

## Possible effects of elastic anisotropy on mechanical properties of ceramics

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Intrinsic microstructural stresses, i.e. stresses arising from intrinsic property differences between adjacent grains in an essentially single-phase body or between adjacent grains of different phases in mixed-phase or composite bodies, have been attracting increasing attention [1-3]. An extensively studied source of intrinsic microstructural stresses is that due to phase transformation of metastable tetragonal  $ZrO_2$  particles, grains or portions thereof. Considerable attention has also been directed towards evaluation of effects of impurity particles on mechanical properties, and particularly, more recently, on effects of dispersed second phases on mechanical properties of resultant composites. Considerable attention has also been given to intrinsic stresses from thermal expansion anisotropy (TEA) due to anisotropic thermal expansion in non-cubic crystal structures [1, 4]. Thus, for example, microcracking has been attributed to microstructural stresses generated as a result of phase transformations, thermal expansion mismatches between different phases as well as due to TEA. More recently, it has been argued that stresses from either phase transformations or TEA can increasingly contribute to mechanical failure as the flaw size decreases closer to the scale of the grain structure [2, 5]. Such effects have been related not only directly to the strengths of bodies but also to a decreasing scale of mist and hackle boundaries relative to the strength of the body as strength increases. In all of these cases, these microstructural stresses are determined entirely by the basic properties of the material and are not dependent upon a macroscopic, e.g. external, stress applied to the body. TEA stresses occur extensively with non-cubic materials, and many transformation stresses also commonly involve non-cubic structures.

Another intrinsic source of microstructural stresses is elastic anisotropy (EA) which occurs in both cubic and non-cubic crystal structures [6]. However, in contrast to the above stresses which exist in the absence of any applied stress, elastic anisotropy can only redistribute, on a local microstructural scale, the applied stresses. Thus, the magnitude of microstructural stresses associated with EA are directly related to the applied stress.

Hasselmann [7] previously presented analysis showing basic EA-grain structural effects on mechanical properties of ceramics. His model shows that stress concentrations from EA in polycrystalline ceramics increases with the ratio of the maximum to minimum Young's modulus of the crystal structure ( $E_{max}/$

$E_{min}$ ) and as the anisotropy of the grain shape increases. The orientation of the long axis of elongated grains is important. Thus, the maximum stress concentrations occur when: (1) the grain long axis and the uniaxial stress axis are parallel with a crystal direction having a Young's modulus greater than the polycrystalline modulus, or (2) the stress is normal to the long grain axis which is parallel to a crystal direction having a Young's modulus less than the polycrystalline modulus. The nature of the stress also has some effect, e.g. ~25% less stress concentration for biaxial versus uniaxial stress. Hasselmann showed that, in the limit of highly elongated grains for the above two limiting orientations, the resultant stress concentration approaches  $E_{max}/E_{min}$  (which as shown later can be 2 or more). He argues that EA affects the stress (and scatter) for failure at room temperature, the character of biaxial failure, as well as high temperature failure (although the temperature dependence of EA was not considered), again with grain elongation (e.g. of isolated large grains) being important.

Subsequently, EA has been cited as a possible factor in other mechanical behaviour of ceramics, most often based on a general analogy between TEA and EA stresses [8]. Examples are as a factor in positive slopes of the finer grain size branch of strength-(grain size)<sup>-1/2</sup> plots, large grain polycrystalline strengths greater than those of the weakest oriented single crystals (with comparable surface finish) [9], spalling along grain boundaries around hardness ( $H$ ) indents and associated  $H$  minima [10], and grain size dependence of polycrystalline fracture toughness/energy of cubic ceramics [11]. For the latter, it was suggested that the modest maxima indicated in fracture toughness or fracture energy of some cubic ceramics versus grain size might be due to EA effects similar to often much more pronounced maxima in  $K_{IC}-G$  trends for non-cubic materials. The above  $H$  minima tended to correlate inversely with EA of cubic MgO, SiC,  $MgAl_2O_4$  and ZnS, i.e. lower relative  $H$  minima with higher EA. Similarly, for non-cubic materials,  $Al_2O_3$  and BeO with moderate TEA and EA had similar  $H$  minima as MgO and SiC, while  $TiB_2$  with higher TEA and EA had a more pronounced  $H$  minimum.

This paper compiles data on EA of ceramics, and suggests some other possibilities of EA effecting mechanical behaviour of ceramic materials. Effects in both cubic and non-cubic materials are considered after first summarizing elastic anisotropy behaviour.

While one of the purposes of this paper is to stimulate further interest and study in possible effects of EA on mechanical behaviour, another is to emphasize the fact that mechanical behaviour often depends on more than one parameter, and EA may often be one of these.

Chung and Bussem [6] define the percentage EA,  $A^*$  for cubic materials as

$$A^* = \frac{3(A-1)^2}{3(A-1)^2 + 25A} \quad \left( A = \frac{2C_{44}}{C_{11} - C_{12}} \right) \quad (1)$$

where the above Cs are the usual single crystal elastic coefficients. Their review shows high  $A^*$  values (35–50%) are mainly for alkali metals and heavier alkali halides, with most cubic ceramics having  $A^* < \sim 10\%$  (Table I). Among cubic ceramics,  $\beta$ -SiC, ZnS, ZnSe, UO<sub>2</sub>, MgAl<sub>2</sub>O<sub>4</sub>, and ZrO<sub>2</sub> [11, 12] have high EA (the latter also shows significant compositional dependence, Fig. 2). For non-cubic materials, characterization of EA requires two parameters, rather than one:  $A^*$  compression and  $A^*$  shear (Table I). Of these,  $A^*$  shear (which involves  $A$  of Equation 1), should be more important to tensile failure since it correlates with  $E_{\max}/E_{\min}$  in the same fashion for cubic materials (Fig. 1). Compared to cubic materials, most non-cubic

ceramics have relatively low EA values, e.g. Al<sub>2</sub>O<sub>3</sub> and BeO, reinforcing the implications that their TEA is more important in their properties (Table II), while EA may be more important for cubic ceramics. However, the elastic anisotropy of some non-cubic materials is often not negligible and can be substantial (e.g. TiO<sub>2</sub>, Table I).

Plotting available  $A^*$  data for ceramics and related materials versus temperature shows a range of behaviour. Some materials of higher EA (i.e.  $A^*$ ), e.g. ZrO<sub>2</sub>, have significant  $A^*$  increases with temperature, as does  $A^*$  of some materials starting from more modest levels (e.g. MgO and LiF, Figs 2 and 3). Some materials of relatively high  $A^*$  show limited change (e.g. SiC, Fig. 3). On the other hand, some show substantial decreases with increasing temperature (e.g. BaO, Fig. 2, and KCl and KBr, Fig. 3). These differences cannot be attributed to differences in homologous temperatures since, for example, LiF, KCl, and KBr all have melting points in the 730–845 °C range.

The primary mechanical effect of elastic anisotropy is to significantly perturb the level of applied stress at or near grain boundaries. Thus, a key to identifying possible effects of EA on mechanical behaviour is to consider behaviour that is associated with substantial intergranular failure. Clearly, a

TABLE I Elastic anisotropy of ceramic and related materials at 22 °C

(A) Cubic materials	$A^*$	$E_{\max}/E_{\min}$	Material	$A^*$	$E_{\max}/E_{\min}$
Material	(%)			(%)	
Si	2.4	1.45	SrO	0.0–2.1	–
Ge	3.0	1.50	ThO <sub>2</sub>	2.9	1.52
C (diamond)	0.4–0.7	1.14–1.21	ZrO <sub>2</sub>	6–10	~2.2
ZnS	8.4	2.1	UO <sub>2</sub>	6.8	1.93
ZnSe	11.7	2.4	MgAl <sub>2</sub> O <sub>4</sub>	6.9	1.91
CaF <sub>2</sub>	3.53	1.56	Garnets	0.01–0.1	1.02–1.09
SrF <sub>2</sub>	0.8	1.23	BaTiO <sub>3</sub>	8.8	2.05
BaF <sub>2</sub>	0.1	1.03	SrTiO <sub>3</sub>	0.3	1.1–1.4
BaO	2.1	–	NbC	1.8	–
CaO	0.0	–	$\beta$ -SiC	7.3	1.9
CoO	1.5	–	TaC	–	2.3
FeO	0.0	–	TiC	0.1–0.9	1.1
MgO	2.3	1.42	UC	4.3	1.67
MnO	2.2	–	ZrC	0.3–1	1.2
NiO	1.6	–			
(B) Non-cubic materials		$A^*$ comp		$A^*$ shear	
1. Hexagonal		(%)		(%)	$E_{\max}/E_{\min}$
BeO		0.02		0.4	1.24
$\beta$ -Quartz		0.09		1.2	
ZnO		0.05		0.5	
BaTiO <sub>3</sub>		0.06		0.5	
TiB <sub>2</sub>		4.4		6.3	2.0
$\alpha$ -SiC		0.01		1.1	1.3
H <sub>2</sub> O		0.004		1.1	
2. Trigonal					
CaCO <sub>3</sub>		4.1		16.1	
$\alpha$ -Al <sub>2</sub> O <sub>3</sub>		0.1		1.7	1.2–1.4
$\alpha$ -Fe <sub>2</sub> O <sub>3</sub>		0.5		1.5	
$\alpha$ -Quartz		0.6		7.6	
3. Tetragonal					
BaTiO <sub>3</sub>		5.4		8.0	
TiO <sub>2</sub> (rutile)		1.8–2.1		11.3–10.9	2.1
ZrSiO <sub>4</sub>		5.0		8	

Most data from Chung and Bussem [6], other values from references 12–20

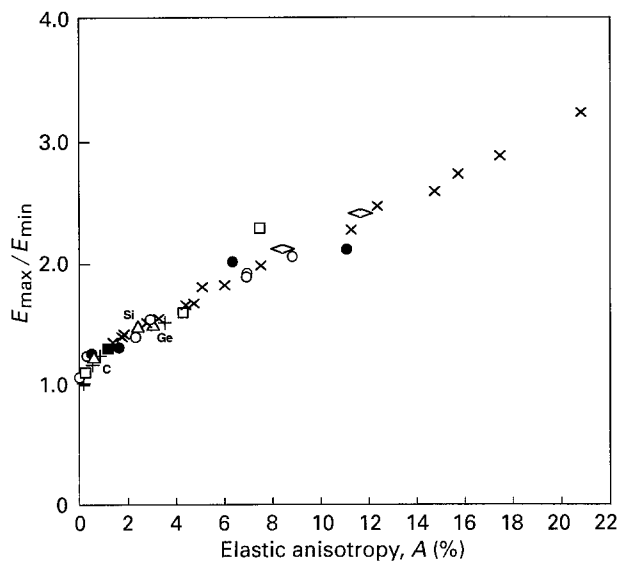


Figure 1 Plot of the ratio of maximum to minimum Young's modulus in a given material crystal structure versus percentage elastic anisotropy ( $A^*$ ). No continuity of correlation for both cubic and non-cubic materials (using  $A^*$  shear for the latter). ( $\times$  Alkali halides (cubic);  $+$  Alkaline earth halides (cubic);  $\square$  Carbides (cubic);  $\blacksquare$  Carbides (non-cubic);  $\circ$  Oxides (cubic);  $\bullet$  Oxides (non-cubic);  $\triangle$  Diamond (c), Ge, Si;  $\diamond$  Chalcogenides (cubic).

critical issue in relating behaviour to intergranular failure is the effects of grain boundary impurities, which can be quite prevalent.

Several materials exhibiting slow crack growth (SCG) due to moisture effects do so predominantly, if not exclusively, via intergranular failure. This raises the question of whether grain boundary stresses due to EA (TEA, or both) may be a contributor to, i.e. enhance, such failure. Thus note that cubic ZnSe has quite high elastic anisotropy and exhibits SCG via entirely intergranular failure [23, 24]. (Since samples tested were of very high purity, made by chemical vapour deposition for infrared window applications, the probability that such intergranular failure is due to grain boundary impurities seems limited [23].) Substantial EA in cubic ZnS (Table I) would suggest evaluation of whether this material exhibits SCG and, if so, whether it is predominantly or exclusively by intergranular failure in similar high purity, e.g. IR window material. Similarly, it should be asked if relatively high EA or cubic  $\text{BaTiO}_3$ ,  $\text{MgAl}_2\text{O}_4$  and  $\text{UO}_2$  as well as SiC could contribute to SCG in them.

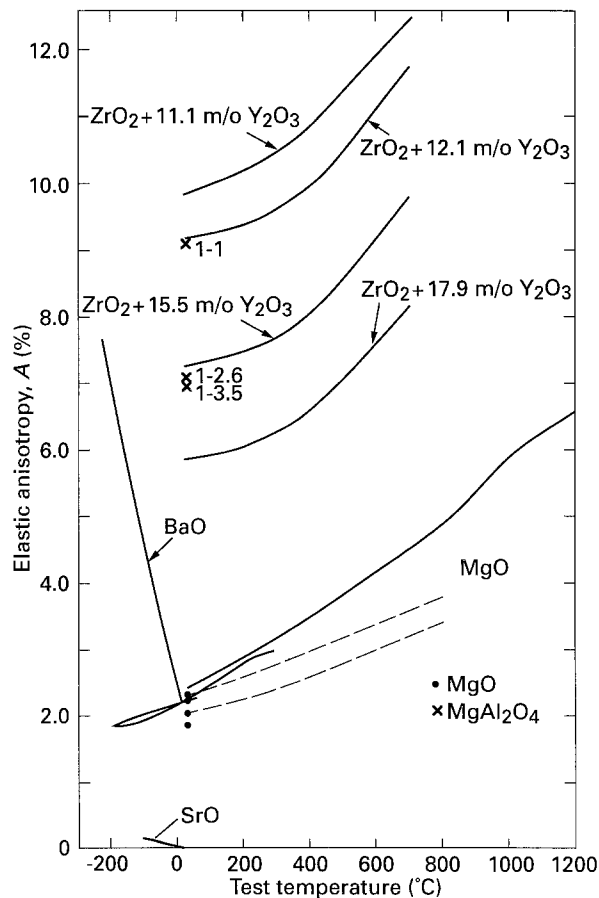


Figure 2 Elastic anisotropy ( $A^*$ , %) versus test temperature for oxides. Note decreases of  $A^*$  with increasing temperature (at lower temperatures) of SrO and especially BaO versus increases with increasing temperature (especially at higher temperatures) for MgO and  $\text{ZrO}_2$ . Note also the trend for increasing EA with decreasing  $\text{Y}_2\text{O}_3$  content of the latter.  $\bullet$  MgO;  $\times$   $\text{MgAl}_2\text{O}_4$ .

However, there is limited or no SCG data on most of these materials, and where there is SCG data, the fracture mode is often not characterized in detail or not given at all (especially fracture mode differences between failure initiation and more general crack propagation). Such enhancement of SCG by EA requires that SCG exist in the material, presumably intergranularly (whether due to intrinsic or extrinsic boundary factors). It also presumes that there are a sufficient number of contiguous or nearly contiguous grain boundaries on which EA enhances the applied tensile stress. However, the occurrence of transgranular, i.e. single crystal, SCG, and the ease and

TABLE II Crystallographic thermal expansion and elastic anisotropy of three non-cubic oxides

Material	Crystal structure	Thermal expansion <sup>a</sup> coefficient ( $10^{-6} \text{ C}^{-1}$ )	Young's modulus <sup>b</sup> (GPa)
$\text{Al}_2\text{O}_3$	Corundum (trigonal)	$\alpha_a = 8.6$	425
		$\alpha_b = 8.6$ $av = 8.9$	$425/E_p = 420$
		$\alpha_c = 9.5$	$461/E_{min} = 372$
BeO	Wurtzite (hexagonal)	$\alpha_a = 9.6$	397
		$\alpha_b = 9.6$ $av = 9.2$	$397/E_p = 400$
		$\alpha_c = 8.5$	$450/E_{min} = 380$
$\text{TiO}_2$	Rutile (tetragonal)	$\alpha_a = 8.3$	153
		$\alpha_b = 8.3$ $av = 8.9$	$153/E_p = 285$
		$\alpha_c = 10.2$	385

<sup>a</sup>From 20–1020 °C, after Bayer [21]

<sup>b</sup>From Ingel [22],  $E_p$  = polycrystalline Young's modulus

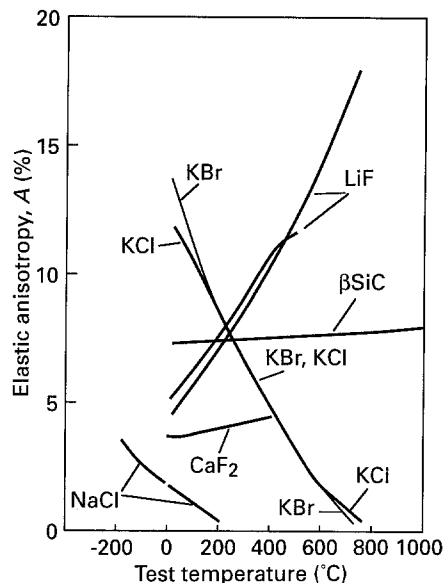


Figure 3 Elastic anisotropy ( $A^*$ , %) versus test temperature for non-oxides. Note diversity of trends, i.e. increases, decreases, or roughly no change with temperature.

multiplicity of such transgranular fracture opportunities, can limit, or preclude such intergranular SCG. For example,  $MgAl_2O_4$ , which has reasonably high EA, shows intrinsic SCG in single crystals on {100} and {110} planes [25]; SCG on these cleavage planes in polycrystalline grains probably precludes much, if any, grain boundary SCG. Thus, the presence of two cleavage planes, both exhibiting SCG, may limit intergranular SCG.

Next consider broader aspects of intergranular failure. Fracture initiation at  $\sim 22^\circ C$  often occurs from grain boundaries, e.g. in PSZ materials [26–28] as well as some  $MgAl_2O_4$  [26], in contrast to most subsequent crack propagation being transgranular [24]. This may indicate possible preferential intergranular fracture initiation in such materials with relatively high EA. This appears to entail effects beyond SCG, since the extent of intergranular fracture significantly rises over a few hundred degrees Centigrade, especially for  $ZrO_2$  where it increased intergranular fracture often accompanied by considerable Young's modulus and flexure strength decreases [25], e.g. those of Drachinskii et al. [29]. While this has been related to changes in  $ZrO_2$  lattice defects [26], corresponding EA changes may be occurring and hence be a mechanism of how such defect changes effect mechanical behaviour.

Changes in transgranular fracture of other ceramics with temperature further indicates correlation with accompanying EA changes. Thus, at  $22^\circ C$ , total transgranular fracture initiation was observed in flexure testing of fully dense, high quality MgO and CaO, e.g. as obtained by recrystallizing single crystals [27, 30, 31], consistent with the modest EA of MgO and the even lower EA of CaO. As test temperatures increased, some intergranular failure initiation (but not general crack propagation) begins to occur in such MgO and CaO, but much less than in less pure materials. However, similar levels of

intergranular fracture require higher temperatures in such higher purity CaO than MgO. Thus, such MgO shows measurable increases in failure initiation (but not subsequent crack propagation) along grain boundary surfaces at up to  $1300^\circ C$ , while CaO is just beginning to show such behaviour at around  $1300^\circ C$ . The fact that such increases in intergranular fracture initiation in these latter two materials commences at substantially higher temperatures than in  $ZrO_2$  is consistent with the EA of MgO and especially CaO, starting at substantially lower levels than  $ZrO_2$ , thus requiring higher temperatures to reach EA levels approaching those of  $ZrO_2$  (and  $MgAl_2O_4$ ) at room temperature. High EA at higher temperatures may also contribute to grain boundary sliding and related phenomena. While grain boundary impurities may commonly be a factor in such intergranular fracture, and are typically cited (often without specific proof) as sources of such intergranular fracture, the increase in EA with temperature, though not apparently previously considered, deserves much more attention. Clearly such EA and impurity effects can be synergistic.

Note that one might possibly expect the increasing ease of slip in materials such as MgO and CaO with increasing temperature to ameliorate the propensity for intergranular failure. This is not necessarily a valid assumption, since grain boundaries are important barriers to slip, and slip pile-up at grain boundaries creates stresses there. This is strongly supported by the fact that room temperature slip-induced fracture is always observed to initiate from slip bands blocked at grain boundaries [30].

Consideration of the effects of elastic anisotropy on mechanical behaviour for non-cubic materials presents two complications. The first is that non-cubic materials require for EA parameters ( $A^*$  compression and  $A^*$  shear) rather than one. However, as noted earlier, of these  $A^*$ , shear should be pertinent to tensile failure, since it shows the same correlation with  $E_{max}/E_{min}$  as for cubic materials (Fig. 1).

The second, more basic and complex, complication is the issue of interaction between EA and TEA (Table II). While for many non-cubic materials this is not a serious issue since, as shown in Table I, many non-cubic materials have quite limited EA, so any effects of microstructural stresses on the mechanical behaviour must be dominated by their TEA. However, some non-cubic materials, e.g.  $TiB_2$ ,  $CaCO_3$ ,  $\alpha$ -quartz,  $BaTiO_3$ ,  $TiO_2$  and  $ZrSiO_4$  have rather substantial elastic anisotropies, in particular unusually high  $A^*$  shear values. At modest temperatures, the issue of the interaction between EA and TEA is thus important for such materials. Thus, EA may moderate or enhance microstructural stresses along a given grain boundary as a result of it, respectively, decreasing or increasing TEA stresses. That other factors, possibly EA interacting with TEA, may play a role in non-cubic materials is suggested by the observation that  $Al_2O_3$ , BeO and  $B_4C$  all have similar levels of TEA, but quite wide variations in their degree of intergranular fracture

[26]. Thus,  $\text{Al}_2\text{O}_3$  typically ranges from 50% to 100% intergranular fracture [27], while  $\text{B}_4\text{C}$  typically exhibits very little and most commonly no intergranular fracture, with  $\text{BeO}$  intermediate between these two. Clearly there may be other factors impacting these differences, for example,  $\text{B}_4\text{C}$  often exhibits extensive mechanical twinning and alumina some and apparently  $\text{BeO}$  little or none [26]. Similarly, while  $\text{TiO}_2$  has somewhat higher TEA than the above three materials, it also exhibits considerable transgranular failure, including more transgranular microcracks spontaneously generated when the grain size reaches the critical value [4, 32]. Whether the high EA of  $\text{TiO}_2$  interacts with its substantial TEA is unknown, but deserves further investigation. However, a complication is that EA does not totally correlate with TEA. Thus, while the maximum Young's modulus of  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$  correspond with the maximum thermal expansion coefficients, the opposite is true for  $\text{BeO}$  (Table II). Further, the minimum Young's moduli for  $\text{Al}_2\text{O}_3$  and  $\text{BeO}$  do not correlate with any of the three principal axes. This is not surprising since cubic materials have EA, often substantial EA, yet are isotropic in thermal expansion.

As temperature increases, TEA stresses must inherently decrease, while presumably the EA of non-cubic materials often increase in some fashion similar to that often occurring for cubic materials. Thus, where EA and TEA work together to enhance microstructural stresses, these opposite temperature dependencies would tend to reduce the rate of decrease of microstructural stresses, thus possibly increasing a probable propensity for intergranular failure as temperature increases.

Consideration of possible EA effects on mechanical behaviour in ceramics suggests three possible effects. The first is a probable correlation between increasing elastic anisotropy (e.g. in  $\text{CaO}$ ,  $\text{MgO}$ , and  $\text{ZrO}_2$ ) with increasing temperature and associated respective increasing intergranular fracture. It is clear that grain boundary impurities to which such increased intergranular failure with increasing temperature is generally attributed, are also commonly a factor. However, it is now clear that one should not arbitrarily assume that grain boundary phases are entirely the cause of such intergranular failure and neglect EA. There are clearly other complications, e.g., while  $\text{MgAl}_2\text{O}_4$  has high EA, it has two low  $K_{\text{IC}}$  cleavage planes ( $\{100\}$  and  $\{110\}$ ) rather than one like  $\text{CaO}$  and  $\text{MgO}$  ( $\{100\}$  and  $\text{ZrO}_2$  (presumably  $\{111\}$ ), thus potentially enhancing opportunities for transgranular fracture in  $\text{MgAl}_2\text{O}_4$ . Another, more general complication is the interaction of EA and TEA in non-cubic materials. Thus, while EA may also enhance intergranular fracture in such materials, effects of impurities, twinning, the ease of cleavage, the extent of correlation of EA and TEA, and possible opposite temperature trends of EA and TEA, can all be complications.

More speculative, but still important to consider and conduct more study on, are the issues of EA possibly contributing to enhanced slow crack

growth, mainly via intergranular fracture, in the presence of active species such as moisture. Such study should be less complicated in cubic materials, but possible interactions of TEA and EA need to be addressed in non-cubic materials, again recognizing the above-noted complications. Possible effects of EA on fracture toughness or energy as a function of grain size deserve further attention in cubic materials, and possible interactions with TEA effects in non-cubic materials deserve attention as well.

Possible effects of EA further reinforce the argument that there may often be several factors contributing to mechanical behaviour and that assumptions of a single dominant mechanism may often be a substantial oversimplification in two fashions. The first is that EA may be a factor in some mechanical behaviour. Second, it is clear that EA can interact with TEA in a variable fashion, i.e. either enhancing or diminishing of TEA effects at low temperatures, but the two phenomena have opposite temperature dependencies. Clearly, interactions with other effects, e.g. of twinning and cleavage, must be considered.

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