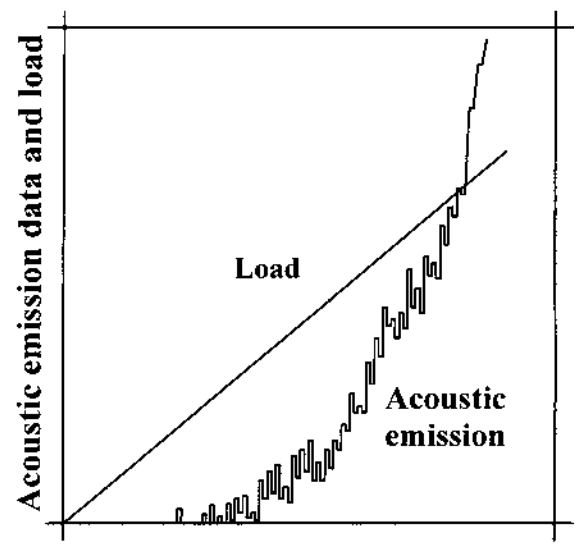

Fig. 3. Acoustic emission data of flexural test for porcelain bars [5].

3. Strength of ceramic-matrix layer: The effect of grain size dispersion on single-phase ceramic strength

The mechanical behaviour of single-phase ceramic layer is researched for the statistical distributions of normalized sizes l/l_m of structural elements with different l_{max}/l_m. The calculations were performed for a polycrystalline material when l_m is the average grain size. The materials suppose an internal stress free materials. The interfaces between the structural elements are assumed to be unfractured. The statistical distributions of normalized sizes l/l_m of structural elements for the materials are shown in Fig. 4.

The dependences of relative strain energy density on the volume fraction of fractured structural elements are presented in Fig. 5. The relative strain energy density increases when l_{max}/l_m decreases. The small structural elements are stronger than the big structural elements. It is resulted from the local criterion of failure and Eq. (10). The dependences of the normalized stress on the normalized strain are shown in Fig. 6. The stress maxima increase when l_{max}/l_m decreases.

The dependences of relative strain energy density at microcracking initiation q_0^*, at fracture termination Q_c^* and at stress maximum Q_{cm}^* on l_{max}/l_m are presented in Fig. 7. These parameters are same and maximum when there is no dispersion of structural element sizes. Then the difference between the parameters increase. For big l_{max}/l_m the difference decreases. If the difference is zero the total fracture is without microcracking. Hence, there is critical parameter l_{max}/l_m and the fracture is total catastrophical (from first crack) when the dispersion of grain sizes is more than critical value.

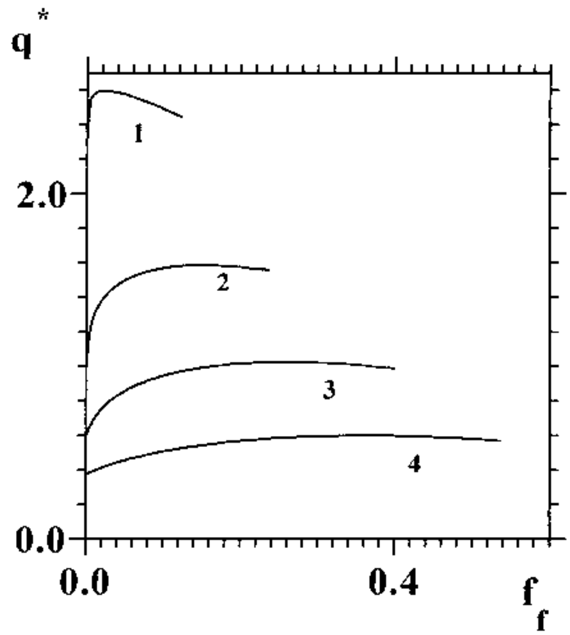

Fig. 5. The dependences of relative strain energy density on the volume fraction of fractured structural elements for a single-phase material: (1) $l_{max}/l_m = 1.3$; (2) $l_{max}/l_m = 3$; (3) $l_{max}/l_m = 5$; (4) $l_{max}/l_m = 8$.

The dependences of volume fraction of fractured structural elements at fracture termination f_c and at stress maximum f_{cm} on l_{max}/l_m are shown in Fig. 8. These dependences are non-monotonic. The parameters

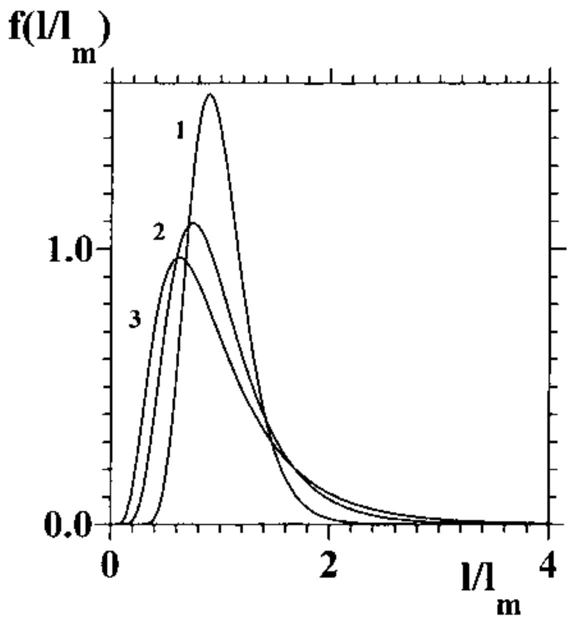

Fig. 4. The statistical distributions of normalized size l/l_m of structural elements for a single phase material: (1) $l_{max}/l_m = 3$; (2) $l_{max}/l_m = 5$; (3) $l_{max}/l_m = 7$.

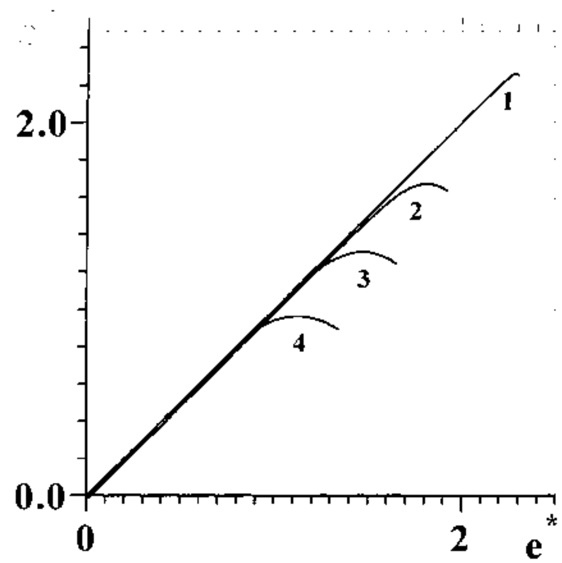

Fig. 6. The dependences of the normalized stress on the normalized strain for a single-phase material: (1) $l_{max}/l_m = 1.3$; (2) $l_{max}/l_m = 3$; (3) $l_{max}/l_m = 5$; (4) $l_{max}/l_m = 8$.

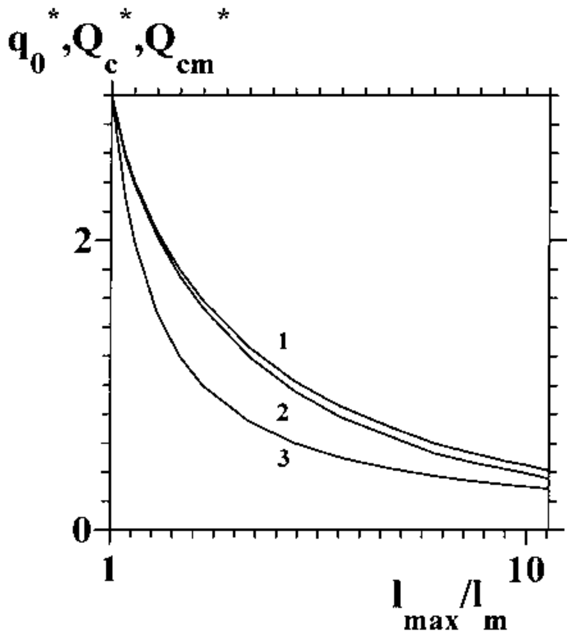

Fig. 7. The dependences of relative strain energy density at microcracking initiation q_0^*, at fracture termination Q_c^* and at stress maximum Q_{cm}^* on l_{max}/l_m: (1) Q_c^*; (2) Q_{cm}^*; (3) q_0^*.

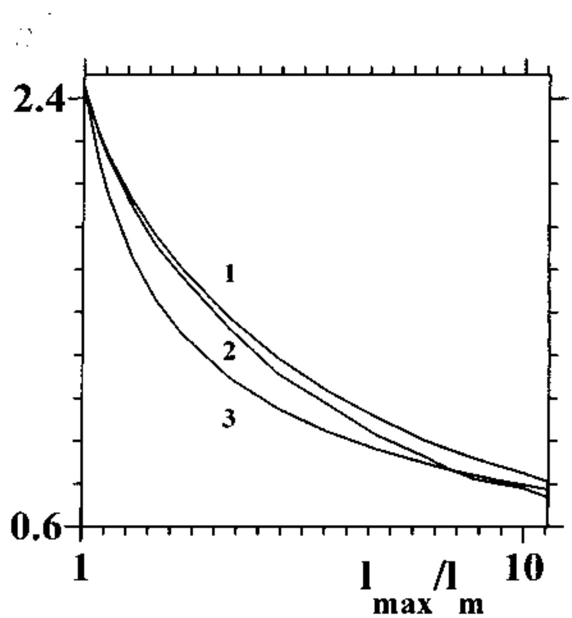

Fig. 9. The dependences of the normalized stress on l_{max}/l_m: (1) stress maximum; (2) at fracture termination; (3) at microcracking initiation.

f_c and f_{cm} are equal to zero when there is no dispersion of structural element sizes. These parameters increase when the dispersion of grain sizes is less than some value and decrease when the dispersion is more than this value.

The dependences of the normalized stress on l_{max}/l_m are presented in Fig. 9. The dependence of stress maximum, stress at fracture termination and stress at microcracking initiation are shown. These stresses are same and maximum when there is no dispersion of structural element sizes. Then the difference between the parameters increase. For big l_{max}/l_m the difference decreases. It is as same as for the relative strain energy density. The dependences of the normalized strain on l_{max}/l_m are shown in Fig. 10. The strain at fracture termination and at stress maximum are presented. These parameters decrease when l_{max}/l_m increases.

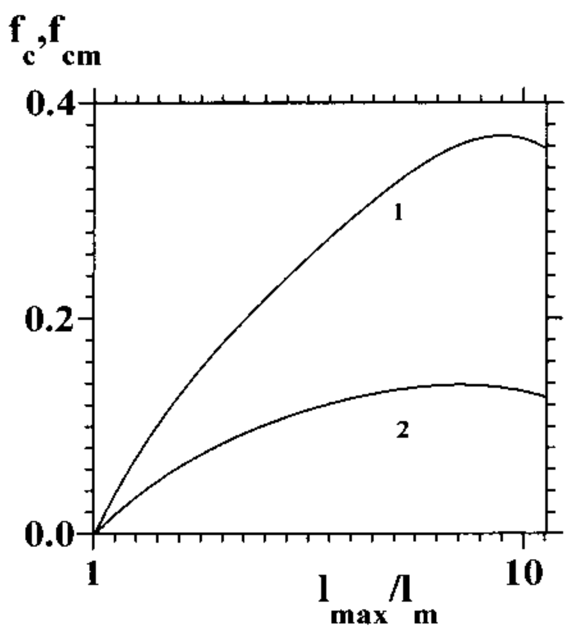

Fig. 8. The dependences of volume fraction of fractured structural elements at fracture termination f_c and at stress maximum f_{cm} on l_{max}/l_m: (1) f_c; (2) f_{cm}.

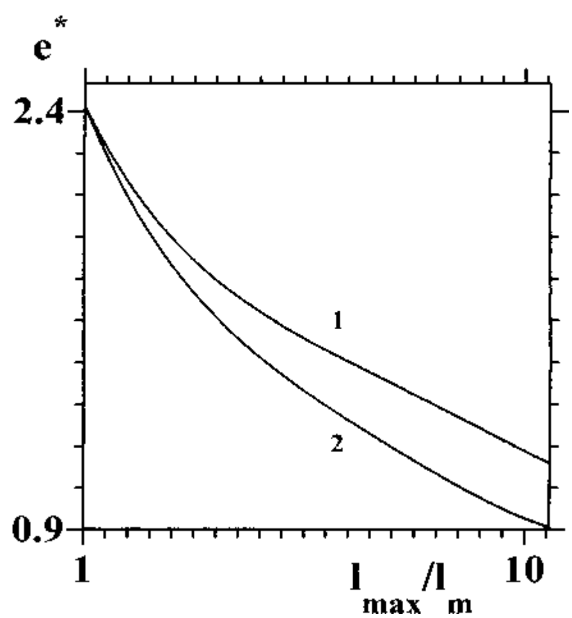

Fig. 10. The dependences of the normalized strain on l_{max}/l_m: (1) at fracture termination; (2) at stress maximum.

4. Conclusions

The model of failure was considered which can be applied to n-phase brittle materials (in particular to ceramics). The authors try to solve a physical problem of the description of failure of a microinhomogeneous solid as stochastic process of the cracking of separate structural elements. The particular modeling was executed for $n=1$ (case of single-phase ceramics). The above mentioned model was applied for the description of mechanical behaviour of single-phase ceramic layer with various statistical distributions of the grains sizes.

The proposed model provides prediction on the dependence of the critical parameters of fracture for single-phase and multi-phase solids with inhomogeneity and the mechanical behaviour of loaded structural elements.

There are four stages of process. First stage is loading without microcracking. Second stage is a stable non-localized microcracking before stress maximum. Third stage is a stable localized microcracking after stress maximum. Fourth stage is a unstable (catastrophic) fracture.

There is critical parameter $l_{\max}/l_m$. The fracture is total catastrophical (from first crack) when the dispersion of grain sizes is more than critical value.

Acknowledgements

This work was supported by Swiss National Science Foundation. It is the part of the Program of Cooperation in Science and Research with Central and Eastern European Countries and Newly Independent States (1996–1998), the Project ‘‘Ceramic-Matrix Layered Composites for Engineering and Tooling Application’’.

References

[1] Davidge RW, Green TJ. The strength of two-phase ceramic/glass materials. J Mater Sci 1968;3:629–34

[2] Lang FF. In: Broutman LJ, editor. Composite materials, fracture and fatigue, vol. 5. New York: Academic Press, 1974:1–44

[3] Schultrich B. Mechanical properties of brittle materials. Modern theories and experimental evidence: wiss. akad. der wissenschaften der DDR, zentralinstitut fur festkorperphysik und werkstofforschung 1986;N32:229–66

[4] Podrezov YuN, Lugovoy NI, Slyunyaev VN, Minakov NV. Statistical failure model of materials with micro-inhomogeneity. Theoretical and Applied Fracture Mechanics 1997;26:35–40

[5] Evans AG, Linzer M. Failure prediction in structural ceramics using acoustic emission. J Am Ceram Soc 1973;56:575–80