ON THE FRACTURE TOUGHNESS OF BRITTLE MATERIALS

J. D. B. VELDKAMP *) and N. HATTU

Abstract
This paper deals with the influence of several structural parameters, such as porosity, grain boundaries and second-phase inclusions on the fracture toughness of brittle materials. Experiments have been carried out with regard to the influence of grain size, grain-boundary segregation and second-phase inclusions on fracture toughness. Experimental data from the literature on the influence of for instance porosity on fracture toughness have also been used. The main tool used for the explanation of the phenomena observed, is the process zone of the propagating crack, which is defined here by an effective size and an effective surface area per unit volume created during propagation. It is shown that the parameters often have counteracting effects and that it is difficult to vary one particular parameter. Porosity will always have a negative influence. Instead of the grain size the fraction of transgranular fracture on the fracture surface appears to determine the fracture toughness. Moreover, the toughness of the boundaries and the anisotropy of the grains are important. The influence of grain-boundary segregation cannot be neglected and needs further investigation. Tough inclusions can under certain conditions have a positive effect which can be further enhanced by thermal stresses. The same holds for microcracking in the matrix material but this effect also needs further investigation.

1. Introduction

Until about twenty years ago the investigation of the mechanical properties of brittle materials was relatively undeveloped. Reasons were the uninteresting low tensile strength of these materials and the wide scatter in this property, which more or less confined their application to constructions in a state of compressive stress. The control of the microstructure, particularly with respect to mechanical properties, was unsatisfactory, due to the absence of good tools for characterization. Since then the knowledge about sintering and the physical description of the microstructure have greatly improved, partly due to techniques like transmission electron microscopy, scanning electron microscopy and Auger electron spectroscopy. Control of the microstructure has consequently been improved as well with the aid, for instance, of new powder preparation techniques, better controlled furnaces and new densification techniques like hot pressing 1). As a result materials with improved and better reproducible properties have become available. Some major breakthroughs have also been achieved

in the understanding of the mechanical properties of brittle materials, amongst others in the description of cracking and fracture. In this paper brittle materials are assumed to show no bulk plastic deformation in tension at room temperature, but brittle fracture at a certain tensile stress. Moreover, it is also assumed that no plastic deformation takes place at the crack tip. Particularly due to the work of Irwin \(^2\) the Griffith equation \(^3\) for the fracture stress of an infinitely extended flat plate provided with a central transverse crack can be written in another way:

\[
K_c = \sigma (\pi c)^{\frac{1}{2}} = (G_c E)^{\frac{1}{2}} = (2 \gamma_c E)^{\frac{1}{2}},
\]

where

- \(K_c\) is the critical stress-intensity factor,
- \(\sigma\) is the stress applied to the ends of the plate at the moment of fracture,
- \(c\) is half the length of the crack,
- \(E\) is Young's modulus,
- \(G_c\) is the critical crack energy release rate and
- \(\gamma_c\) is the effective crack-surface energy.

At high crack speeds, i.e. generally in the case of brittle fracture, \(G_c\) or \(\gamma_c\) can be considered as material properties \(^4\); \(\gamma_c\) can include several energy dissipating mechanisms, such as the formation of new surface, dislocation movement and creation, phase transformations, etc. Generally, \(\gamma_c\) is given for the main crack plane (see fig. 1a); thus \(\gamma_c\) is also increased by a roughening of the crack surface and of the crack tip with respect to that plane. Because \(E\) is a material property, so too \(K_c\) is a material property.

On the basis of calculations of the particular stress distributions and experiments, several test samples have been developed \(^4\) which, under standard conditions, provide reasonably reliable values for \(K_c\).

At first sight, the strength of a product can now be estimated when the stress distribution \(K_c\) and the size of the fracture-initiating flaw are known.

In the shaping of brittle products the use of surface grinding is common. When \(K_c\) is low and the hardness relatively high, then the fracture-initiating flaw will often be caused by this grinding treatment \(^5,6\). When \(K_c\) is high and the hardness low, structural flaws often initiate fracture \(^7\), in spite of the grinding.

The size of the fracture-initiating flaw is not easy to detect \(^7\) and can vary considerably. The resulting variation in strength can be described statistically with the Weibull method \(^8\). Another complication in the determination of the length of the fracture-initiating flaw is found particularly in oxidic materials, where stress corrosion cracking due to water takes place. Cracks grow slowly at a \(K\) value which is lower than \(K_c\) (ref. 9), which means that a surface flaw due to
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Fig. 1. Schematic cross-sections of a crack at the tip: (a) crack with main crack plane, the actual crack path, microcracks, and size of the process zone $2\delta$; (b) idealized crack with main crack plane and asperities of width $2b$ and angle $\varphi$.

grinding becomes more severe when the product is stressed \(^{10}\), but also a structural flaw near the surface increases in length. In practice this means that after a certain time fracture occurs at a certain stress.

All the considerations discussed above form part of linear elastic fracture mechanics; they can lead to the prediction of the strength of a brittle product and also of a safe lifetime \(^{11}\).

There is still a lot of confusion about the influence of structural parameters on the material property $K_c$ in brittle materials. First it is necessary to distinguish between the influence of different structural parameters on $E$ and on $\gamma_c$ (eq. (1)) and then to determine which parameters are really important.

In contrast with the review paper by Rice \(^{7}\) an attempt is made in this paper to study the influence of each structural parameter independent of the other known parameters. The investigation is carried out with the aid of experimental results obtained by other workers and by ourselves. In explaining the observed effects we could use an extension of expression (1). The elaboration of expression (1) is given in sec. 2.

For the investigation of the influence of grain size and second phase on $K_c$ we felt the need for additional experiments, because results obtained from the literature were found to be rather confusing. The preparation of the materials for these investigations and the experiments are described in sec. 3. Because we applied a standard method for the determination of $K_c$\(^{4}\) it was decided that only results from the literature in which similar methods had been applied, could be used for our investigation.
All the relevant results on the influence of structural parameters on $K_c$ are given in sec. 4. They are related to porosity (secs 4.1.1 and 4.1.2 for Young's modulus and crack-surface energy, respectively), grain size (sec. 4.2.1), grain-boundary segregation (sec. 4.2.2), different phases (influence on Young's modulus, crack-surface energy and internal thermal stresses in secs 4.3.1, 4.3.2 and 4.3.3, respectively) and microcracking (sec. 4.3.4). The results are also discussed in these sections with the aid of expressions (1) and (4) (see sec. 2). Conclusions are drawn in sec. 5.

2. The process zone at a crack tip and the crack-surface energy

By analogy with brittle crack propagation in plastics and metals Clausen et al.\textsuperscript{12} propose a process zone which runs in front of the crack tip. Within this zone the dissipation of energy takes place, which, according to the idea of Irwin\textsuperscript{2} contributes to the height of $G_e$ or $\gamma_e$ (eq. (1)). In the case of highly brittle materials where the yield stress is not exceeded at the crack tip, possible mechanisms for this dissipation are for example phase transformations, deviations of the main crack plane (see figs 1a and 1b) and microcracking. In the last two examples the process zone will result in a zone of a certain thickness $2\delta$ adjacent to the main crack plane, which contains deviations of the actual crack path from the main crack plane and additional microcracks of a deviating orientation (see fig. 1a). The size of the process zone is the average maximum height of the zone in which new surface is created. The average size of the process zone can be determined in fig. 2 from the quotient of the shaded area and the thickness of the plate (see sec. 1).

Let the cross-sectional shape of the process zone at the crack tip be semi-circular (fig. 1a) with a radius $\delta$. The crack-surface area per unit volume created during propagation is denoted by $A_e$. Thus the surface area at the crack tip in the semicircular zone is

$$\frac{1}{2} \pi \delta^2 A_e d,$$

where $d$ is the thickness of the plate.

Fig. 2. Schematic longitudinal section perpendicular to the main fracture plane of a crack at the tip. Indicated are the main crack plane, the actual crack path, microcracks and the area over which the size of the process zone has to be determined (see sec. 2).
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The energy dissipated in that zone $W$ is thus

$$W = \frac{1}{2} \pi \delta^2 A_c \gamma,$$  \hspace{1cm} (2)

where $\gamma$ is the crack-surface energy of the crack surfaces created within the zone. One may also say that the energy dissipated at the crack tip is

$$W = 2 \delta \gamma_c d,$$ \hspace{1cm} (3)

where $\gamma_c = \text{the effective crack-surface energy.}$ Combining expressions (2) and (3) gives

$$\gamma_c = \frac{1}{4} \pi \delta A_c \gamma.$$ \hspace{1cm} (4)

For $K_c$ we can therefore write

$$K_c = (2 E \gamma_c)^\frac{1}{2} = (\frac{1}{4} \pi E \delta A_c \gamma)^\frac{1}{2}.$$ 

The process zone develops due to the microstructure of the material.

For a polyphase or porous material $E$ can be calculated for instance with the aid of the method developed by Veldkamp$^{13}$, when the volume fractions, shapes and distributions of the phases are known.

For a material with a randomly distributed second phase where the actual crack path and the microcracks proceed at random through the two phases we may write

$$\gamma = V_1^\frac{1}{d} \gamma_1 + (1 - V_1^\frac{1}{d}) \gamma_2,$$

where

$\gamma_1$ is the crack-surface energy for phase 1,
$\gamma_2$ is the crack-surface energy for phase 2 and
$V_1$ is the volume fraction of phase 1 ($V_1^\frac{1}{d}$ is the surface fraction of phase 1).

In this case one can also write

$$\gamma_c = V_1^\frac{1}{d} \gamma_{c1} + (1 - V_1^\frac{1}{d}) \gamma_{c2}$$ \hspace{1cm} (5)

or for a porous material

$$\gamma_c = (1 - V_p^\frac{1}{d}) \gamma_{c0},$$ \hspace{1cm} (6)

where

$V_p$ is the volume fraction of the pores,
$\gamma_{c0}$ is the effective crack-surface energy for zero porosity.
3. Experimental

We prepared and investigated three materials: alumina, nickel–zinc ferrite and strontium hexaferrite. Sintering of the alumina powder *) (99.995% pure) was carried out with the aid of analytical-grade magnesium acetate **) in the manner described by Peelen 14).

For the preparation of the nickel–zinc ferrite a powder of the composition \( \text{Ni}_{0.36}\text{Zn}_{0.64}\text{Fe}_2\text{O}_4 \) was used. Sintering was carried out according to the description given by Gorter 15).

The strontium hexaferrite was prepared from \( \text{Fe}_2\text{O}_3 \) (99.9% pure) and \( \text{SrCO}_3 \) (99.9% pure) in accordance with the method described by van den Broek and Stuijts 16). Stoichiometric strontium hexaferrite has the formula \( \text{SrFe}_{12}\text{O}_{19} \), which can be written as \( \text{SrO-6(Fe}_2\text{O}_3) \). When the 6 is replaced by \( n \) one can obtain different volume fractions of stoichiometric strontium hexaferrite in a more or less continuous strontium-rich phase by varying \( n \) (\( n < 6 \)). Prior to sintering the hexagonal c-axes of the grains were aligned in a magnetic field.

The grain sizes, densities and impurities of the sintered materials are listed in table I. The chemical analyses were carried out by X-ray fluorescence and atomic absorption analysis.

For the determination of \( K_c \) the standard three point bend test 4) was utilized. At the ends of the notch the samples were precracked on both sides with the aid of cracks made by a sharp indenter 17). The fracture surfaces were studied by means of scanning electron microscopy.

4. Results and discussion

4.1. Porosity

4.1.1. The influence of porosity on Young's modulus

A method that can be used to calculate the influence of porosity on Young's modulus \( (E) \) has been given by Veldkamp 13). For this calculation it is necessary to know the volume fraction, the shape and the distribution of the pores and Young's modulus of the fully dense material. Figure 3 shows calculated curves for \( E \) as a function of volume fraction of the pores for a face-centered cubic array of spheres in the direction of one of the cube axes and for a “close-packed” hexagonal array of spheres in the direction of one of the axes in the basal plane. Also indicated are experimental results by Coble and Kingery 18) for a random array of isolated pores. An example of such a microstructure can be seen in fig. 4a. The calculated curve for the cubic array gives \( E \) values which

*) Rubis de synthétique des Alpes, France.
**) Merck, Germany.
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### TABLE 1

Densities, grain sizes and impurities in the sintered materials

<table>
<thead>
<tr>
<th></th>
<th>Density (%)</th>
<th>Grain size (μm)</th>
<th>Impurities (%)</th>
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<tr>
<td><strong>Al₂O₃</strong></td>
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<tr>
<td></td>
<td>97.9</td>
<td>2.5</td>
<td>Fe 0.0005</td>
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<td></td>
<td>99.1</td>
<td>5</td>
<td>Ga 0.0004</td>
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<td>99.3</td>
<td>6</td>
<td>Si 0.003</td>
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<td></td>
<td>99.6</td>
<td>14</td>
<td>Na 0.001</td>
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<td></td>
<td>99.9</td>
<td>20</td>
<td>K 0.003</td>
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<td></td>
<td>99.9</td>
<td>25</td>
<td>Ca 0.001</td>
</tr>
<tr>
<td><strong>Ni₀.₃₆Zn₀.₆₄</strong></td>
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<td></td>
<td>Pb 0.0007</td>
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<td>Mg 0.0007</td>
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<tr>
<td></td>
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<td>Cl 0.0006</td>
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</tbody>
</table>
are slightly higher than the experimental data; for the hexagonal array the agreement between calculation and experiment is fine. One should take into account that the assumption of spherically shaped pores is not always correct, particularly at relatively high porosities when the pores tend to connect and are situated along the grain boundaries (see fig. 4b). In such a case the porosity can be expected to have a much greater influence on \( E \), which is illustrated by the calculation carried out by Ondracek \(^{19})\) for disc shaped, relatively very compliant inclusions:

At a volume fraction of 0.3 he found \( E \) to be about 20\% lower than according to the hexagonal array line in fig. 3.

4.1.2. The influence of porosity on \( \gamma_\varepsilon \)

Here too, it is necessary to know the fraction, shape and distribution of the pores.

The experiments reported by Evans and Tappin \(^{20})\) are, to our knowledge, the only three measurements that have been made with porosity as the only structural parameter. Their results are shown in fig. 5 together with the curve according to expression (6); the calculated curve falls within the error bars of the experimental points.
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Fig. 4. Microstructure of porosities: (a) isolated pores in a manganese-zinc ferrite, (b) open porosity with elongated pores in an yttrium zirconate.
4.2. Grain boundaries

4.2.1. Grain size

In this section grain boundaries are considered without any segregation. In such a material the grain boundaries have hardly any influence on Young's modulus, so that this effect is left out of account.

In the case of a proceeding crack, weak zones in a completely dense and pure polycrystalline material are the grain boundaries and the cleavage planes. It has been shown \(^{21}\) that even in a material with pronounced cleavage planes, like MgO, the tendency of the crack to follow one of these planes in each grain is only very limited. The fracture surface is irregular due to curved fracture lines and many steps. For materials with not very pronounced cleavage planes, like nickel–zinc ferrite, it can be observed that the transgranular fracture path is even smoothly undulating within the grains (fig. 6).

Due to the stress distribution around the crack tip the crack can follow the weakest path within a zone of limited thickness adjacent to the main crack plane. When a grain boundary is considered as such a weak path the crack can be expected to follow the grain boundaries when the grains are small. When the grain size increases, the grain boundaries become too long with respect to the extent of the stress field, and therefore the crack will increasingly favour the path through the grains \(^{22}\). This can also be understood more analytically.

Consider a grain-boundary material with a size of the process zone \(\delta_b\), a crack-surface area per unit volume \(A_{eb}\) and a crack-surface energy \(\gamma_b\). Then the
energy dissipated in the process zone $W_b$ is

$$W_b = \frac{1}{2} \pi \delta_b^2 A_{cb} \gamma_b$$

see expression (2) (7)

Similarly, the energy dissipated in the process zone of the grain material $W_g$ will be

$$W_g = \frac{1}{2} \pi \delta_g^2 A_{cg} \gamma_g,$$

where

- $\delta_g$ is the radius of the process zone at the crack-tip for the grain material,
- $A_{cg}$ is the crack-surface area per unit volume for the grain material and
- $\gamma_g$ is the crack-surface energy for the grain material.

Let the average width within the main crack plane of the asperities that built up the surface roughness be $2b$ ($b_b$ for intergranular cracking, $b_g$ for transgranular cracking) and the average angle of the asperities with respect to the main crack plane be $\varphi$ ($\varphi_b$ for intergranular cracking, $\varphi_g$ for transgranular cracking). When microcracking is absent, one can write for the radius of the
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process zone (see fig. 1b)

\[ \delta = C_1 b \tan \varphi, \]  

where \( C_1 \) is a constant.

Similarly, for the crack surface per unit volume \( A_c \)

\[ A_c = \frac{C_2}{b \sin \varphi}, \]  

where \( C_2 \) is a constant.

When expressions (9) and (10) are substituted in expression (4) one obtains

\[ \gamma_c = \frac{\pi C_1 C_2 \gamma}{4 \cos \varphi}. \]  

For pure intergranular cracking or for transgranular cracking with unpronounced cleavage planes \( \gamma_c \) (and \( K_c \)) is thus independent of the size of the asperities within the main crack plane. Consequently \( K_c \) is thus independent of grain size for such materials. Expression (7) becomes after substitution of (9) and (10)

\[ W_b = \frac{1}{2} \pi (c_1 b_b \tan \varphi_b)^2 \frac{C_2}{b_b \sin \varphi_b} d \gamma_b, \]  

and expression (8)

\[ W_g = \frac{1}{2} \pi (c_1 b_g \tan \varphi_g)^2 \frac{C_2}{b_g \sin \varphi_g} d \gamma_g. \]  

At the grain size where the transition from intergranular to transgranular cracking takes place one must have

\[ b_b \tan \varphi_b \gamma_b = b_g \tan \varphi_g \gamma_g. \]  

\((\tan \varphi_b / \cos \varphi_b) \gamma_b \) is independent of grain size when the shape of the grains is independent of their size. The same holds for the right hand term of eq. (14) for materials with unpronounced cleavage planes, hence with a roughness that is determined by curved fracture lines, steps etc. Equation (14) is thus only valid for one grain size. But because there is a grain size distribution, the size of the grains, where the transition of intergranular to transgranular fracture takes place, must correspond to a certain fraction of intergranularly cracked grains.
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Though at the transition the energy dissipated in the process zone is equal for transgranular and intergranular cracking, the two effective crack-surface energies differ because $\delta$ is different for the two “phases” (see expression (4)). This means that the two “phases” have different $K_c$ values (expression (1)). The transgranular $\gamma_c$ value will be generally higher than the intergranular one. The height of $\gamma_c$ is therefore determined by the transgranular fraction of the fracture surface via the rule of mixtures.

In figure 7 our results on $K_c$ (or $\gamma_c$) of $\text{Al}_2\text{O}_3$ as a function of grain size are given. On the fracture surface no major grain-boundary segregation could be detected with Auger electron spectroscopy. $K_c$ remains constant within the range of grain sizes investigated. The transgranular fraction (see fig. 8) as a function of grain size remains also constant, which is in agreement with the above reasoning. Certainly, we had expected the same result for the cubic nickel–zinc ferrite. Here, however, $K_c$ decreases with increasing grain size. We will return to that observation in section 4.2.2.

Grain boundaries can also be looked upon as the centre of a stress field that is brought about by the mismatch in thermal expansion between the crystal faces of neighbouring grains. In the case of a homogeneous distribution of grain sizes and grain orientations, one can expect these stresses to be neutralized with respect to a proceeding bulk crack front, due to the interactions of the neighbouring grains. This situation changes when the thermal mismatch is high and when a grain boundary shows such poor coherence that a crack is formed in the boundary. For the formation of such a crack a certain minimum strain energy is required, which is only available above a certain grain size $23)$. Thus this effect leads in the case of intergranular fracture to a lowering of $\gamma_c$ with increasing grain size. In our experiments with alumina, having grains of a rather low anisotropy, even the maximum grain size is apparently insufficient to obtain this effect.

4.2.2. Grain-boundary segregation

Grain-boundary segregation may be called a chemical composition of the grain boundaries which differs from the composition of the grains and which cannot be detected microscopically or with the aid of microprobe analysis, but which has to be detected with Auger electron spectroscopy or with transmission electron microscopy.

An example of the influence of grain-boundary segregation on fracture energy and thus on $K_c$ is MgO hot-pressed with LiF. It was found by Kessler et al. $24)$ that the crack-surface energy (see eq. (1)) doubled when LiF was removed by annealing the material. This increase was accompanied by a transition from intergranular to transgranular fracture. Johnson et al. $25)$
Fig. 7. $K_c$ of $\text{Al}_2\text{O}_3$ as a function of grain size.

Fig. 8. SEM picture of fracture surface of $\text{Al}_2\text{O}_3$. One stripe is ten microns.
showed by means of Auger electron spectroscopy that apart from Ca, Ti and Si, an F-enrichment of the grain boundaries was indeed present in the unannealed material. A tentative explanation is that, due to annealing, F and its weakening influence gradually disappear, which leads to an increasing value of $\gamma_b$ in eq. (13). Consequently the transition grain size decreases and a larger transgranular fraction is present on the fracture surface. This means that increasing values for $\gamma_e$ are obtained.

In an experiment that was meant to demonstrate the expected influence of grain size on $\gamma_e$ of a cubic material (see sec. 4.2.1) we found to our surprise a decrease of $K_c$ with increasing grain size in a nickel–zinc ferrite (see fig. 9). In this case too, no significant change in the fraction of transgranular fracture (26%) was found with varying grain size. A representative fracture surface is shown in fig. 6.

Auger electron spectroscopy revealed that chlorine and calcium from the minute bulk amounts were segregated on the grain boundaries of the fine grained materials. In the coarse grained materials no chlorine and calcium could be detected. Though chlorine can be expected to weaken the grain boundaries the experimental toughness results contradict these expectations. An explanation might be found in deviations of stoichiometry which are well-known in these ferrites. For instance the volume fraction of the Ni- and Zn-rich second phase increases slightly and the shape of the second phase changes when the sintering time is increased (relatively larger grain sizes). Moreover, the oxygen concentration in grains and grain boundaries changes as a function of sintering time. An unambiguous explanation of the observed effect cannot yet be given. It is also conceivable that segregations take place, which strengthen the grain boundaries.

![Fig. 9. $K_c$ of nickel–zinc ferrite as a function of grain size.](image-url)
4.3. Different phases

In this section only a combination of two phases is considered. One of the phases consists of isolated particles.

The coherence between the two phases can play a role in the fracture toughness of the material, because in the case of a limited coherence cracks can increase their surface area by partly following the interface which can lead, depending on the shape of the inclusion, to retardation of the crack front or even to crack stopping. For example in the case of fibre-reinforced materials such a mechanism can lead to very high values for fractures perpendicular to the fibre direction 26).

In this section we will further assume a good coherence to occur between the two phases.

4.3.1. The influence of different phases on $E$

The influence of $E$ on $K_c$ can directly be seen in eq. (1). The influence of a second phase on $E$ can be calculated when the volume fraction, the shape and the distribution of the second phase in the main phase are known 13). In figure 10 the calculated effective stiffness of a material consisting of WC spheres in a Co matrix is given as a function of the volume fraction of the WC. Experimental results 27) are also indicated. Generally one can say that a second phase which is stiffer than the matrix leads to a stiffer material and vice versa. In the

![Graph](image)

Fig. 10. Calculated effective stiffness $E_{cz}$ with respect to the stiffness of the matrix phase $E_m$ as a function of volume fraction $V_1$ of cemented carbide. Inserted are experimental data by Hashin 27).
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case of a random distribution of the second phase in the form of isolated particles, the influence will only be marked at high volume fractions. When the second phase is flaky or fibre-like and is oriented, a considerable effect is already to be expected at low volume fractions.  

4.3.2. The influence of a second phase with a different \( \gamma_e \) value

For such materials the same approach can be used as in sec. 4.2.1 where we already assumed two "phases" to exist: the grain-boundary phase and the grain phase. Let us assume that the crack proceeds through the weak continuous phase and that the inclusions have a higher toughness and a higher stiffness than the matrix. When the size of the inclusions is equal to or smaller than the usual roughness of the crack surface of the weak phase then no influence of the size of the second phase can be expected. When the inclusion size becomes larger than the surface roughness then one can say that \( \bar{\delta} \) in expression (4) increases so that also \( \gamma_e \) will increase. As soon as the inclusion size dominates the surface roughness, no major changes of \( \gamma_e \) with inclusion size can be expected according to expression (11). However, it can be expected that the crack will traverse the inclusions above a particular inclusion size, according to an equation similar to eq. (14). This is indeed observed by Lange in a material consisting of Al\(_2\)O\(_3\) spheres in a glass. It can also be expected that above that particular inclusion size a relatively high fracture energy is obtained, in agreement with the theoretical expectations and experimental observations of sec. 4.2.1. This has also been experimentally confirmed by Lange. When there is a distribution of inclusion sizes the fracture-surface energy will increase proportionally with the fraction of the inclusions, which are traversed by the crack (see also sec. 4.2.1). On increasing the volume fraction of the inclusions one can expect, for fracture around the inclusions, that \( \bar{\delta} \) in expression (4) remains virtually constant but that \( A_e \) and so \( \gamma_e \) increase. This expectation is experimentally confirmed for instance by Swearangan et al. When the crack propagates through the inclusions it is also to be expected that \( \gamma_e \) will increase with increasing volume fraction. For fracture fully through the inclusions, and disregarding roughness contributions, expression (5) must approximately hold.

We investigated the oriented two-phase material strontium hexaferrite. For the material described in sec. 3 the inclusions are the hexaferrite, the matrix is the Sr-rich phase. In a commercial strontium hexaferrite with a grain size (inclusion size) of about 1.0 \( \mu \)m, the crack always propagates through the weak phase both for fracture parallel and perpendicular to the texture direction. In the material prepared for this investigation, with a grain size of about 5 \( \mu \)m, fracture was for a considerable part through the inclusions. As expected, \( K_e \) increases with volume fraction of the "inclusions" or with \( n \) (fig. 11). However,
above approximately \( n = 5.6 \) a decrease is observed. This might be related to a lack of the Sr-rich phase which promotes sintering above 1200 °C by the formation of a liquid phase. The absence of the Sr-rich phase should then prevent accommodation of the thermal stresses so that during cooling down microcracking occurs resulting in a relatively low \( \gamma_c \) (or \( K_c \)) value. Fracture surfaces parallel and perpendicular to the c-axis for \( n = 5.8 \) and \( n = 4.4 \) are shown in figs 12a and b and 13a and b, respectively. Microcracks in the case of \( n = 5.8 \) can be seen clearly.

### 4.3.3. The influence of a thermal mismatch between the two phases

Though it might be superfluous to state it is emphasized here that the average thermal stresses have to be of the same order of magnitude as the average crack-tip stress in order to get a significant influence on \( K_c \). For an estimate of the average crack-tip stress one can derive with the aid of Inglis expression \(^{30}\) for the maximum stress at the tip of an elliptically shaped notch (crack) the following equation (see eq. (1)):

\[
K_c = \sigma (\pi C)^{\frac{1}{2}} = \frac{1}{2} \bar{\sigma}_t (\pi R)^{\frac{1}{2}}, \tag{15}
\]

where

- \( \bar{\sigma}_t \) is the average crack-tip stress and
- \( \bar{R} \) is the average crack-tip radius.
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Fig. 12. SEM pictures of representative fracture surfaces of strontium hexaferrite for $n = 5.8$: (a) parallel to c-axis, (b) perpendicular to c-axis.
Fig. 13. SEM pictures of representative fracture surfaces of strontium hexaferrite for $n = 4.4$: (a) parallel to $c$-axis, (b) perpendicular to $c$-axis.
For $R$ one can take as an estimate $\delta$ from expression (4). For instance for the $\text{Al}_2\text{O}_3$ investigated here $\delta$ is approximately equal to half the grain size, say $20 \mu\text{m}$ so that $\sigma_1$ becomes about $1.5 \times 10^9 \text{ N/m}^2$. In this section secondary effects which result from the thermal stresses, such as microcracking, are disregarded.

A rough estimate of the average thermal stresses for a limited difference in Young's modulus and thermal expansion coefficient between the two phases can be made in the following way. Suppose that relatively stiff and tough particles are distributed in the matrix material. One assumes a representative volume element with a parallel connection between particle and matrix material; then holds that

$$\alpha_1 \Delta T + \frac{\bar{\sigma}_1}{E_1} (1 - 2 \theta_1) = \alpha_2 \Delta T + \frac{\bar{\sigma}_2}{E_2} (1 - 2 \theta_2),$$

where

- $\alpha_1$ is the coefficient of thermal expansion of the particles,
- $\alpha_2$ is the coefficient of thermal expansion of the matrix,
- $\bar{\sigma}_1$ is the average stress in the particles,
- $\bar{\sigma}_2$ is the average stress in the matrix,
- $\theta_1$ is Poisson's ratio of the particles,
- $\theta_2$ is Poisson's ratio of the matrix,
- $E_1$ is Young's modulus of the particles,
- $E_2$ is Young's modulus of the matrix,
- $\Delta T$ is the difference between preparation and room temperature.

The forces in particle and matrix within the representative element have to balance each other thus

$$\bar{\sigma}_1 V_1 + \bar{\sigma}_2 (1 - V_1) = 0.$$

The result is

$$\bar{\sigma}_1 = - \frac{\Delta T (\alpha_2 - \alpha_1) E_1 E_2 (1 - V_1)}{(1 - V_1) E_2 (1 - 2 \theta_1) + E_1 (1 - 2 \theta_2) V_1} \quad (16)$$

and

$$\bar{\sigma}_2 = \frac{\Delta T (\alpha_2 - \alpha_1) E_1 E_2 V_1}{(1 - V_1) E_2 (1 - 2 \theta_1) + E_1 (1 - 2 \theta_2) V_1}. \quad (17)$$

After calculating these stresses one can conclude that thermal mismatch is not likely to have had any significant effect on $K_c$ in the experiments of Swearangan et al. 29).

Say we have a material consisting of inclusions of a $K_c$ value very
much higher than the $K_c$ value of the matrix and where due to cooling-down from the preparation temperature the inclusions are roughly under tensile stress. However, this thermal tensile stress is not high enough for the proceeding crack front to fracture the inclusions. The matrix is roughly under compression, so that the proceeding crack requires a higher $\bar{\sigma}$ (see eq. (15)) with respect to the matrix value in order to have crack propagation. Moreover, the crack propagates through the matrix along the particles so that surface roughening takes place, which leads to a further contribution to $K_c$. The $K_c$ value of this material can thus be expected to be considerably higher than $K_c$ of the matrix material. An example of such a material is partly or unstabilized ZrO$_2$ particles in an Al$_2$O$_3$ matrix.

In a second example the $K_c$ value of the inclusions is much lower than that of the matrix and a thermal compressive stress is present in the inclusions. Nevertheless the crack may be expected to propagate through the inclusions due to their low $K_c$ value. A further negative effect is the state of tensile stress in the matrix so that the proceeding crack needs a lower $\bar{\sigma}$ value than the matrix for propagation. The result is a material with a distinctly lower $K_c$ value than the matrix.

Similar considerations may hold for other cases.

4.3.4. Special effects

Any energy-dissipating mechanism can contribute to $K_c$. In the case of inclusions with a lower coefficient of thermal expansion than the matrix, Lange$^{34}$ showed that a critical inclusion size must exist over which cracks in the matrix can be formed. These microcracks can contribute to a $K_c$ value which is higher than the matrix value. Microcracking increases the size $\delta$ of the process zone (see expression (4)). Moreover, $\lambda_c$ may also be expected to increase. However, it is also likely that the effective stiffness, particularly in the zone of microcracking, will be relatively low. In addition the microcracking lowers the tensile prestress in the matrix.

It has been reported by Clausen et al.$^{12}$ that a.o. in the system Al$_2$O$_3$–ZrO$_2$, the positive effects, which in our view $\lambda_c$ and $\delta$ have on $K_c$ (which are clearly demonstrated by these authors) exceed the negative influence of the lowering of the effective stiffness. The critical particle size in their system was relatively low owing to the tetragonal to monoclinic phase transformation which provides an extra contribution to the tensile stress in the matrix. Moreover, a favourable influence of the thermal stresses alone also cannot be excluded with this material (see sec. 4.3.3.). Due to this small inclusion size the lowering in strength with increasing volume fraction, which generally occurs in such systems, was very restricted.
The extremely small size of the inclusions virtually rules out the occurrence of microcracking in the material developed in the cubic-tetragonal ZrO$_2$ system$^{32}$, where the tetragonal ZrO$_2$ particles change into monoclinic ones under the influence of stress. Furthermore, because of this small size, surface roughening cannot make a major contribution to $K_c$. We believe that in this case at least a contribution is made to the strength by abrasion of the sample surfaces, so that the phase transition causes a compressive layer on the surface.

5. Conclusions

(1) Many, often counteracting, parameters determine the value of $K_c$ in brittle materials, which lead to confusing results and considerations.

(2) To study the influence of separate parameters it is necessary to take great care over the preparation of the materials and over the investigation of the actual events that take place during propagation of the crack.

(3) It is shown that the concept of a process zone with a certain average size and a certain surface area per unit volume created by the crack provides at least a qualitative tool for the description of the influence of several parameters on $K_c$.

(4) The influence of porosity on $K_c$ is always negative.

(5) $K_c$ is roughly independent of grain size for pure transgranular or intergranular fracture, provided that no microcracking occurs. Microcracking takes place beyond a certain grain size and leads to a lowering of $K_c$.

(6) $K_c$ is proportional to the transgranular fraction of the fracture surface.

(7) Grain-boundary segregation either due to impurities or to non-stoichiometry can have a considerable influence on $K_c$. This phenomenon needs further investigation.

(8) The incorporation of relatively tough and stiff particles increases $K_c$ owing to an increased effective stiffness and to an increased effective crack-surface energy which is either due to surface roughening for the case of interparticle cracking or to the contribution in crack-surface energy of the particles for the case of transparticle cracking.

(9) An extra contribution to $K_c$ can be given to the material from conclusion (8) when the particles are subjected to a thermal tensile stress and the matrix to a thermal compressive stress.

(10) The optimum density of microcracks in two-phase material for an enhanced $K_c$ needs further investigation.

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