

Impact Strength of Thick-Interlayer Composites

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Synopsis

The impact strength of a thicker interlayer fiber filled composite material is evaluated as a function of both interlayer thickness and glass transition temperatures (T_g). The results confirm that for maximum strength, an optimum interlayer thickness is required. It is shown that this result is a function of the interlayer's T_g . Comparison with studies on other rubber reinforced systems are noted.

INTRODUCTION

Interest in improving the mechanical properties of composite materials is increasing rapidly. Normally rigid fillers are added to polymeric materials so as to increase modulus, strength, and high-temperature performance. The effect of fillers on composite performance can be further increased by the modification of the boundary between the filler and matrix material. Thus it has become common practice to use coupling agents to improve the adhesion between these two phases.¹ Several theories² have been proposed to explain the function of coupling agents, but complete elucidation of the "bonding" mechanism has not been achieved. Recently a technique for producing thick-interlayer composite materials was described.³ Through the use of electrostatic forces, substrates (used as filler systems) were encapsulated with uniform polymer layers. Composites containing filler materials coated with a thick interlayer resulted in improvements in mechanical properties of the composite.³ The impact resistance values of these materials will be discussed here. As will be noted, the impact strength is a function both interlayer thickness and glass transition temperature (T_g).

EXPERIMENTAL

The method used for the placement of polymer coatings onto a glass surface has been described previously.^{3,4} Alternate layers of negatively and positively charged colloidal particles can be deposited from dilute sols, thus forming a "film" on the substrate surface. The method described here is based on the fact that the surface of glass is charged (usually anionically) and that oppositely charged particles can be attracted to this surface. A monolayer of particles is formed because further particle deposition is inhibited by electrostatic repulsion. Multiple layers can be formed by placing between each layer of like charges a layer of oppositely charged particles.

Iler's method⁴ for the placement of colloidal particles onto a glass surface, as noted previously,³ is a relatively simple matter. For latex particles (normally a negatively charged species) to be attracted to a sized fiberglass material, the neutrally charged coupling agent has to be removed so as to expose the glass

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surface. This clean surface is then placed into a 0.5% aqueous solution ($\text{pH} \approx 7$) of a colloidal alumina ("Baymal," E. I. DuPont de Nemours & Co., trademark for colloidal alumina), which "reverses" the anionic to a cationic charge. The glass is subsequently removed from this solution, rinsed under distilled water, and air dried. This positively charged glass is then dipped into a 5% latex solution, the pH of which is adjusted to 2. Again the glass is rinsed under distilled water and air dried. In all cases, the solutions were quite dilute ($\leq 5\%$), and the entire coating process took place at 25°C . After air drying, the latex-coated fibers were heated in an air-circulating oven for 20 min at 155°C . The properties of the latex materials used in this study are given in Table I. As stated previously,³ multiple latex layers can be deposited by alternating the layers with oppositely charged species (colloidal alumina).

It should be noted that the surface charge on metal oxides and silicates can be easily changed. The surface of these materials in equilibrium with water are made up of hydrated hydroxyl groups. A zeta potential can be imposed, which is determined by the OH^- and H^+ concentration in the adjacent aqueous solution. The pH at which this potential is zero is defined as the isoelectric point of the surface (IEPS). Bascom⁵ noted that the IEPS represents the H^+ or OH^- concentration necessary to suppress ionization of the surface OH groups. At a pH (in an aqueous medium) lower than the IEPS, the surface will be cationic; while at a higher pH, the surface will be anionic. The IEPS of the oxide surface of Al^{3+} and Si^{4+} is a 9.1 and 2.2, respectively.³ Thus by modifying the pH of the coating solutions, one can change the particle-attracting ability of the glass surface. The significance of the pH used in both coating solutions previously described should be apparent.

A cationic-sized fiberglass (supplied by Owens Corning) was used here so as to eliminate the need to expose the glass surface. A negatively charged latex could be directly placed onto the glass without the initial layer of colloidal alumina. The procedure for placing a multilayer latex coating on the surface follows the recipe previously described. Burning off of the coupling agent causes a deterioration in the properties of the composite material, presumably due to scratching of the glass fibers by other fibers.

Finally, the latex-coated (monolayer or multilayer) fiberglass is placed into an epoxy matrix (Shell's Epon 815). This mixture is degassed and cured as previously described.³ In all cases, the epoxy resin system (resin and curing agent) penetrated the interlayer to an approximately equal extent. About 20% by weight Epon 815 resin diffused into the interlayer region.

The three lower T_g acrylate latices deposited onto the surface of glass as individual particles but on drying would coalesce into a film. The higher T_g polyvinyl chloride (PVC), however, retained its particle structure after complete

TABLE I
Physical Properties of the Latex Used in Study

Latex composition	Average particle diameter, Å	T_g , °C
Poly(vinyl chloride)	900	+80
Poly(methyl acrylate)	1100	+10
Poly(ethyl acrylate)	1000	-14
Poly(butyl acrylate)	1000	-56

drying. Thus to insure complete coverage of the surface by PVC, these fibers were placed into a vacuum oven at a temperature of 95°C. This temperature was sufficient to allow the individual PVC particles to form a uniform film on the glass.

The notched Izod measurements follow the procedure outlined in ASTM-D256.

RESULTS AND DISCUSSION

As shown in a previous publication,³ the impact strength of thick-interlayer composites is a function of the thickness of the interlayer. In those materials studied, the latex-coated fiberglass composites had greater impact resistance than an uncoated fiberglass material. As we shall see, the impact strength is both a function of interlayer thickness and interlayer T_g . With a T_g greater than room temperature, the impact resistance is less than the completely uncoated glass. This trend is brought out in Figure 1.

Figure 1 shows the notched Izod values of four interlayer epoxy composites as a function of both T_g and interlayer thickness. An increase in T_g causes a decrease in impact strength at each level of interlayer thickness. The best impact values are obtained with a T_g of -56°C. At each T_g level one also observes that the impact values increase up to an interlayer thickness of approximately 2000 Å. A decrease is found with thickness greater than 2000 Å. The impact values, in all cases, are also observed to intersect the ordinate at the unsized and uncoated fiberglass material. Epoxy composites containing interlayers possessing a T_g greater than room temperature causes no appreciable change in impact resistance. This interlayer apparently does not allow for any new energy-absorbing mechanisms to appear as in the other three lower T_g materials.

As stated previously, a cationic size is used to attract the initial layer of latex particles. The thickness of this coating³ is about 275 Å. The impact strength of an epoxy composite containing this cationic glass is found to be 1.62 ft lb.³ Thus it seems that this cationic coating has little effect on the impact strength whenever it is coated over by an interlayer. The properties of these interlayer composites is essentially controlled by the thickness and T_g of the latex interlayer. In previous studies⁶ on rubber-toughened polymeric materials, the T_g

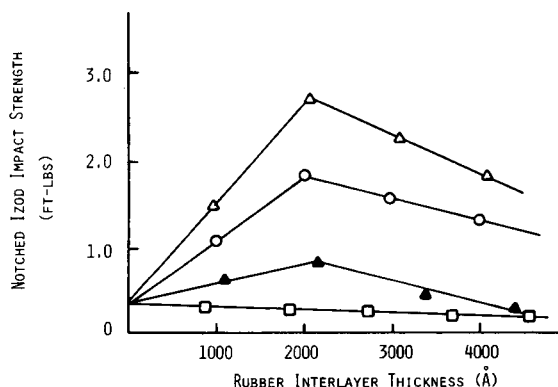


Fig. 1. Impact strength of a 30 v/o randomly oriented fiberglass composite possessing an interlayer structure of varying thickness at the following T_g (°C): -56 (Δ), 14 (○), 10 (▲), and 80 (□).

of the elastomeric phase had a significant effect on impact properties. This phase should have a low T_g so as to remain elastomeric at the specific loading rate and testing temperature. The elastomeric phase must be able to relax under the very high loading rates found in an impact test. Through this mechanism the impact energy is dissipated. These same considerations apply to the interlayer composites produced in this work.

Up to this point, little attention has been given to the rather abrupt change in impact strength in the interlayer thickness region of 2000 Å (0.2 μm). There are indications in the literature⁷⁻¹⁰ on rubber-reinforced polystyrene that an optimum particle size is needed to ensure maximum impact strength. These particles should have diameters in the range of 2-5 μm.⁸ The size of the rubber particles does vary depending on polymerization conditions. In ABS the maximum rubber latex particle size that ensures high impact is 18,000 Å.¹⁰ The results shown in Figure 1 parallel these studies. The maximum in toughness (as observed in Fig. 1) is due to the effect of interlayer thickness on the extent of craze initiation and craze termination within the epoxy matrix. When a balance is achieved between these two factors (dependent on a specific particle size), the strength is at a maximum value. Furthermore, recent calculations by Sudduth¹¹ have shown that a critical particle size exists below which impact strength increases with a particle size increase and above which strength decreases with an increase in particle size. Variations in graft thickness change the value of the critical particle size.

CONCLUSIONS

The major conclusion that can be drawn from this work is that optimum mechanical properties (specifically toughness) are obtained when there is a uniform rubbery interface region located between glass fibers and a rigid epoxy in a composite material. The properties are dependent on both the interlayer T_g and thickness.

Many other factors could be controlled in order to further modify thick-interlayer composite properties. These include modifications in the matrix molecular structure, matrix crosslink density or crystallinity, filler type and size, interlayer molecular structure, interlayer crosslink density, diffusion rate of matrix into interlayer, and chemical reactions between coupling agent and interlayer. These factors are, of course, not completely independent of each other. Further experimentation is, of course, necessary so that their effect on the composite's mechanical properties can be measured.

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