Authors' Reply to the Remarks made by Dr. S. Paipetis Concerning Their Paper Entitled: "Glass Transition Behavior of Particle Composites Modelled on the Concept of Interphase" [J. Appl. Polym. Sci., 27(8), 3019–3025 (1982)]

It will be clarifying to introduce this reply by the observation that Dr. Paipetis came to our laboratory some 15 years ago, worked for more than 8 years with me and my associates on glass transition phenomena of composites, published several papers with us, and continued such studies after he left to take over another position.

During the same period—after 1978—additional experiments on the same subject were continued at our laboratory, which Dr. Paipetis used in his article without, in my opinion, giving adequate credit to our contributions.

In the above-mentioned papers our key approach was to describe and to interpret the behavior of particular composites in the glass-transition region by the existence of an *interphase* between particles and matrix. It goes without saying that this concept has been treated many times very systematically from our research group. Dr. Paipetis, however, who was well aware of all this work, misinterprets our aim by pretending that we did not give a sufficiently detailed description of this interlayer. Hence, it seems that he did not understand the main scope of our publication, which was to emphasize the very decisive role of the interphase, by introducing it, even through the simplified term $E_i v_i$ into the equation for the effective modulus of the composite material.

Dr. Paipetis, however, takes the position that—for the explanation of an increased value of T_g of the composite—it suffices to make recourse only to a more rigorous mathematical expression for the modulus. All data in our papers indicate that the increase in T_g is due to the existence of an intermediate layer between filler and matrix, which is developed during the preparation of the composite material. In this way our analysis, based on the influence of this interphase, explains not only the possibility of an increase but also that of a decrease of T_g of the composite.

It is also worth noting that Dr. Paipetis, in order to prove his suggestions, recommends as a rigorous expression for the particulate composites the Hashin equation

$$E_a = v_m E_m + v_p E_p + \frac{4_1 v_m v_p (\nu_p - \nu_m)^2}{v_m/k_p + v_p/k_m + 1/c_m}$$

which obviously refers to fiber composites, as first used by Hashin. Then, he asks for a better expression and approximation of this incorrect formula!!

Apart from this fact, even if we accept the validity of this expression also for particulate materials, we have to make the following remarks on the use of this equation:

a. The third term

$$\frac{4v_m v_p (\nu_p - \nu_m)^2}{v_m/k_p + v_p/k_m + 1/c_m}$$

refers to contributions coming from the differences in the Poisson ratios between fiber and matrix $(\nu_p - \nu_m)^2$. These contributions are not larger than 1% and so the contribution of this third term could not explain any substantial increase of the glass-transition temperature.

b. This term has nothing to do with the interphase. It improves only slightly the results derived from the simple two-term formula. Dr. Paipetis in his criticism, however, does not have a clear view, and his position at this point is dubious. Does he believe that the existence of such an intermediate phase is irrelevant for the detailed description of the model, or not?

Journal of Applied Polymer Science, Vol. 31, 2845–2846 (1986)
 CCC 0021-8995/86/082845-02$04.00

We think that he appeared to be a little bit confused when he criticized us for not using an extensive and detailed description of this boundary layer, a fact which is unimportant for our study, if we consider the average values of the moduli and Poisson's ratios of this variable with the thickness layer.

We believe that our thesis is obvious, that for our analysis the complete description of the interphase is irrelevant, and that it does not influence the results. The interesting point is that, by accepting its existence with its average characteristic values as a physical reality, we may improve its influence on the phenomenon of the displacement of the position of the glass-transition temperature in the temperature scale.

Referring now to the points which Dr. Paipetis accuses as "verbatim reproduction" of his publication, we want to say:

i. The way of defining the glass-transition frequency ω_g for harmonic loading, through the loss factor $\eta(\omega)$, by the equation

$$\left.\frac{\partial \eta(\omega)}{\partial \omega}\right|_{\omega = \omega_g}$$

is widely used, has been established long before our publications, and is included in many textbooks; it does not represent a contribution of Dr. Paipetis.

ii. The parts of our article which he calls "verbatim reproductions" of his article are stereotype expressions which have been used many times in the past and are contained in older articles written from our group, eventually even in collaboration with Dr. Paipetis.

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Received November 28, 1984
Accepted January 17, 1986