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Fracture in precursor-derived Si–C–N ceramics – Analysis of crack roughness and damage mechanisms

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ABSTRACT

Roughness analysis of fracture in precursor-derived amorphous and phase segregated Si–C–N ceramics using fractal methods is reported, towards examining the possible correlations between fractal scaling of roughness and fracture properties as well as fracture damage mechanisms. Topography of the fracture surfaces created at a crack velocity of $\sim 10^{-4}$ m/s was recorded using atomic force microscopy, and analyzed using RMS roughness and second order height–height correlation functions. The evolution of roughness was well correlated with the evolution of structural and compositional inhomogeneities in the amorphous materials, and the formation of second phases in the phase segregated materials. All the investigated fracture surfaces displayed self-affine scaling with a correlation length of ~50–100 nm and a roughness exponent of 0.8 ± 0.1, commensurate with the universal exponent conjectured by Bouchaud et al. corresponding to dynamic damage regime. No correlation was observed between the roughness exponents and the fracture toughness of the corresponding materials. Examination of the crack opening near the tip region revealed no persistent damage cavities assignable to 'plastic deformation' preceding fracture, suggesting that the fracture in the Si–C–N ceramics proceeds in a brittle manner at the employed crack velocities.

1. Introduction

In a previous paper, the evaluation of crack tip toughness (K_{I0}) of Si–C–N ceramics synthesized from a polysilazane precursor was presented [1]. The net change in crack resistance in the investigated range of materials was discussed in terms of the phase evolution at various degrees of ceramization. Depending on the pyrolysis temperature during synthesis, the derived Si–C–N materials possess an amorphous structure with an appreciable quantity of bonded hydrogen below 1000 °C and convert into nano-crystalline ceramics with the loss of hydrogen at higher annealing temperatures [2]. The observed variations in K_{I0} were identified with the progressive increase in the network connectivity, promoted by the stripping of onefold coordinated (OFC) hydrogen in amorphous materials, and the formation of turbostratic graphite (TG) and nano-crystalline SiC (n-SiC) in the phase segregated materials [1]. Concomitant to the overall increase in K_{I0} a monotonous increase in the root mean square roughness (RMS roughness, R_q) of the fracture surfaces was also recorded, proceeding with the ceramization and phase segregation.

Traditionally, an increase in fracture surface roughness is associated with increased fracture toughness. However, a detailed consideration [3–5] of the crack tip processes, microstructure and the energetics of the constituent phases and interfaces reveals that such a generalization would be misleading. Nevertheless, fractographic analysis of fracture surfaces can be usefully applied (i) to identify fracture mechanisms and energy consuming processes e.g., in metals, ceramics and composites [6] and (ii) with necessary additional considerations, towards a quantitative evaluation of the toughening mechanisms [7,8]. Attempts to the former with respect to amorphous ceramics e.g., silicate glasses however have proved to be non-trivial and are a subject of intense discussion recently [9–13].

Analysis and quantification of the fracture surface topography with first order roughness parameters such as R_q is inadequate towards understanding the physics of the fracture process, as it

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does not reflect the spatial correlation of the morphological features contained in the fracture surface. This inadequacy is fulfilled with second order statistical functions such as height-height correlation function (structure function), autocorrelation function and power spectral density (PSD) function [14]. PSD function completely describes the fracture surface, as the height distributions in the fracture surface show Gaussian behavior. A PSD plot in double logarithmic coordinates groups the individual strengths of the fracture surface undulations in inverse length space, enabling analytical comparison and characterization of different fracture surfaces. Extended linear regions in the PSD plot have been shown to signify fractal character of fracture surfaces, associated with a fractal dimension D (or alternatively a roughness exponent ζ) in the respective length scales. Roughness analysis of the fracture surfaces using fractal approach was initiated by Mandelbrot et al. [15] and has been applied [16–20] to understand the fundamental physical process of fracture at the crack front or process zone towards establishing correlations to the gross fracture behavior. (Definitions and methods of fractal analysis are presented in the next section.) Mecholsky et al. have addressed the problem through the inclusion of D (or precisely, the fractal dimensional increment D^*) as a sub-parameter within the critical surface energy term, based on experiments and modeling. The experimental data supporting their formulation suggest a positive correlation of D^* with K_{IC}, with an implicit notion that K_{IC} scales with crack tortousity [16,17]. Borodich proposed a different formulation for accounting the fractal character of fracture surfaces, where the surface energy term in the Griffith energy balance equation is defined in terms of the 'specific energy absorbing capacity of the fractal surface', which corresponds to the amount of elastic energy spent on forming a unit of fractal surface area [18]. On the other hand, Bouchaud et al. conjectured that the fractal scaling of fracture surfaces from a wide range of materials e.g., glasses, metals and wood displays a universal roughness exponent ($\zeta \sim 0.8$), and that the correlation to the fracture properties is reflected in the observed correlation lengths ξ of the fractal regimes in the respective materials [19]. Additionally, depending on the crack velocity, two regimes of damage processes (quasi-static and dynamic) were discussed, exhibiting two universal roughness exponents *viz*., 0.5 and 0.8 in short and large length scales, respectively. The crossover lengths ξ_c between the two regimes were discussed to be both crack velocity and material dependant, the latter dependency arising from the size of the heterogeneities in the material structure [20,21]. Concerning the investigation of the damage mechanisms at the crack-tip/process zone, formation and linkage of damage cavities akin to plastic deformation was proposed, even for nominally brittle materials such as silicate glasses and amorphous ceramics. Further, the roughness exponent within the damage cavities was related to a quasi-static damage process, while that arising from the linkage of cavities was related to a dynamic damage process [20]. This interpretation derived strong motivation from the results of MD simulations of fracture in silica glass and amorphous Si_3N_4 [22,23]. Subsequently, *in situ* high-resolution AFM imaging of propagating cracks in silicate glasses were presented to support the formation of 'damage cavities' [9–11].

In light of the above background, it is worthwhile to examine the fractal scaling of roughness in the fracture surfaces of the precursor-derived Si–C–N amorphous and nano-crystalline ceramics possessing a progressively varying material structure in terms of the number and size of the heterogeneities. The objectives of this work are twofold: (a) examination of the possible correlations of fractal scaling parameters and fracture behavior and (b) examination of the damage mechanisms at the advancing crack front that lead to the fracture surface roughness. Towards the first objective, the roughness analysis of the fracture surfaces of Si–C–N ceramics is presented, performed using second order statistical functions to evaluate the fractal scaling, relevant length scales of fractal regimes and attendant roughness exponents. These results are then discussed with reference to previously developed understanding of the subject from literature as above to examine possible correlations between fractal scaling and fracture behavior. The second objective is pursued using high-resolution AFM imaging of the crack-tip process zones and crack faces of the critically loaded cracks, where the possibility of the formation of damage cavities is investigated.

2. Theoretical background

Fracture surfaces are self-affine objects displaying scale invariance under an affine transformation [18,19]:

$$x' = \lambda x, \quad y' = \lambda y \quad \text{and } z' = \lambda^{\zeta} z. \tag{1}$$

They are characterized by their non-integer fractal dimensions D, in contrast to Euclidian objects. The fractal dimension of a fractal surface lies in the range $2 < D < 3$ and that of the fractal curve, in the range $1 < D < 2$. To illustrate, if N is the number of balls or cubes with diameter Δ required to cover a fractal object and if the increase in N due to a corresponding decrease in Δ is proportional to Δ^{-D} (i.e., $N \propto \Delta^{-D}$), then the object is said to have a fractal dimension of D (more specifically, the box dimension D_B) [18]. To enable comparison of the fractal dimensions derived from surfaces and curves, the fractal dimensional increment D^* is defined such that $D = 2 + D^*$ for a fractal surface and $D = 1 + D^*$ for a fractal curve [16–18]. An increase in fractal dimension is physically manifested as an increased tortousity of the fracture surface profile trace. Considering the self-affine rather than self-similar scaling that is observed in fracture surfaces (Eq. (1)), the more appropriate parameter that describes the fractal scaling in fracture surfaces is the roughness exponent ζ (also known as self-affine exponent or Hurst exponent), which varies between 0 and 1 [19]. The roughness exponent ζ and (box) fractal dimension D_B of fracture profile are related to each other as $\zeta = 2 - D_B$ in the limit where the horizontal increment of the fracture profile data is small compared to the typical range of profile heights, i.e., $\delta x << |z_{\mathrm{Max}} - z_{\mathrm{Min}}|$. Thus, a higher fracture profile tortousity corresponds to a higher D but a lower ζ. Unlike the ideal fractal objects, fractal character of real fracture surfaces is limited to regimes of finite length scales, characterized by their corresponding correlation lengths ξ_i. The fractal dimension D_i or the roughness exponent ζ_i is defined as valid within ξ_i, beyond which either a different fractal regime or a Euclidian dimension is realized.

Several methods are described in literature for the evaluation of fractal scaling of fracture surfaces, and include slit island analysis (SIA) [16], variable bandwidth method [24], return probability method [19,24] and power spectrum method [19,21,24]. The analytical procedures and reliability issues of these methods have been extensively reviewed [24,25]. Generally, the analysis is performed on the fracture profile curves obtained by sectioning of the fracture surface, either along the plane of the surface (as for SIA, yielding island curves) or normal to it (e.g., in power spectrum method). In this work, fractal scaling is evaluated in terms of the roughness exponent ζ from fracture profile curves obtained from vertical sectioning of the fracture surface, using the following relations:

(1) from the variational Rq method [24,26]:

$$R_q(r) \propto r^{\zeta}, \quad \zeta = \left(d \log R_q(r) / d \log r\right); \tag{2}$$

(2) from the structure function S(r) [19,20]:

$$S(r) = \langle |h(x+r) - h(x)|^2 \rangle \propto r^{2\zeta},$$
$$\zeta = (1/2) * (d \log S(r) / d \log r); \tag{3}$$