

### Comments on "Thermal stresses in rubber-modified glassy polymers"

In a recent paper [1], Pavan and Ricco have considered the distribution and magnitude of thermal stresses developed around a rubber particle embedded in a glassy matrix during cooling of the composite material. They have shown that as glassy material is transferred in the form of occlusions into the interior of the rubber particle, the level of thermal stress decreases at, e.g. the particle-matrix interface. A uniform state of triaxial hydrostatic tension is maintained, as is the case for a simple rubber particle. By referring to the work of Schmitt, which indicates that triaxial tension in rubber particles is essential for toughening, the authors imply that the tension-reducing effect of introducing glassy occlusions ("subinclusions") into rubber particles is detrimental to toughness. They conclude that "for a given rubber content, the level of thermal stresses decreases as more glassy polymer is transferred from the matrix to the dispersed phase". It must be pointed out, however, that for a "given amount" of rubber, the introduction of glassy occlusions is necessarily accompanied by an increase in volume fraction of dispersed phase. Pavan's analysis deals with a model system in which rubber is clearly not conserved, and it is not valid for him to extrapolate

the results of such an analysis to a situation where rubber is by definition conserved during the introduction of glassy occlusions. It should be pointed out that the insertion of glassy occlusions under conditions where rubber is conserved means that in unit volume of composite material there is an increase in the number of thermally stressed "matrix shells" one of which is associated with each discrete phase particle. The amount of matrix stress necessary for effecting any level of toughening is not quantified in the work of Schmitt [2] and Beck *et al.* [3] and until such information appears, it would seem reasonable that the implications of Pavan and Ricco's analysis, and the reservations which attach to their conclusions, be set out.

### References

1. A. PAVAN and T. RICCO, *J. Mater. Sci.* **11** (1976) 1180.
2. J. A. SCHMITT, *J. Appl. Polymer Sci.* **12** (1968) 533; *J. Polymer Sci.* **C30** (1970) 437.
3. R. H. BECK, S. GRATZ, S. NEWMANN and K. C. RUSCH, *Polymer Letters* **6** (1968) 707.

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### Reply to "Comments on 'Thermal stresses in rubber-modified glassy polymers'"

The comments on our paper seem to have originated from two unjustified assumptions.

(1) In the commentator's reasoning it is tacitly assumed that the particle size is invariant. This is completely unjustified. The introduction of glassy occlusions in *preformed* rubber particles (a situation of practical interest whenever the dispersed phase is formed in emulsion in two stages), for instance, affects the size of the particles, while it leaves the number of particles per unit volume of composite material unaffected. Thus, while it is perfectly true that "for a given amount of rubber, the introduction of glassy occlusions is necessarily accompanied by an increase in volume fraction of dispersed phase",

the next two statements are not consequential, and are not true. It is not true that in our model, "rubber is (clearly) not conserved", because it is not true that "the insertion of glassy occlusions... necessarily "means that... there is an increase in the number...". In our model, rubber is clearly conserved during the introduction of a glassy domain of volume  $\frac{4}{3}\pi b^3$  into a rubber particle, if the outer radius of the particle,  $a$ , is thought of as varying with  $b$  so that the difference  $\frac{4}{3}\pi(a^3 - b^3)$  is kept constant. The increase in volume fraction of dispersed phase in the actual material will reflect the increase in size of each dispersed particle.

(2) Our quoting of papers dealing with thermal stresses in relation to toughness, must not be taken as implying that we share (or do not share) their authors' view on the mechanism of toughening

and the role of thermal stresses. Our paper was declaredly aimed at taking cognizance of the general presence of glassy occlusions in rubber particles and at assessing its effect on thermal stresses. Accordingly, the only conclusion that we drew from our analysis related to the distribution and magnitude of these stresses. Any possible implication as to toughness depends on which mechanism is believed to be responsible for the toughening, and we leave to others to draw their own conclusions depending on their own beliefs.

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