Letters

Impossibility of fragmenting small particles: brittle–ductile transition

In the comminution of particles a critical particle size is reached below which further fragmentation is impossible and at which the particles begin to flow plastically [1–3]. It is suggested in this letter that the final lower limit to the critical particle size for transition to a completely ductile behaviour is determined by conditions for crack formation or nucleation rather than by conditions for their propagation. The critical specimen size then depends only on the critical stress intensity factor, K_{IC}, and the hardness, H, of the material and the predicted specimen sizes show reasonable agreement with those observed in practice.

Situations arise in the testing of materials in which a solid may undergo a ductile–brittle or brittle–ductile transition, depending on the specimen size or volume of material and the strain rate or temperature. The transition of interest in this letter is the one associated with specimen size. Most brittle solids exhibit ductile behaviour if the specimen size or volume of material stressed is very small (e.g. hardness indentations in diamond). The inherent ductility of small particles and the critical particle size for the onset of this ductility is, however, not well understood [4–8]. As the particle size decreases the transition to ductile behaviour is obscured and is complicated by the fact that associated local yielding which occurs can nucleate cracks, so that brittle fracture may continue after plasticity. In such situations of localized elastic–plastic contacts, specimen fragmentation or beam splitting (such as those discussed by Kendall [6] and Karihaloo [7]) may still occur from plastically-induced cracks and in the absence of any long pre-existing cracks. The only condition for these cracks to be nucleated is for enough elastic energy to be available in the elastic matrix.

Once the condition of localized deformation is invoked, the problem of fragmentation, in the most general case of elastic–plastic contact, is the same as, and consists of, the *nucleation* and *propagation* of crack systems, both median and radial, around plastic indentations: a fact recognised by Rumpf and Schonert [8]. The critical specimen size for ductile–brittle (or brittle–ductile) transition may then be deduced from the threshold conditions for crack nucleation under plastic indentations as follows. No assumptions are made about the specimen size or the initial crack length; the only requirement is for the load to be localized enough to induce a limited amount of plastic deformation at the contact.

At a certain stage of the indentation various crack systems are nucleated by plastic processes in the deformed zone; interaction of dislocations and shear flow-lines in crystalline and some amorphous materials, respectively, are responsible for crack nucleation [9, 10].

The micromechanics of the nucleation process have been considered recently by Lawn and Evans [11] and Hagan [10]. The analysis of Hagan shows that the threshold load, P_c, to nucleate a crack, and the size of the crack, c, are given respectively by

$$P_c \approx 880\,(K_{IC}/H)^3 K_{IC} \qquad (1)$$

and

$$c \approx 30\,(K_{IC}/H)^2 . \qquad (2)$$

Because the analysis treats the crack nucleation phase of the fragmentation, both equations depend only on the critical stress intensity factor, K_{IC}, and the hardness, H.

The suggestion of this letter is that the largest flaw-size predicted by Equation 2 essentially determines the critical specimen size for the ductile–brittle (or brittle–ductile) transition in comminution. This is, however, only true at or near the threshold specimen sizes at which the transition is to be expected. It is argued that if the crack nucleated by the plastic processes is equal to, or larger than, the specimen size then splitting occurs. If, however, the plastically-induced crack is smaller than the specimen size then the specimen cannot be fractured because the maximum available energy is insufficient for fracture: i.e. not enough elastic energy can be stored in the specimen to cause splitting. Therefore specimens below this specimen size will not fracture but will continue to flow plastically.

The critical specimen sizes of 0.6 μm and 0.5 μm for MgO and glass, respectively, predicted by

0022–2461/81/102909–03$02.30/0

TABLE I Comparison of the brittle–ductile transition models for different materials

Materials	Critical size for brittle–ductile transition	
	Present model (μm)	Kendall's model (μm)
Polystyrene	2800	4500
MgO	0.6	1.8
KCl	295	944
NaCl	32	102
SiO_2	1.1	3.6
Al_2O_3	3.3	10.6
Soda-lime glass	0.5	1.8

the above analysis are in excellent agreement with those observed in practice [12. 13]. Some other values are compared with those given by the model of Kendall in Table I.

It must be emphasised that the above argument is only valid at or near the threshold condition for crack nucleation. If such critical flaw sizes (Equation 2) are, however, nucleated in larger specimens, fragmentation by the applied load is still possible. The problem then becomes one of crack propagation instead of nucleation. The expression given by Lawn and Fuller [4] for the propagation of a fully-developed indentation (median) crack of size c may be re-expressed as

$$K_{IC} = \chi P/(a + c)^{3/2}, \quad (3)$$

where χ is a constant and a is the radius of the plastic contact; c is measured from the edge of the plastic contact. For values of c much smaller than a, Equation 3 reduces to $K_{IC} = \chi P/a^{3/2}$. On making the substition of $a = \alpha(K_{IC}/H)^2$ from [10], Equation 3 becomes

$$P = \beta(K_{IC}/H)^3 K_{IC}, \quad (4)$$

where β is a constant. It is interesting to note that Equation 3 reduces to the threshold conditions for the ductile–brittle transition as the propagation stresses become dominated by the deformation stresses.

Now let us consider the two models by Kendall [6] and Karihaloo [7] on comminution. Karihaloo has suggested that the incorrect predictions about the critical load, P_c, to propagate a crack and the specimen size, d_{crit}, for the brittle–ductile transition given by the Kendall analysis, being independent of crack length may be resolved by incorporating tractions at the specimen–platen interface in the analysis. However, this does not, for the following reasons, resolve the apparent discrepancies between their models. Firstly it may be shown [15] that the Kendall analysis rather fortuitously correctly predicts the threshold conditions for ductile–brittle transitions. This is because, for crack nucleation, only the shear stresses are required so that traction at the specimen–platen interface normal to the prospective crack path play little or no part in the actual nucleation process [16, 17] and may be ignored. Thus Kendall's predictions are correct only if they are interpreted as representing threshold nucleation conditions instead of crack propagation conditions, as is assumed in his analysis; in this former case the critical parameters should be independent of crack length.

These threshold nucleation conditions have to be extended, however, to predict the load to propagate the crack once it has been nucleated. This, in reality, is what the modification of Karihaloo does, and the propagation condition will be expected to depend on the crack length for the specimen geometry and loading conditions analysed. It is worth pointing out that the origin and magnitude of the adjustable constraint factor, α, in the analysis of Karihaloo are uncertain. It appears, however, that this constraint factor is probably derived from, and is comparable with, the coefficient of friction and it ought to be larger than 0.005 since the average value of the coefficient of friction even for diamond on diamond is 0.1 [18].

In conclusion, it must be appreciated that the critical specimen sizes for the brittle–ductile transition in the above analysis are only approximate. They are, however, useful for comparison and also as a guide to the limiting sizes to be expected in practical situations. The analysis in this letter removes the limiting features of large inherent flaw sizes in the Kendall–Karihaloo models and provides a mechanism for producing large flaws for fragmentation in the general case of elastic–plastic contact.

Acknowledgements

I am grateful to Dr J. E. Field, Dr E. Yoffe, Dr M. M. Chaudhri and Mr S. van der Zwaag for their useful comments.

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Received 13 January
and accepted 26 February 1981

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Lattice thermal expansion of cupric thiogallate

The I–III–VI_2 ternary compounds which crystallize in the uniaxial calcopyrite structure are potentially interesting as non-linear optical materials as well as semiconductors [1, 2]. The compound cupric thiogallate, Cu–Ga–S_2, a member of this class of materials, is probably the most interesting because of its potential use for many technological applications [2, 3]. Under a programme of studies on some chalcopyrite ternary semiconducting compounds by X-ray diffraction analysis the authors have previously reported the lattice thermal expansion of a number of compounds within this class of materials [4–8]. The present note gives the results of a similar X-ray study of cupric thiogallate. While the work was in progress, Yamamoto *et al.* [9] reported the temperature variation of lattice parameters and the average coefficients of thermal expansion of Cu–Ga–S_2 using X-ray diffraction analysis. In their study they made use of a Rigaku high-temperature powder camera. However, in their work [9] no details of the accuracy of their results was given and, further, the accuracies with which the lattice parameters and the temperatures were determined were poor compared to our earlier studies [4–8]. Hence, it was thought worthwhile to proceed and to determine the accurate lattice parameters and the coefficients of thermal expansion for Cu–Ga–S_2 at various temperatures, as a part of the programme.

The Cu–Ga–S_2 sample used in the present study was kindly supplied by Dr B. Tell of Bell Laboratories. The details of the growth method

TABLE I Lattice parameters of Cu–Ga–S_2 at room temperature

Lattice parameter		Reference number
a (nm)	c (nm)	
0.5349	1.047	[13]
0.5351	1.0484	[10]
0.534 741 ± 0.000 007	1.047 429 ± 0.000 006	[14]
0.5347	1.0474	[15]
0.5359	1.049	[3]
0.534 74 ± 0.000 01	1.048 25 ± 0.000 02	Present study

