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On Microstructural Residual Stresses in Particle Reinforced Ceramics

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Abstract

Residual microstructural stresses are shown to influence the crack propagation in particle reinforced ceramics. The dispersion of second phase particles with different coefficients of thermal expansion generates a spatially fluctuating stress field. In this paper deterministic and statistical solutions are considered for the determination of residual stresses. Numerical results are presented for SiC-platelet reinforced Si_3N_4 *and correlated to mechanical properties. It is concluded that the toughening mechanisms of particle reinforced composites are influenced decisively by the spatially fluctuating stress field.*

Der Rißfortschritt in partikelverstärkten Verbundwerkstoffen wird entscheidend von den mikrostrukturellen Eigenspannungen bestimmt. Verschiedene Berechnungsverfahren zur Bestimmung der Eigenspannungen werden vorgestellt und mit grundlegenden Überlegungen zum Rißfortschritt in keramischen Verbundwerkstoffen verknüpft. Am Beispiel von SiC-Platelet verstärktem Si_3N_4 *wird der Einfluß von räumlich fluktuierenden Eigenspannungen auf die Verstärkungsmechanismen diskutiert.*

On montre ici que les contraintes résiduelles dans la microstructure jouent un rôle important dans la propagation des fissures pour les céramiques renforcées par des particules. La dispersion de ces particules comme seconde phase ayant un coefficient de dilatation thermique différent génère un champ de contraintes spatiales oscillantes. On rapporte dans cet article des solutions statistiques et précises pour définir le rôle des contraintes résiduelles. On présente des résultats numériques concernant les céramiques Si_3N_4 *renforcées par des plaquettes en SiC et la corrélation avec les propriétés mécaniques. Enfin, on a pu conclure que les mécanismes de propagation de fissures, de ces composites dépendent fortement du champ de contraintes spatiales oscillantes.*

1. Introduction

There are many projects aimed at the optimization of the microstructure of ceramics in order to obtain outstanding mechanical properties. Two different pathways can be distinguished. (1) Minimize the defect size by way of better manufacturing. Typically this leads to high demands with respect to purity and quality of the raw materials. Furthermore, advanced processing techniques like colloidal pressing and hot-isostatic pressing are required in order to get 'defect-free' structures. However, these materials are highly sensitive to loads exceeding tolerable limits and offer low reliability. (2) Tolerate certain defects but increase the reliability by the development of non-linear ceramics (which show non-linear fracture behaviour). This approach can be realized by the incorporation of second phase particles like platelets, whiskers or fibres. In order to do this effectively it is necessary to develop theoretical models of the relevant micromechanisms to supplement empirical and experimental research.

The relevant micromechanisms in particle reinforced ceramic composites can be classified as follows:[1,2]

—Crack tip mechanisms
—Crack bridging mechanisms
—Formation of a process zone (energy dissipation zone)

Normally the three mechanisms will act simultaneously, and a mutual amplification may occur (synergism). Because of this superposition of different mechanisms, the mechanical properties of reinforced ceramics exhibit relatively complicated dependencies on microstructural parameters. Especially the residual stress distribution affects all stages of the crack propagation process and the effect of toughening mechanism. Therefore, in the present work, the authors concentrate on the influence of residual stresses on crack propagation. Firstly the influence of residual stresses on the crack propagation process is discussed by considering some bounds. Then the principal methods for the calculation of residual stresses in heterogeneous materials are presented and some examples are given for SiC-platelet reinforced Si_3N_4. Finally the calculated residual stresses are correlated with mechanical data.

2 Influence of Internal Stresses on Crack Propagation

Considering particle reinforced ceramic composites an inhomogeneous stress field caused by the differential thermal expansion of the constituents must be expected. The stress field due to an external load will also be inhomogeneous because of the heterogeneity of the microstructure. When a crack propagates through such a fluctuating stress field, the following processes may occur:

—Crack deflection
—Crack deceleration or even crack arrest
—Crack acceleration

These principal possibilities of interaction between crack propagation and internal stresses will now be discussed in a general form. Without considering fracture surface energies and environmental conditions, the following general crack propagation equation[3] can be assumed:

$$\dot{a} = f(G) \tag{1}$$

Here $\dot{a}$ is the local crack velocity and G the local energy release rate. Both quantities fluctuate randomly according to the heterogeneous microstructure. The function f may be approximated for some problems by a simple step function; in general, it is a monotonously increasing function of G.

For a simple approximation it is assumed that the practically relevant average crack velocity is determined by the average of G:

$$\langle \dot{a} \rangle = f(\langle G \rangle) \tag{2}$$

Here the averaging has to be taken over the actual crack path, which is not flat and regular. In the following the averages for a certain characteristic volume in front of the crack tip are considered.

By using the well-known relationship $G = K^2/E$, where K denotes the stress intensity factor (for this treatise a global view is used and no distinction is made between K_{I}, K_{II}, K_{III}), then

$$\langle G \rangle = \langle K^2 \rangle / E = \langle (K^e + K^i + K^{ia})^2 \rangle / E \tag{3}$$

In this equation the stress intensity factor has been decomposed according to the superimposing stress fields:

K^e—caused by the external loading
K^i—caused by internal stresses (residual stresses due to the heterogeneity of the material)
K^{ia}—caused by interaction stresses which originate since the crack front is not straight

Now two limiting cases are considered:

(i) It is assumed that the crack randomly propagates through regions with tension and compression.

Generally:

$$\begin{aligned} \langle (K^e + K^i + K^{ia})^2 \rangle &\geqslant (\langle K^e + K^i + K^{ia} \rangle)^2 \\ &= (\langle K^e \rangle + \langle K^i \rangle + \langle K^{ia} \rangle)^2 \end{aligned} \tag{4}$$

Here it can be put that $\langle K^i \rangle \to 0$, since the crack prefers neither tension nor compression. Moreover it may be assumed that $\langle K^{ia} \rangle \to 0$, since overloaded and unloaded parts of the crack front should be balanced. Thus, from eqn (3) with eqn (4):

$$\langle G \rangle \geq \langle K^e \rangle^2 / E \tag{5}$$

That means the average energy release rate is increased in the presence of residual stresses and according to eqn (2) crack propagation is accelerated.

(ii) Picture a situation of a material with locally fluctuating tensile and compressive stress fields associated with locally fluctuating crack resistances. If, on average, compressive stress regimes are predominantly associated with low crack resistance domains, then a non-planar crack might be able to run favourably in compressive zones, hence $K^{i} < 0$. In this case it is possible that

$$K^{e} + K^{i} + K^{ia} \leq K^{e} \quad (6)$$

Then from eqn (3)

$$\langle G \rangle \leq \langle K^{e} \rangle^{2}/E \quad (7)$$

i.e. the average crack velocity decreases.

Of course, the two mechanisms discussed have to be analysed in more detail when they are applied for practical purposes. In particular one has to take into account the triaxiality of stresses, the form of the function f as well as the actual stress intensity factors, but already this simple approximation shows that residual internal stresses have a distinct influence on the crack propagation in heterogeneous materials. Therefore it is obvious for the understanding of reinforced composites that the interaction between crack propagation and residual stresses should be analysed very carefully.

3 Determination of Residual Stresses

The theory of internal stresses in heterogeneous solids is a very complex topic which cannot be discussed in the scope of this paper. In the following, the basic equations of continuum mechanics and some solutions which are relevant for the calculation of particle reinforced ceramics are summarized. An extensive review may be found in Ref. 4, for example.

Generally the equilibrium condition must hold for the stress field:

$$\operatorname{div} \sigma(x) = 0 \quad (8)$$

The properties of the material appear in the thermoelastic stress–strain relation

$$\sigma = C(\varepsilon - \varepsilon^{T}) \quad (9)$$

where C is Hook's tensor of the elastic constants. The thermal strain ε^{T} is determined by the thermal expansion coefficients α (which may depend on temperature T) and possibly by a transformation strain ε^{tr}, which may occur in connection with the tetragonal → monoclinic transformation in zirconia-containing ceramics, e.g.:

$$\varepsilon^{T} = \int_{T_0}^{T} \alpha(\mathrm{T}')\,\mathrm{d}T' + \varepsilon^{tr} \quad (10)$$

Viscoelastic behaviour can be represented by a similar equation:

$$\sigma(t) = \int_{0}^{t} C(t, t') \frac{\mathrm{d}}{\mathrm{d}t'} (\varepsilon(t') - \varepsilon^{T}(t'))\,\mathrm{d}t' \quad (11)$$

Here $C(t, t')$ describes the temporal relaxation behaviour of the material, which is important for ceramics at elevated temperatures.

From eqns (8)–(11) together with the compatibility condition (i.e. the strain field must be derived from a displacement field) and corresponding boundary conditions, one gets a complete mathematical problem for the calculation of internal stresses.

A general solution can be given by means of the Green's function method. Let G^{0} be the Green's tensor of a homogeneous reference medium C^{0}, then one has[4]

$$\sigma(x) = \int G^{R}(x, x')(\delta C(x')\varepsilon(x') - C(x')\varepsilon^{T}(x'))\,\mathrm{d}x' \quad (12)$$

with

$$\delta C(x) = C(x) - C^{0}$$

Of course this is still a formal solution, since the unknown strain field appears also on the right-hand side of the equation. The resultant eqn (12), however, can be used as a basis for certain approximate solution schemes. In general two different methods are distinguished.

3.1 Deterministic solution

Here one simplifies the heterogeneous microstructure of the material in a way that an analytical solution for the stress field becomes possible. A well-known example is the description of two-phase materials by assuming certain shapes of the second phase particles. The stress field solution for ellipsoidal inclusion is given in Ref. 5 and transferred to elastic stress transfer in composites by Hsueh.[6] The numerical calculation scheme by the finite element method provides a solution for the deterministic approach, too.

3.2 Statistical solution

Contrary to Section 3.1 one does not try to calculate the local peculiarities of the stress field. Instead the fluctuating internal field is described by means of probability distributions, correlation functions and statistical expectation values. Due to the non-regular microstructure of most materials, this solution often yields a better description of the problem. The basics of this approach are reviewed in Ref. 4.

4 Examples of Stress Calculations

In the following the calculated residual stresses of Si_3N_4 composites containing SiC-platelets are presented using the two basic approaches mentioned in the previous section. The calculations were performed using the following elastic constants and thermal expansion coefficients:

	K(GPa)	G(GPa)	E(GPa)	ν	$\alpha(10^{-6}\,K^{-1})$
Si_3N_4	236·4	121·9	312	0·28	3·4
SiC	208·3	169·5	400	0·18	4·4

For an assessment of stresses, a temperature difference of about 1000 K between room temperature and the stress-free state at elevated temperature is assumed.

Firstly results obtained by means of the statistical approach are considered. Figure 1 shows the residual stresses in SiC-platelets and Si_3N_4-matrix as a function of platelet volume fraction. Beside the averages the mean square fluctuation of the stresses is plotted. This statistical fluctuation is caused by the spatial variation of the stress-field due to the non-regular microstructure. The results indicate that the Si_3N_4-matrix is, on average, under moderate compression, which is in accordance with experimental results.[7] Nevertheless, there are also tensile stress components, especially at the interface between particle and matrix. Since the statistical theory gives a more global view on the stress field, this local tension is reflected by the statistical fluctuation. The peculiarities of the local stress field

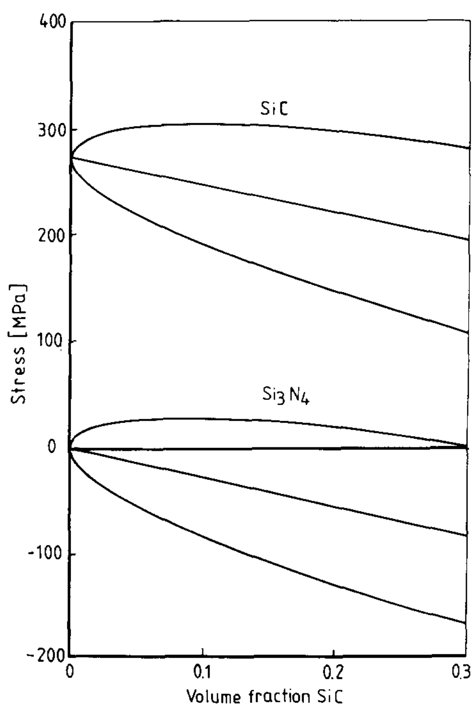

Fig. 1. Residual stresses in SiC-platelets and Si_3N_4-matrix as a function of particle volume fraction. The normal stress σ_{11} is shown. Note that one obtains the same results for σ_{22} and σ_{33}, although these stresses need not be equal because of statistical fluctuations. In order to visualize the statistical variation, the mean square fluctuation has been plotted below and above the average stress.

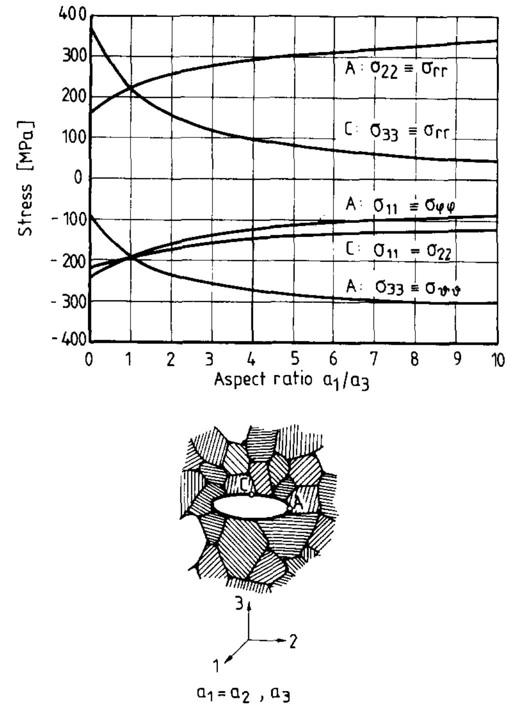

Fig. 2. Influence of particle shape on the residual stresses in SiC-platelet and Si_3N_4-matrix at the interface (thermal stress in matrix $V_p = 0{\cdot}2$). The aspect ratio of the platelets is described by the ratio between the axes $a_1 = a_2$, a_3 (rotational symmetry around the x_3 axis). The stresses were calculated for positions $A = (0, a_2, 0)$ and $C = (0, 0, a_3)$. Note that the normal stress components σ_{rr} must be continuous at the interface, i.e. they equal the stress in the platelet. The isotropic part of the stress tensor is equal to the averages shown in Fig. 1.

will be discussed later. The SiC-platelets, however, are in tension of 100–300 MPa, which is mainly triaxial, although there is also some fluctuation.

Besides the statistical features of the residual stress it is also important to know some local stress values at the particle matrix interface. Here it is recommended that a deterministic theory is applied. By assuming spheroidal particles and an effective interaction field one can calculate the stress in the platelet (which is homogeneous) as well as the stress in the matrix at all positions at the interface. Figure 2 shows some results.

In addition to the calculation of stresses by means of a fixed setting temperature (i.e. one assumes that below this setting temperature no relaxation takes place), it is important to study the influence of viscoelastic stress relaxation at high temperatures. A composite consisting of Si_3N_4 grains which are bonded with a glassy phase is now considered. A typical temperature dependent viscosity is assumed for the glassy phase:

$$\eta(T) = \eta_0 T/T_0 \exp\left(Q/R(1/T - 1/T_0)\right) \qquad (13)$$

with $\eta_0 = 12{\cdot}7\ 10^{12}$ Pas, $T_0 = 1225$ K, $Q = 500$ kJ mol^{-1}. The elastic constants of the glass were chosen

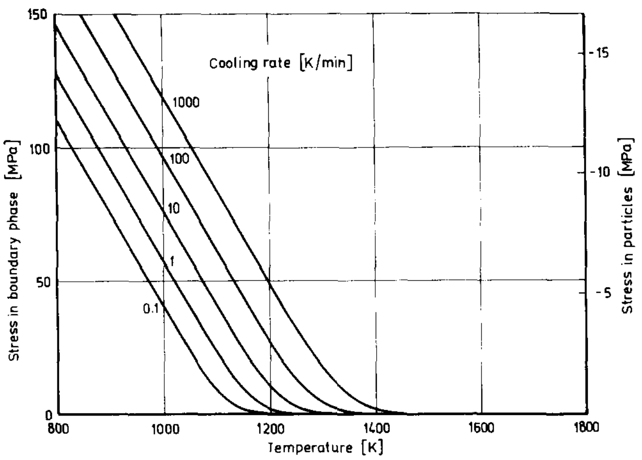

Fig. 3. Viscoelastic stress relaxation in Si_3N_4 caused by a glassy interphase. Average tensile stresses in the glassy phase (left-hand scale) and compression stresses in the Si_3N_4-particles (right-hand scale adjusted to match that on the left-hand scale). The results were obtained for 10 vol.% interphase and plotted for different cooling rates.

as $K = 93{\cdot}9$ GPa, $G = 70{\cdot}4$ GPa, and a thermal expansion coefficient $\alpha = 6{\cdot}1 \times 10^{-6}\,K^{-1}$ was used (the data for Si_3N_4 are given at the beginning of this section).

By using the elastic–viscoelastic analogy it is possible to calculate the time- or temperature-dependent stresses similar to elastic stress calculations (for details see Ref. 8). The calculation indicates again moderate compression stresses for Si_3N_4 but relatively strong tension in the glassy interphase (Fig. 3). Note that the average tension as shown in Fig. 3 is realized by biaxial tension parallel to the particle–glass boundary and compression perpendicular to the boundary. The calculated residual stresses at room temperature for the glassy phase are 270–450 MPa and for Si_3N_4 30–50 MPa, respectively. Obviously there may be a risk of spontaneous microcrack formation in the glass phase. On the other hand, the thin glassy interphase may be quite strong. So this problem deserves further investigation.

In Fig. 3 the influene of SiC-platelets is neglected. Additional calculations indicate that in Platelet reinforced Si_3N_4 the level of residual stress in the glassy phase is in the same range as in the monolithic material. However, this is only true for the composites investigated and one will get different results for composites with other values of elastic moduli and thermal expansion coefficients.

5 Correlation to Mechanical Properties

The mechanical properties at room temperature of SiC-platelet reinforced Si_3N_4 are given in Fig. 4.

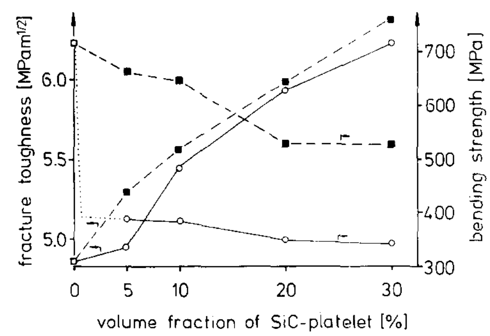

Fig. 4. Fracture toughness and strength of SiC–Si_3N_4 composites are plotted for ○, large (I, American Matrix, USA, average diameter 70 μm) and ■, small platelets (III, C-axis, Canada, average diameter 12 μm). Note that the aspect ratios of both platelet grades are similar (~10).

Fully dense (>99% T.D.) composites with random distribution of either large (I, average diameter 70 μm) or small platelets (III, average diameter 12 μm) were fabricated by reaction bonding and subsequent hot isostatic pressing of SiC–Si powders using 7 wt% Y_2O_3–Al_2O_3 as sintering additives. Fracture toughness was determined by indentation strength in bending and modulus of rupture in four-point bending.

The incorporation of platelets causes a reduction of the bending strength. This result was foreshadowed in the previous section where the conclusion was reached that the glassy phase is under tension. It can be assumed that the crack resistance of the glassy phase at the interface platelet–matrix is low compared to both platelet and matrix. If the Griffith approach is used, the preloading weak interface can be considered as the strength-controlling defect. Therefore, the strength of the composite is partly determined by the diameter of platelets added, as shown in Fig. 4. For example, the incorporation of large platelets results in a strength degradation of ~50% even at low platelet content. It is obvious that a reduction of the residual stress in the glassy phase by changing the composition would improve the strength.

On the other hand, fracture toughness increases close to linearly with increasing volume fraction of platelets, in a first approximation independent of platelet size. Under the influence of external loading crack propagation occurs predominantly at the weak interface. Therefore, the crack is deflected at platelets with a certain misorientation to the axis of external loading as confirmed by fracture surface investigations (Fig. 5(a)). By further propagation the crack is deflected several times and forms a rough fracture surface (Fig. 5(b)). However, the tangential compressive and radial tensile stresses shown in Fig. 2 disturb the crack deflection and, consequently,

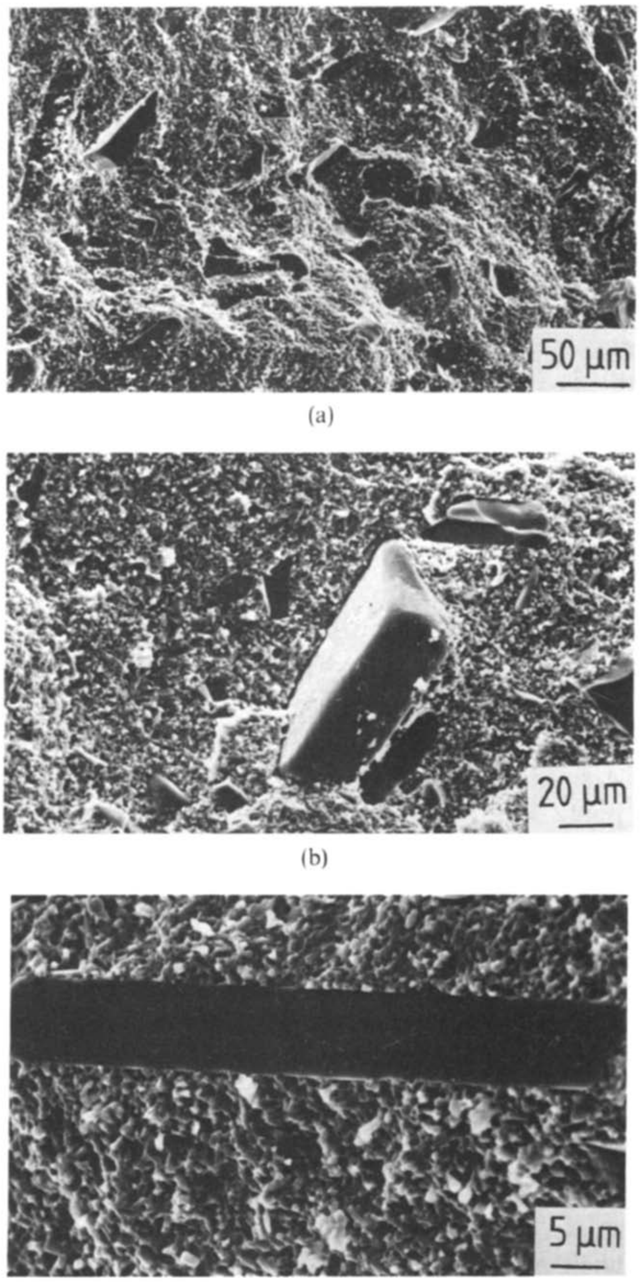

Fig. 5. Fracture surface of SiC-platelet reinforced Si_3N_4. (a) Crack deflection at a platelet with a certain misorientation. (b) Rough fracture surface formed by alternating crack deflection. (c) Rupture of platelet.

favour the rupture of some platelets during crack propagation (Fig. 5(c)). Additionally, the spatial fluctuation of the residual stresses shown in Fig. 1 results in a complex crack propagation through the heterogeneous microstructure. In-situ crack observations showed that the main crack is partly accelerated and partly decelerated or even arrested as foreshadowed in Section 2.

These results are reflected in accompanying *R*-curve measurements, calculated using the compliance function of notched DCB specimens (Fig. 6). The crack tip resistance is found to be decreased

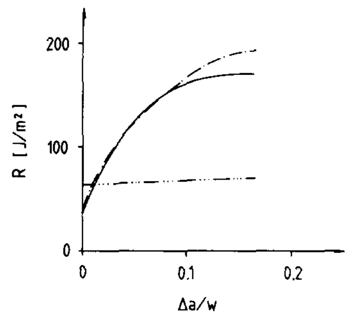

Fig. 6. Crack resistance as a function of normalized crack length for –···–, monolithic Si_3N_4 and composites with –·–·–, 20 vol.% small (III) and ——, large (I) SiC-platelets.[8] Note that *R*-curves of the sample with large starter cracks ($a/w \sim 0{\cdot}7$) are presented.

(presumably as a consequence of a weak platelet–matrix interface), with a strongly rising *R*-curve following thereafter. Only a small difference between large (I) and small (III) platelet inclusions is observed. The low resistance of the composites near the notch mouth supports the suggestion that the platelet–matrix interface acts as flaw origin during cracking. The increase of crack resistance by further crack propagation originates from crack closure forces behind the crack tip, mainly by mechanically interlocking bridges in the crack wake. Additionally, the fluctuating residual stress field may cause microcracks in a process zone around the crack tip. In general, the residual stresses favour the formation of crack closure stresses in SiC-platelet reinforced Si_3N_4 containing glassy phase and partly cause the observed *R*-curve behaviour.

6 Conclusions

For Si_3N_4-ceramics reinforced with SiC-platelets the residual stresses have been calculated using a deterministic as well as a statistical approach. The spatial fluctuation of these stresses results in locally variable crack velocities during fracture. The Si_3N_4-matrix is, on average, under moderate compression. However, tension occurs in the platelets and at the interface between platelet and matrix, which causes crack deflection at the interface. This can also be important for the formation of microcracks in a process zone in the neighbourhood of a macroscopic crack. In general, the residual stresses affect all stages of the crack propagation process and the effect of toughening mechanism. The statistical fluctuation of these stresses can cause locally different processes during crack propagation, and one has to expect complex reinforcement effects which cannot be understood by qualitative considerations based on averages. At the present time, the available methods for the theoretical calculation of residual stresses are useful to improve the understanding of the micromechanic mechanisms in particle-reinforced ceramics. However, the quantitative prediction of mechanical properties by a theoretical approach requires extended modelling of heterogeneous structures, e.g. by Monte-Carlo simulations in combination with residual stress calculations and theories describing crack propagation.

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