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Influence of Microstructure on Crack Tip Toughness of α-Sialon Ceramics

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Abstract

For two different α-Sialon ceramics, one with a stoichiometric composition and the other one with an extra amount of yttria, the fracture toughness (SEVNB) and the crack opening displacements (COD) in the crack tip (CT) region were measured. The fracture toughness values were 3.0 MPa√m for the material without excess Y_2O_3 and 4.5 MPa√m for the material with a residual grain boundary phase. In contrast, the crack tip toughness of α-Sialon with the stoichiometric composition was 1.35 – 1.68 MPa√m and 0.95 – 1.01 MPa√m for the α-Sialon with an excess amount of yttria. Crack behavior in α-Sialon with an excess amount of yttria is both intergranular and transgranular owing to the formation of large elongated grains, and mainly intergranular in α-Sialon with stoichiometric composition. The different crack behavior and the resulting crack tip toughness are the result of the different chemistry and changed microstructure of the materials.

Keywords: Sialon, microstructure-final, toughness and toughening, mechanical properties, crack tip toughness

I. Introduction

The development of α-and α/β-Sialon ceramics has gained increasing interest over the last few years owing to their higher hardness and higher chemical resistance in comparison with those of β-Si_3N_4 materials and their correspondingly better wear properties in cutting tool applications [1–3]. α-Sialon ($M_xSi_{12-(m+n)}Al_{m+n}O_nN_{16-n}$, where x = m/v and v is the valance of the cation M^{+v}; M = Li, Mg, Ca, Y and some rare earth elements Z > 60) has the same structure as α-Si_3N_4 [1]. It is well established that the densification of β-Si_3N_4 and of α-and β-Sialon materials is a liquid phase sintering process. Above 1250 °C (depending on additives) an oxide liquid is formed with increasing temperature in which Si_3N_4 is dissolved and depending on the composition β-Si_3N_4 or β-or α-sialon precipitates. In the case of α-Sialon in the liquid dissolved rare earth oxides, AlN/Al_2O_3 are precipitated together with the Si_3N_4 to form the α-Sialon phase. By this chemical reaction, the amount of liquid is strongly reduced or completely disappears. If stoichiometric amounts of rare earth oxides are used, a fast consumption of the liquid results in retardation of the densification and the formation of small equiaxed grains. Therefore materials with m and n values near 1 are difficult to densify completely without the application of external pressure [1, 4–5]. The addition of some excess rare earth oxides strongly improves the densification behavior and causes growth of elongated grains. The resulting α-Sialon ceramics have a fracture toughness similar to β-Sialon materials [1–6]. Nevertheless there are still several open questions concerning the toughening mechanisms in Sialon materials. These materials are characterized by an increasing crack growth resistance, K_R, with increasing crack propagation and extension, Δa, (R-curve behavior) [7]. The rising R-curve of Si_3N_4 and other ceramics is mainly due to the grain bridging effects behind the propagating crack [8–13]. In order to determine the R-curve of a ceramic, conventional R-curve measurements are performed based on determination of the load displacement and the crack length displacement. The initial point of the R-curve, the so-called crack tip toughness, K_{I0}, is important for indicating whether an increasing R-curve behavior exists and for predicting the strength of ceramics with natural μm size flaws [12,13]. The estimation of the crack tip toughness (initial point of the R-curve) by means of conventional R-curve measurements is complicated by measurement difficulties including: a) the accurate measurement of micron size initial crack propagation, from K_{I0} to saturation value is very short, about 10 μm [11] and b) the determination of the load deflection point.

Schneider and Fünfschilling *et al.* [11,12,19,20] have proposed a method to determine the K_{I0} by measuring the opening displacement of the cracks introduced by Vickers indents near the crack tips. They have evaluated the method for different Si_3N_4 materials.

In the literature, measurements of the crack tip toughness, K_{I0}, were performed for some ceramics: alumina [14–16], soda-lime glass [17] and silicon nitride [18–19,21]. There is no data available in the literature about the crack tip toughness of Sialons.

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In this work, the crack tip toughness and the fracture toughness of two different α-Sialons, one with a stoichiometric composition and the other one with an extra amount of yttria, were measured with the proposed method of measuring the opening displacement of the cracks near the crack tip. The resulting K_{I0} values were analyzed and interpreted in respect of the different observed microstructures.

II. Materials and Method

(1) Materials

Two α-Sialons with different chemical composition were used in the present study to measure the crack tip toughness. As given in Table 1, one α-Sialon ceramic had a stoichiometric composition with m = 1.2 and n = 1 (sample Y1210-SC) whereas the other α-Sialon ceramic had a similar composition but 1.5 wt% Y_2O_3 in excess (sample Y1210-EY). Densification of the materials was performed by hot pressing at 1800 °C and 30 MPa for 30 min, followed by additional heat treatment at 1825 °C for 90 min in nitrogen atmosphere at a gas pressure of 50 bar. The resulting density, as measured by Archimedes method, was 3.28 g/cm^3 (99.7 % relative density) for the stoichiometric-composed Sialon (sample Y1210-SC) and 3.30 g/cm^3 (99.7 % relative density) for the material with the excess amount of yttria (sample Y1210-EY). Detailed information about the sintering and characterization of the α-Sialon ceramics used for investigations can be found in a previous paper [4].

The fracture toughness of the two investigated materials was measured using the SEVNB method (Single-Edge V-Notch Bending) in a 4-point-bending set-up on the basis of three single measurements/material and calculation in accordance with ISO 23146:2008. The notch radius was less than 20 – 30 μm.

(2) Crack Tip Toughness Measurement

A method described in [19] and comprehensively discussed in [11] was adapted for the measurement and calculation of the crack tip toughness of the investigated α-Sialons. Therefore Vickers indentations were made with 98.8 N load and 15 s dwelling time on the polished surface of the two α-Sialon materials. The resulting indentations and cracks were then investigated with a Field Emission Scanning Electron Microscope FESEM (ULTRA 55, CARL ZEISS, Germany) at magnification up to 120kx.

For each sample two indentation diagonals (2b) and the crack length (c) were measured. For the evaluation of the crack tip toughness, the crack opening displacements (2δ) were measured as a function of the measured distance (x) from the crack tip. Two cracks, resulting from two different indentions, were evaluated for each sample. Normally, owing to the measurement difficulties, 2δ's were measured starting as close to the crack tip as a distance x of about 50 nm up to about 20 μm in about 20 – 25 points. Using the measured crack opening displacements 2δ at distance x from the crack tip, the crack tip fracture toughness (K) was calculated according to the following equations (1 – 3) from [11, 19] which were analytically derived in [20]:

$$\frac{\delta}{\kappa} = \frac{\sqrt{b}}{E'}\left(\sqrt{\frac{8}{\pi}}\left(\frac{x}{b}\right)^{1/2} + A_1\left(\frac{x}{b}\right)^{3/2} + A_2\left(\frac{x}{b}\right)^{5/2}\right) \quad (1)$$

with,

$$A_1 \cong 11.7\exp\left[-2.063\,(a/b-1)^{0.28}\right] - \frac{0.898}{a/b-1} \quad (2)$$

$$A_2 \cong 44.5\exp\left[-3.712\,(a/b-1)^{0.28}\right] - \frac{1}{a/b-1^{3/2}} \quad (3)$$

The effective modulus, E' = E for plane stress and E' = E/(1 - ν^2) for plane strain where E is the Young's modulus and ν is the Poisson's ratio of the material. Since plane stress exists on the surface, E' = E. The Young's modulus was set as 320 GPa in the above calculations. In A_1 and A_2: a = b + c whereas b and c correspond to the half indentation diagonal and the crack length, respectively.

III. Results and Discussion

Microstructure investigations by SEM show, that during sintering, α-Sialons with excess amount of Y_2O_3 (sample Y1210-EY) had anisotropic grain growth resulting in an elongated grain structure, while the α-Sialon with the stoichiometric composition (sample Y1210-SC) had less grain growth owing to less liquid phase, which results in an equiaxed grain structure (Figs. 1 a, b). In the material Y1210-SC no triple grain junctions filled with a Y_2O_3 rich grain boundary phase can be found, whereas the material Y1210-EY contains a secondary phase in the triple junctions. No crystallization of this secondary phase could be determined by XRD.

The fracture toughness (SEVNB method) of the two materials was about 3.0 MPa√m for the sample Y1210-SC and 4.5 MPa√m for the sample Y1210-EY (Table 1).

On average, lower crack lengths were measured in the α-Sialon sample Y1210-EY in comparison to those in the material Y1210-SC.

The measured crack opening displacements were plotted as function of the calculated crack opening displacements with K = 1 MPa√m. A linear fit was obtained between these values with the least-squares method, the slope of the line results in the crack tip toughness (Figs. 2, 3; Table 1). In Fig. 2, measured and calculated crack opening displacements are presented for sample Y1210-SC. The slope of the best linear fit gives a crack tip toughness of 1.35 MPa√m (R^2 = 0.94). The results of the measurement on another crack of the α-Sialon with the stoichiometric composition are presented in Table 1 and give a crack tip toughness of 1.68 MPa√m (R^2 = 0.87). In Fig. 3, measured crack opening displacements are presented for the α-Sialon with excess Y_2O_3 (sample Y1210-EY). Using the least squares fit method for the initial portion of the data ($2\delta_{calc}$ < 22 nm), the crack tip toughness of 1.01 MPa√m (R^2 = 0.98) was obtained for one crack (Fig. 3) and 0.95 MPa√m (R^2 = 0.89) for another crack. Table 1 lists all obtained values for the crack tip toughness with the goodness of fit (R^2). Comparing the crack opening displacement diagram of sample Y1210-SC in Fig. 2 to that of the material Y1210-EY in Fig. 3, it is obvious that the entire data is represented by one linear function for α-Sialon with the stoichiometric composition (sample Y1210-SC), but at least two linear functions with different slopes are necessary for the linear

approximation of the α-Sialon with excess Y_2O_3 (sample Y1210-EY): a lower slope linear function near the region of the crack tip and a higher slope linear trend away from the crack tip. This behavior was also reported for crack tip toughness measurements of Si_3N_4 [19] and could be a result of the higher occurrence of bridging effects owing to the elongated grains in α-Sialon with excess Y_2O_3 (sample Y1210-EY). Bridging stresses were evaluated in silicon nitrides with elongated grain structure by Fünfschilling *et al.* in [21]. They found maximum bridging stresses at small, about 0.02 μm, crack opening displacements at the initial part of the R-curve, which may explain the low slope behavior in Fig. 3 at $2\delta \leq 20$ nm. When comparing the results in Fig. 2 and Fig. 3, another important point is that there is a much higher scatter in the crack opening displacement in sample Y1210-SC than in that of sample Y1210-EY. This may be explained by the microstructure and crack growth behavior of these α-Sialons. In sample Y1210-EY the crack growth has mainly transgranular character (Fig. 4), which results in a relatively straight crack, but in sample Y1210-SC, the crack growth is mainly intergranular which results in a zig-zag type crack propagation (Fig. 5). 2δ value depends on x but also on the orientation of the grains when the crack propagates intergranularly. This causes higher scattering in 2δ values in sample Y1210-SC than in sample Y1210-EY, see Figs. 2 and 3.

The obtained crack tip toughness values could not be compared to α-Sialon literature data because no data has been reported yet. At least the resulting crack tip toughness values can be compared to data reported for Si_3N_4 ceramics. In this work for the crack tip toughness of α-Sialon with a stoichiometric composition a value of approximately 1.5 MPa√m was determined while for the α-Sialon with excess Y_2O_3 a value of 1 MPa√m was measured. For sintered, reaction-bonded Si_3N_4 containing yttria and alumina, Kounga Njiwa *et al.* [18] retrieved values of 1.6 MPa√m and 1.94 MPa√m. Fünfschilling *et al.* [19] reported for Si_3N_4 with yttria and magnesia values of 2.33 MPa√m and 2 MPa√m for Si_3N_4 with yttria and alumina, respectively. Our measurements on the crack tip toughness of the α-Sialons are lower but close to the reported values for the Si_3N_4 ceramics.

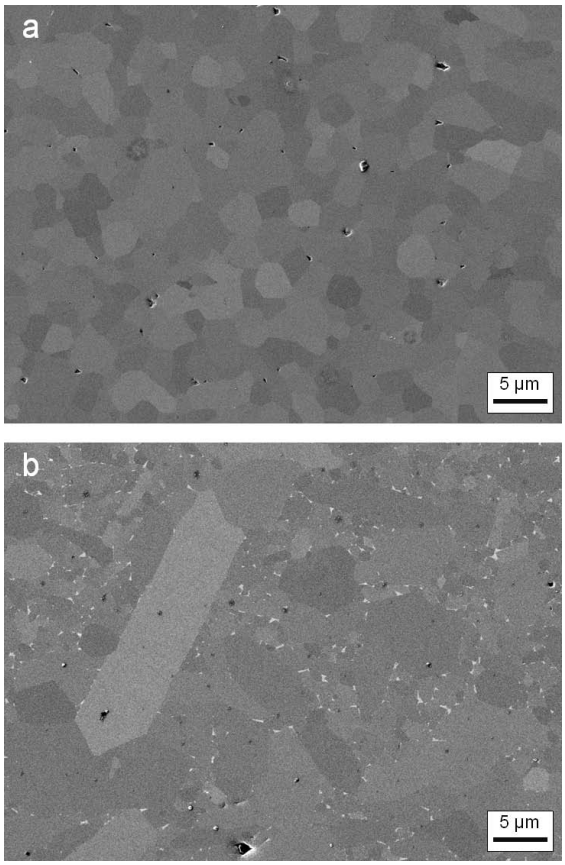

Fig. 1: SEM micrographs of the α-Sialon with stoichiometric composition (Y1210-SC) (a), α-Sialon with excess Y_2O_3 (Y1210-EY) (b) (bright regions: amorphous Y-rich grain boundary phase).

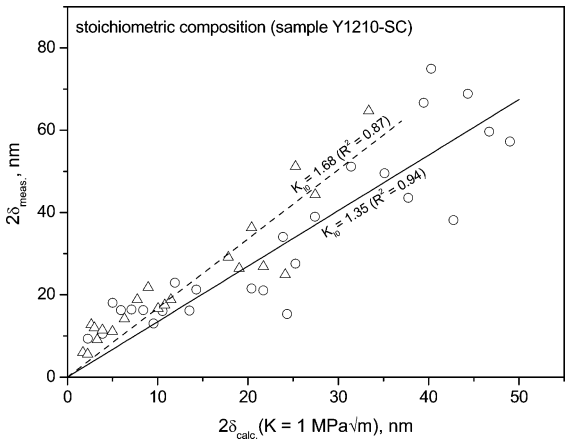

Fig. 2: Measured and calculated crack opening displacement and least-squares fit to determine crack tip toughness for two different cracks of the α-Sialon with the stoichiometric composition (sample Y1210-SC).

Table 1: Compositions and properties of the investigated Sialon materials. ($Y_xSi_{12-(m+n)}Al_{m+n}O_nN_{16-n}$, where x = m/3)

Sample	**m**	**n**	**Composition,** wt% Si_3N_4	AlN	Y_2O_3	Y_2O_3 **(Excess)**	**Density,** g/cm^3 (**Rel. density**, %)	**Hardness HV$_{10}$,** GPa	**K_{IC} (SEVNB),** MPam$^{1/2}$	**K_{I0},** MPam$^{1/2}$
Y1210-SC	1.2	1	77.22	15.24	7.54	0	3.28 (99.7)	18.0 ±0.2	3.0 ±0.1	1.35 ±0.07 (R^2 = 0.94) 1.68 ±0.09 (R^2 = 0.87)
Y1210-EY	1.2	1	76.07	15.01	8.92	1.5	3.30 (99.7)	17.7 ±0.4	4.5 ±0.1	1.01 ±0.03 (R^2 = 0.98) 0.95 ±0.09 (R^2 = 0.89)

The crack tip toughness values of sample Y1210-EY are lower than those of sample Y1210-SC. By comparing the microstructures and indentation crack morphologies of these α-Sialons having different chemistry and microstructure, it was possible to rationalize their different fracture toughness and crack tip toughness values. α-Sialon with excess Y_2O_3 (sample Y1210-EY) has mixed types of grains: large elongated grains, medium equiaxed grains and small equiaxed grains and between these grains triple grain junctions filled with a yttria-rich amorphous secondary phase (Fig. 1 b). In contrast, the α-Sialon with the stoichiometric composition (sample Y1210-SC) has only medium and small equiaxed grains, and no elongated grains and secondary phase are formed in this sample (Fig. 1 a). The crack path in sample Y1210-EY is both transgranular through mainly large grains and intergranular through the secondary phases in the region of the crack tip and away from the crack tip (Fig. 4). In contrast, the crack path in sample Y1210-SC is mainly intergranular (Fig. 5).

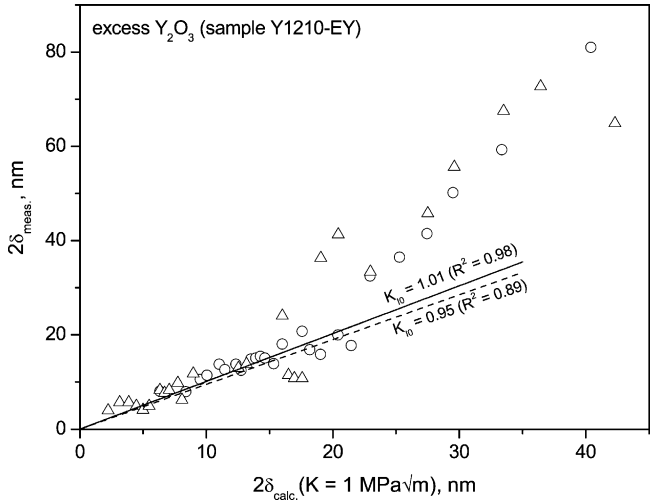

Fig. 3: Measured and calculated crack opening displacement and least-squares fit line of the initial portion of the data to determine crack tip toughness of the α-Sialon with the excess Y_2O_3 (sample Y1210-EY).

The higher fracture toughness (4.5 MPa√m) of sample Y1210-EY and lower crack tip toughness of about 1 MPa√m are explained by the special character of the microstructure of the material. Large elongated grains help to improve the fracture toughness of this α-Sialon with excess Y_2O_3 (Y1210-EY) based on a bridging mechanism, resulting in higher fracture toughness in comparison to the fracture toughness of 3 MPa√m of the α-Sialon with a stoichiometric composition (sample Y1210-SC). In contrast to the fracture toughness values, sample Y1210-SC has a slightly higher crack tip toughness of 1.35 – 1.68 MPa√m than the sample Y1210-EY which is caracterized by a value of about 1 MPa√m. This can be attributed to the bridging effects due to the long elongated grain structure in α-Sialon with excess Y_2O_3 (sample Y1210-EY), which causes lower crack tip opening displacement, meaning lower crack tip toughness. With measurement of the crack tip toughness and fracture toughness, it would be possible to comment on the R-curve behaviour in ceramic materials. Sample Y1210-EY is characterized by a steeper R-curve than sample Y1210-SC. This is again mainly due to the difference in microstructure of these Sialons. Large elongated grains in α-Sialon with excess amount Y_2O_3 (sample Y1210-EY) help to improve the fracture toughness and cause increasing R-curve behavior by bridging effects behind the crack front.

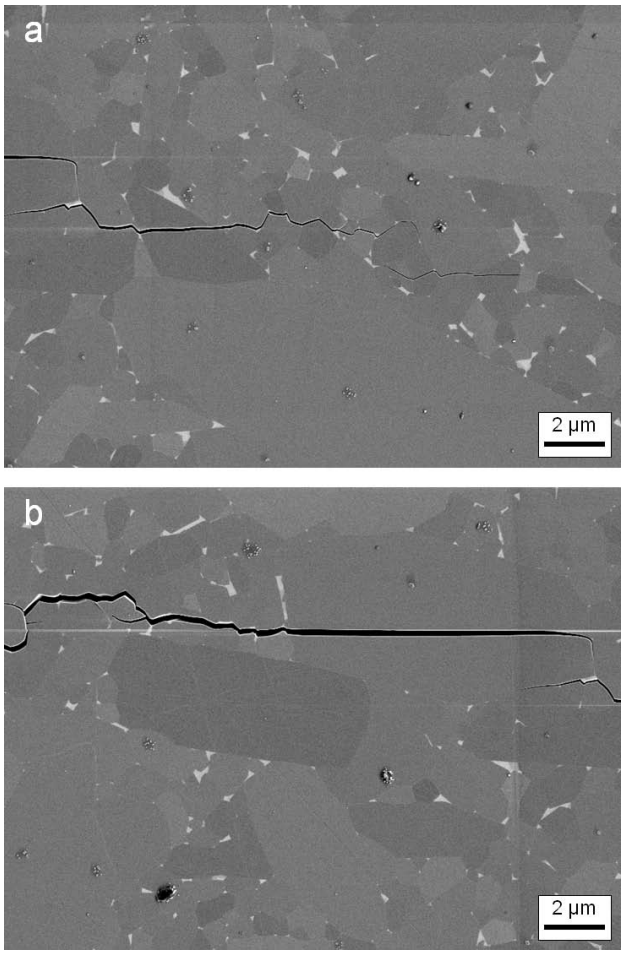

Fig. 4: SEM micrograph of the crack path of α-SialonSialon with excess Y_2O_3 (sample Y1210-EY), a) near the crack tip and b) away from the crack tip.

IV. Conclusions

Crack tip toughness measurements were performed on the basis of Vickers indentation induced cracks in two α-Sialon materials, one with a stoichiometric composition (sample Y1210-SC) and the other one with an excess amount of Y_2O_3 (sample Y1210-EY). Crack tip toughness of about 1 MPa√m and 1.35 – 1.68 MPa√m were measured for sample Y1210-EY and sample Y1210-SC, respectively. Fracture toughness measurements by means of SEVNB resulted in 4.5 MPa√m and 3 MPa√m for the sample Y1210-EY and sample Y1210-SC, respectively. The difference between the toughness values of these two α-Sialons is the result of their different microstructures owing to a variation in their chemistry. The main differences between the microstructures of these α-Sialons are as follows: a) the α-Sialon with excess Y_2O_3 (sample Y1210-EY) has mixed grains, both elongated and equiaxed and a secondary amorphous phase which remains from the liquid phase sintering; b) α-Sialon with stoichiometric composition (sample Y1210-SC) has mainly equiaxed grains. It is well known that an elongated grain structure in ceramics

helps to improve the fracture toughness due to the occurrence of bridging mechanisms.

A larger difference between the fracture toughness and the crack tip toughness of the α-Sialon with excess Y_2O_3 (sample Y1210-EY) and the α-Sialon with stoichiometric composition (sample Y1210-SC) would mean a higher increasing R-curve behavior for the material on the basis of the non-stoichiometric composition.

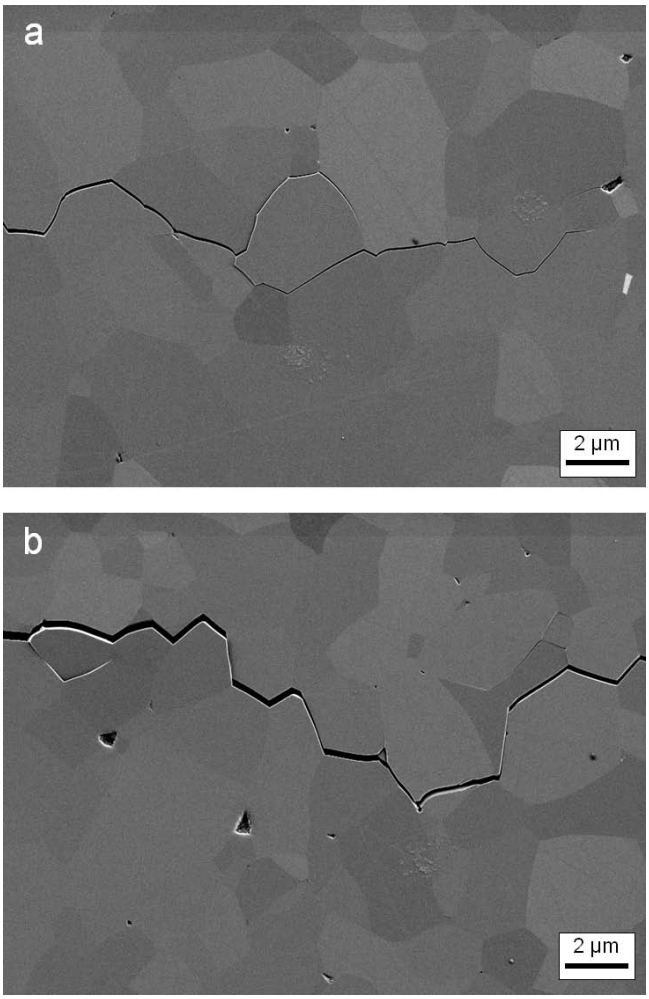

Fig. 5: SEM micrograph of the crack path in the α-SialonSialon with stoichiometric composition (sample Y1210-SC), showing mainly intergranular crack propagation a) near the crack tip and b) away from the crack tip.

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