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Mechanical behaviour of a silicon nitride particulate ceramic composite

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Abstract

Mechanical behaviour of a particulate ceramic composite (Si_3N_4 + SiC) was investigated. Its strength and fracture toughness on heating up to 1300 °C were determined as well as stress–strain curves plotted for this temperature range were analyzed. It is emphasized that this material is not only heterogeneous but also inelastic, and its deformation and fracture behaviour differ considerably from those of conventional ceramics. It was established that SENB fracture toughness measurements on notched specimens in flexure were quite reliable. Thus, there is no need in employing sophisticated standard test methods for this purpose. Fracture resistance estimates by the edge fracture (EF) method demonstrated that this material exhibited a lower barrier to the onset of fracture and a nonlinearly rising R-line, i.e., it displayed the ability to resist crack propagation (R-curve effect). The fracture resistance F_R and initial fracture toughness K_{Ii} were also determined. This information is rather useful for analysis of its actual performance under mechanical loading. The model of a nozzle vane of the gas turbine was employed to illustrate that the EF method was appropriate for evaluating the uniformity of ceramic items by their fracture resistance.

Keywords: B. Composites; C. Strength; C. Fracture; EF method; R-line

1. Introduction

Particulate ceramic composites, being quite promising materials, offer an adequate alternative to conventional ceramics. They are often employed in different fields of technology for manufacturing heavy-duty structural elements. This gave impetus to studies on the mechanical behaviour of those materials, with special emphasis on their ability to resist fracture. But the fact that such a characteristic as the fracture toughness K_{Ic} [e.g., 1,2] is an integral value that estimates the ability of the material to resist the initiation of a dangerous crack and its further propagation escapes the attention of researchers quite often. However, in various applications of examined materials the initial stage of their fracture is also of considerable interest, viz. the extension of existing (or originating) microcracks, which appears as nonlinearity of the stress–strain curve for a solid specimen in flexure and an increase in acoustic emission accompanying deformation, and the origination of a dangerous macrocrack from microcracks with further loading.

The investigation is devoted to studying the mechanical behaviour of a silicon nitride–silicon carbide particulate ceramic composite. Its fracture resistance was evaluated by the alternative edge fracture (EF) method [3]. This additional information on the examined material can probably be not only of scientific interest but also of practical use.

2. Material and methods

A porous reaction-bonded silicon nitride-based (5 μm grains) particulate ceramic composite (pore volume up to 30%) doped with silicon carbide (up to 30–40% of 30–60 μm grains) and magnesia (up to 2%) [4] was the object of investigation. It was termed NKKKM[1] [5]; variations of this material are also known as SNCM [6] and NKM-2 [7]. Specimen blanks (and items) were produced by thermoplastic injection moulding in split metal moulds with a paraffin + 10–12% beeswax slip containing silicon powder. Then green ceramic blanks were treated on induction heating (about 1600 °C) in nitrogen under boron nitride + silicon covering,

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[1] The abbreviation for silicon nitride, silicon carbide, and magnesium in Russian.

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which ensured their reaction bonding and further activated sintering. To optimize properties of these ceramics, they were doped with about 2% boron nitride (NKKKM-83B), which provided their higher homogeneity. However, it affected their corrosion resistance, e.g., in a flow of organic combustion products [8].

The surface of as-produced ceramics was smooth and mat, and ceramic items, being rather dimensionally accurate, were usually used without any machining. Such ceramics were first employed as a material of crucibles for gold melting. It was established that they were quite resistant on multiple heating in air. Then they have found use in engines [6]. They turned out to be also quite appropriate for general refractory applications, e.g., accessories of chambers for high-temperature tests of ceramics without a protective environment (Fig. 1). The studies resulted in several modifications of these ceramics [5] whose strength and fracture toughness did not essentially differ. The most representative one (NKKKM-83B) was chosen for the present investigation, others were examined elsewhere, e.g., [6,9]. Comparative tests also made use of microlaminate ternary carbide Ti_3SiC_2 [10] and other materials studied earlier, i.e., their mechanical behaviour during the tests can be quite predictable.

Basic investigations were performed on ground rectangular bars with 4 mm × 5 mm and 3 mm × 4 mm cross-sections and about 45 mm long, which were cut out from 6 mm × 6 mm blanks.

Mechanical characteristics in ambient environment (e.g., as in [11]) were determined in a multipurpose loading CeramTest device fitted with an independent measurement system and mounted on a universal test machine. This device provided high-precision loading rod displacement. A rigid dynamometer, located under a lower loading support, registered the applied load. Moreover, it recorded acoustic emission accompanying deformation and fracture of specimens. The tests were performed in four-point flexure (20/40-mm span), the support design ensured free displacement of the rollers. The deflectometer with a sensitive LVDT gauge was suspended on the specimen for recording load–displacement curves.

In high-temperature tests, the loading support made of hot-pressed silicon carbide and secured on NKKKM columns (Fig. 1a) was placed into a heating chamber, the LVDT gauge was located outside, with its NKKKM probe being inserted into the heating zone (Fig. 1b) [12]. The cross-head speed of a test machine was 0.5 mm/min.

Strengths, static elastic moduli, ultimate strains, and brittleness measures χ were determined from load–displacement curves. The brittleness measure χ, characterizing the inelasticity of ceramics, was calculated as the ratio of the specific elastic energy, accumulated in a solid specimen in flexure to the moment of its fracture, to the total specific energy spent for its deformation by the same moment [13] (for an elastic material $\chi = 1$).

In our early studies, attempts were made to determine fracture toughness in flexure of ceramic specimens with a sharp crack (SEPB method) and in tension of a double cantilever beam. But because of difficulties associated with accurate measurement of initial crack lengths, which are necessary for calculations of critical stress intensity factors K_{Ic}, the SENB method was adopted instead. In this procedure, specimens with a notch 200 μm wide made with a diamond saw were tested in flexure. However, this method could result in the overestimation of the ability of the material to resist fracture since additional energy is spent for the formation of a sharp crack from the blunt notch, being the stress concentrator in such tests [9].

The attempt was also made to use the SEVNB method [9] that is best suited to fracture toughness estimation of ceramics [14]. For this test, the V-notch was prepared by introducing a 1–3-μm fine-grained diamond paste into a notch 200 μm wide and polishing it out with a razor blade secured in a special machine that provides its reciprocating motion. But because of

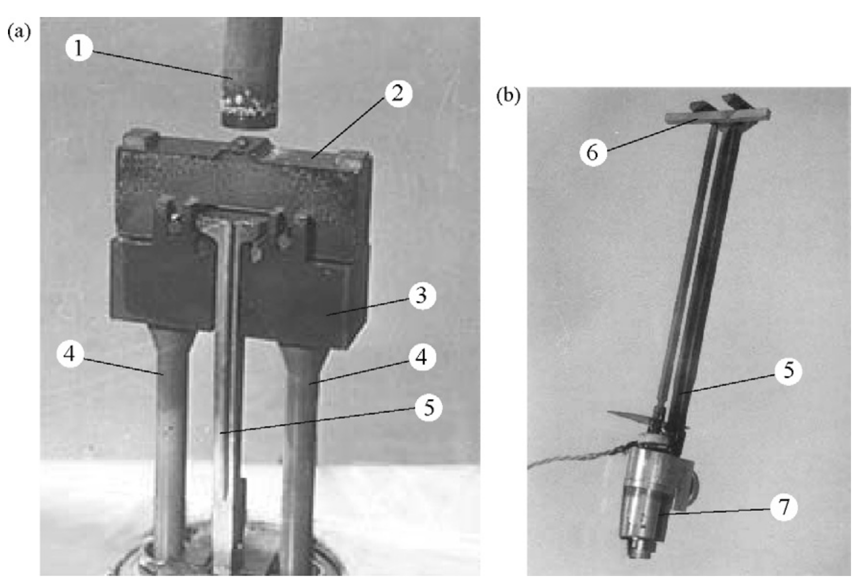

Fig. 1. Loading device for high-temperature tests with elements made of NKKKM ceramics (a) and deflectometer suspended on the specimen (b): (1) loading rod, (2 and 3) upper and lower prismatic supports, (4) supporting rods, (5) deflectometer, (6) specimen, (7) LVDT.

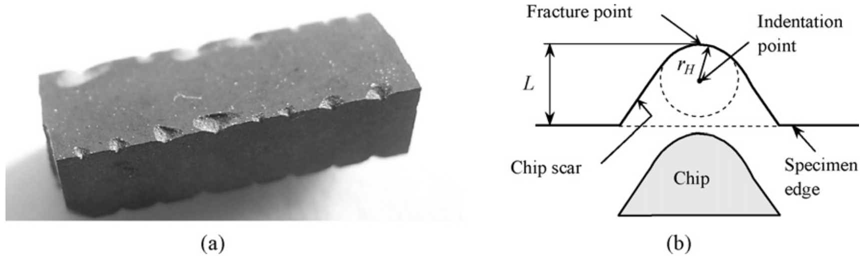

Fig. 2. NKKKM-80 specimen cut out from a nozzle vane (a) with chip scars on its edges (side view) and schematic of the fracture zone after the Rockwell indentation (b): r_H is the radius of a Hertzian ring crack.

considerable differences in hardness of the silicon nitride matrix and coarse reinforcing silicon carbide grains, the formation of the notch of a preset sharpness turned out to be problematic. Both SENB and SEVNB tests were carried out in three-point flexure (20-mm span) with a CeramTest device.

Specimen fragments of NKKKM-83B ceramics, formed in the above fracture toughness tests, were polished and rectangular edges with a radius that did not exceed 20 μm were prepared. They served as specimens for further EF fracture resistance tests [3]. In those tests, the specimens cut out near the leading and trailing edges and body of an NKKKM-80 nozzle vane were also used. Specimen edges were flaked off with a Rockwell C-Scale standard diamond indenter several times (Fig. 2a). The indentation point was chosen with a magnifying glass. The loading support of a CeramTest device was replaced by the indenter holder and *X*–*Y* table (earlier hardness and fracture behaviour of materials were studied in the same way [11,15]). A Hertzian ring crack formed on the Rockwell indentation of the specimen gives rise to its edge fracture. Chip scars acquired a scallop-like shape at the perpendicular displacement of a Rockwell indenter relative to the specimen surface (they took on the shape of Hertzian quasi-cones on glass specimens [16,17]).

The load P_f responsible for the fracture of the specimen edge was registered with a computer. Then the distance from the specimen edge to the extreme point on the chip scar termed the fracture distance L (Fig. 2b) was measured. These data were employed to plot fracture diagrams (fracture distance L–fracture load P_f relations). The ratio of the average P_f load to the average fracture distance L was termed the fracture resistance F_R [18]. The slope of the straight line, approximating the fracture diagram, termed the flaking toughness E_t [3] was an auxiliary characteristic for the present investigation. Fracture resistance F_R–fracture distance L relations termed R-lines [19] were also plotted, they correspond to the initial portion of a conventional R-curve [20].

Unfortunately, the method of local fracture resistance evaluation with a Vickers indenter (IF method [21]) could not be employed for the comparison of results. The first difficulty was associated with polishing the specimen surface to necessary smoothness, it was also impossible to initiate necessary straight cracks near the corners of Vickers indents since they branch running into coarse silicon carbide grains. Moreover, it is quite difficult to establish the location of crack tips.

3. Results and discussion

Strengths, ultimate strains, static elastic moduli, and other characteristics of examined ceramics are summarized in Table 1. Stress–strain curves and fracture toughness values for NKKKM-83B ceramics at different temperatures are presented in Fig. 3, their fracture diagram and R-line are shown in Fig. 4.

Analysis of investigation results was started with examination of the stress–strain curve for inelastic NKKKM ceramics (Fig. 5a). It is seen that these ceramics as a structural material possess the elastic limit σ_e that looks low (their inelasticity is explained by structural microfractures, which is confirmed by AE counts, Fig. 5c). Therefore, the unique design of an all-ceramic nozzle assembly of the gas turbine had to be developed to give the most effective use of these ceramics for the above purpose [22]. In this design, nozzle vanes, freely expanding on heating, were fixed in a ceramic solid outer bearing ring. The roots of those vanes formed the inner ring of the nozzle assembly. Allowable calculated stresses σ_{max} in all the components did not exceed 60 MPa, i.e., they were below the elastic limit σ_e of the examined ceramics (Fig. 5a).

The examination of the diagram in Fig. 3a shows that the strength of NKKKM-83B ceramics at high temperatures also exceeds 60 MPa, which ensured full-scale tests of nozzle assemblies. The corrosion protection of the items before operation is effected by holding them in air at 800–1100 °C, as

Table 1
Characteristics of materials

Material	Density (g/cm^3)	Brittleness measure (χ)	Strength (MPa)		Weibull modulus	Ultimate strain × 10^4 (m/m)	Elastic modulus (GPa)
			20 °C	1300 °C			
NKKKM-80	2.52	1.00	216	133	8–10	11.6	145
NKKKM-83B	2.44	0.88	170	175	18–20	12.0	147

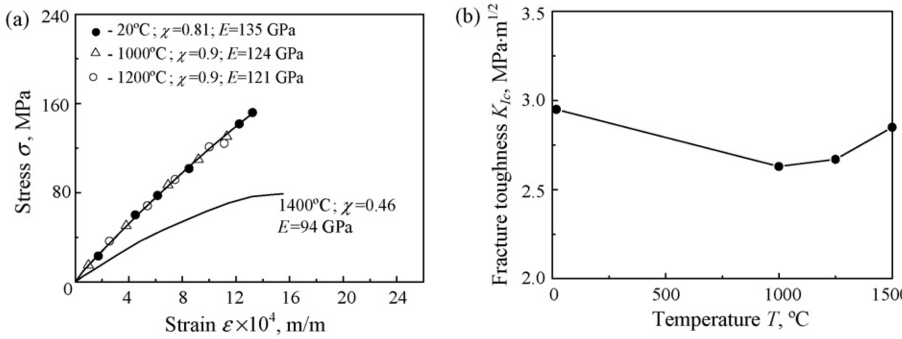

Fig. 3. Temperature effect on the pattern of stress-strain curves (a) and fracture toughness (b) of NKKKM-83B ceramics.

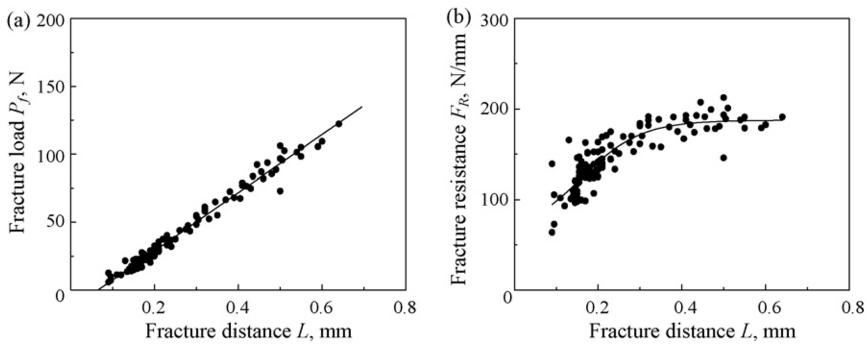

Fig. 4. EF test results for NKKKM-83B ceramics in ambient environment: fracture diagram (a) and R-line (b).

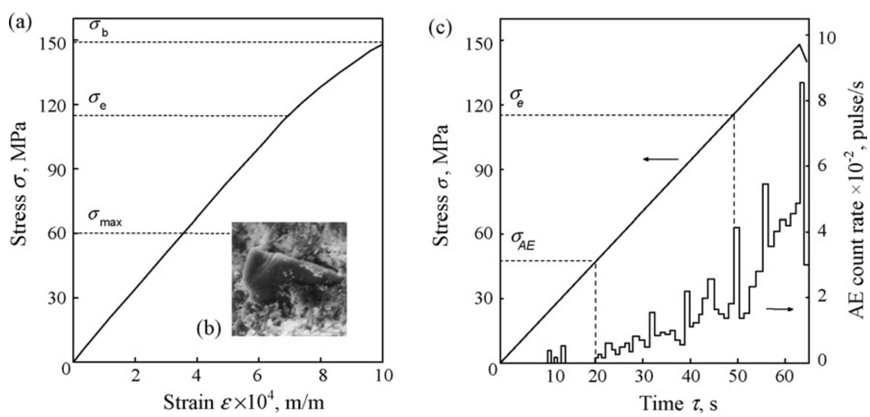

Fig. 5. Test results for NKKKM specimens in flexure: stress–strain curve (a), micrograph of a fracture surface (b), and AE count rate (c): σ_{AE} is the stress at the onset of acoustic emission pulse trains.

a result their open surface pores were filled with amorphous silica and α-cristobalite [8].

The preparation of specimens for SEVNB tests includes making the V-notch. When it is made by the conventional method with a ''floating'' razor blade that distributes diamond paste, the notch width measured as a diameter of the circle inscribed in its tip [9] varied over the thickness of the specimen. The tests demonstrated that examined ceramics displayed very limited sensitivity to stress concentrations (low resistance to the onset of fracture), therefore, the V-notch sharpness exerts inconsiderable influence on fracture toughness values. For example, 20–70-μm notch widths for the specimens cut out from a nozzle vane resulted in an average K_{Ic} value of 2.87 ± 0.11 MPa m$^{1/2}$, when their widths were 320–500 μm, it varied slightly, being 2.98 ± 0.17 MPa m$^{1/2}$ (constant notch

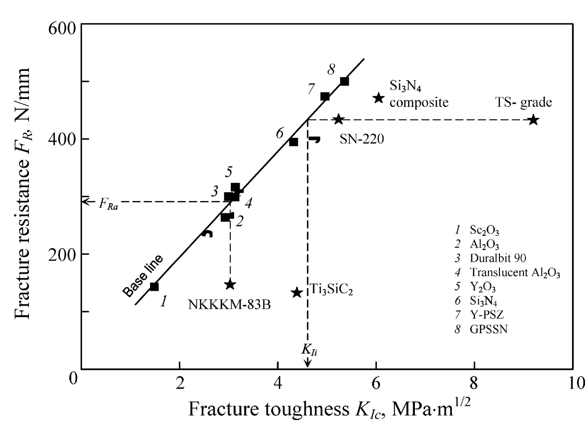

Fig. 6. Base diagram with the base line, initial fracture toughness (K_{Ii}) and arbitrary fracture resistance (F_{Ra}) estimates for different ceramics.

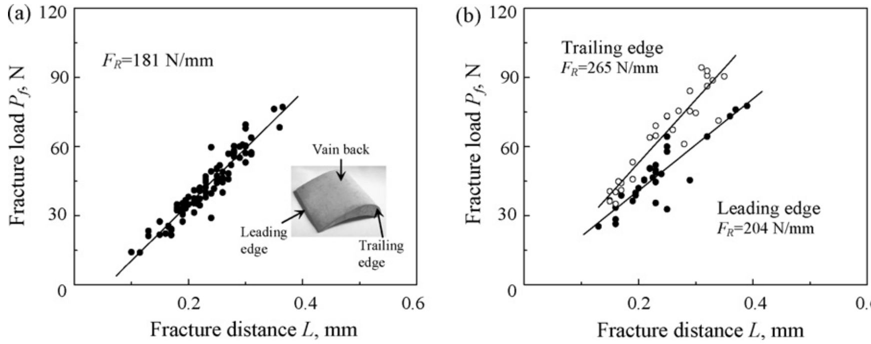

Fig. 7. Fracture diagrams for nozzle vane ceramics: specimen cut out from the vane body (a) and specimens cut out near its leading and trailing edges (b).

widths were not reproduced during their preparation). About the same insensitivity to the V-notch width is exhibited by TS-grade inelastic zirconia ceramics partially stabilized with magnesia, laminated composites $Si_3N_4/Si_3N_4 + TiN$, and microlaminate ternary carbide Ti_3SiC_2 [23], while elastic homogeneous ceramics displayed fracture toughness that could differ more than twice [9,24]. The above data count in favour of the ability of examined ceramics to resist crack initiation.

Fracture toughness estimates of examined ceramics by the SENB method (Fig. 4) did not bear witness to any specific behaviour of this material but an increase in test temperature brings about a slight decrease of its fracture toughness, which is also advantageous.

Analysis of the fracture diagram for NKKKM-83B ceramics plotted by EF test data (Fig. 4a) demonstrates that it is satisfactorily approximated by the straight line. But their fracture resistance F_R and flaking toughness E_t differ considerably (147 ± 31 and 213 ± 4 N/mm, respectively), which is caused by considerable heterogeneity of the material (for homogeneous single-phase ceramics these values are closer [17,19]). As is seen in Fig. 4b, NKKKM-83B ceramics possess the nonlinearly rising R-line, thus, they have the ability to arrest propagating cracks [18,19]. In other words, they exhibit the R-curve effect, which is revealed in a much simpler way by the EF method that can be employed in a materials science laboratory on small-size specimens.

Further analysis of test results was built on the base diagram (Fig. 7) with the base line [19], demonstrating proportionality between F_R and K_{Ic} values for elastic homogenous single-phase ceramic materials, which is not peculiar to other local fracture resistance test methods for brittle materials. The results (data points) for NKKKM-83B ceramics plotted in this diagram fell below the base line. Test results for such heterogeneous materials as microlaminate ternary carbide Ti_3SiC_2 (brittleness measure $\chi < 0.5$), silicon nitride particulate ceramic composites, such as hot-pressed SN-220, reinforced with titanium carbide, and a hot-isostatically pressed Si_3N_4 composite with silicon carbide, and TS-grade ceramics ($\chi = 0.42$), are also plotted in this figure. It is associated with the fact that these materials display a lower resistance to the onset of fracture (termed the fracture barrier in [25]) than conventional linear elastic ceramics, which exhibit uncontrolled crack propagation, originating in their subsurface layer, in the absence of a toughening mechanism (e.g., microfracture), contributing to relaxation of critical tensile stresses in the fracture zone. This effect can be evaluated with the base line, as proposed in [19], i.e., to use average estimates of fracture resistance of ceramics that do not contradict the solid model of linear fracture mechanics. The ratio of a measured F_R value to its arbitrary value, i.e., to its vertical projection onto the base line (F_{Ra}) can be proposed as a numerical characteristic of the fracture barrier effect (Fig. 6). This ratio can be termed the factor of the barrier to the onset of fracture ψ, which is equal to unity for ceramics consistent with the base line. The same estimates are possible by determining corresponding arbitrary fracture toughness values with the base line, as proposed in [17]. The values ψ for NKKKM-83B, Ti_3SiC_2, SN-220, a Si_3N_4 composite, and TS-grade ceramics are equal to 0.51, 0.32, 0.89, 0.83, and 0.51, respectively. But, e.g., for silicon carbide EKasic® T $\psi = 1.3$ and for technical quartz glass $\psi = 3.9$. Thus, the resistance of brittle materials (including particulate ceramic composites) to the onset of fracture can be greatly different, which should be taken into consideration in practice since their mechanical behaviour can significantly differ from that characteristic of ideal ones, often used as a basis for analytical estimates of ceramics.

For the materials, whose test results (data points) fall below the base line, approximate initial fracture toughness (K_{Ii}) values [17,19] can be determined, projecting those results onto the base line, as shown in Fig. 6 by the example of TS-grade ceramics[2]. The values are approximately consistent with initial fracture toughness data on their R-curves [17,18]. The K_{Ii} values for NKKKM-83B, Ti_3SiC_2, and TS-grade ceramics reach 1.6, 1.4, and 4.7 MPa m$^{1/2}$, while their K_{Ic} values are equal to 3.03, 4.39, and 9.20 MPa m$^{1/2}$, respectively.

The EF method was also used to evaluate the homogeneity of NKKKM-80 ceramics, which the nozzle vane is made of, by the ability of these ceramics to resist fracture in flaking. For this purpose, the specimens were cut out from the vane body as well as near its leading and trailing edges and then tested. It was established (Fig. 7) that their fracture resistance was different, thus, the material displayed different properties in various zones of the vane. It is probably associated with different conditions of reaction bonding and activated sintering in those zones.

[2] TS-grade ceramics tested in this investigation differ from similar ceramics examined in [3] (note).

It should be noted that information on the fracture behaviour of ceramics obtained in edge fracture tests complements data usually used in analysis of performance of ceramic materials and assists in evaluation of the potentials of those materials in many applications.

4. Conclusion

The investigation of silicon nitride-based particulate ceramic composites has demonstrated that such a material can be inelastic, which should be taken into consideration in engineering applications, since its operation, especially under cyclic mechanical and thermal loadings and under stresses above the elastic limit leads to accumulation of dangerous defects and can shorten service life of ceramic components. Therefore, it is desirable to complement the specification for examined materials with the characteristic of their inelasticity, e.g., with the brittleness measure χ.

Fracture resistance estimates for particulate ceramic composites, especially for inelastic ones, exhibiting limited sensitivity to initial fracture do not necessarily agree with their fracture toughness values obtained by standard test methods. In this case, a simple test method, such as SENB, can give good results.

The efficiency of the EF method for providing useful information on the fracture resistance of particulate ceramic composites has been demonstrated. It allows data on the onset of fracture and other relevant information to be accumulated, as well as the R-curve effect to be revealed. Such tests with small-size specimens can be performed on the equipment available in a conventional mechanical laboratory. The EF method may also be employed for studying the localized mechanical behaviour of the material on the edges of the items.

Both SEVNB and EF tests would be appropriate for use on the same specimens for estimating the fracture resistance of composite ceramics, i.e., simultaneous employment of the methods of fracture and contact mechanics.

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