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Measurements of Ballistic Impact Response of Novel Coextruded PC/PMMA Multilayered-Composites

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ABSTRACT: Ballistic impact behavior of coextruded composites consisting of alternate layers of ductile polycarbonate (PC) and brittle poly(methyl methacrylate) (PMMA) were investigated. These PC/PMMA composites were 256, 1024, 2048 and 4096 layers; each consisted of various component compositions and their corresponding layer thickness was on the order of micron or sub-micron scale. Individual layer thickness of PMMA was determined to be critical for the impact response of these ductile/brittle hybrids. A brittle mode of failure was encountered as the thickness of PMMA layers reached 0.5 μm or higher, regardless of their composition or layer configuration. Damage zone size increased significantly as the PMMA layer thickness reduced to approximately 0.36 μm; a mixed mode of failure resulted in these PC/PMMA composites. With the PMMA layer thickness further reduced to approximately 0.14 μm, ductile deformation, which was predominant in the pure PC, occurred in these coextruded composites. Ballistic impact energy data showed that the extent of energy absorption or dissipation increased with an increase in either the PC content or the total number of layers. These results further revealed that the individual layer thickness of PMMA was the critical material parameter, and the determined impact energy values were consistently higher for the composites with PMMA layer thickness being 0.14 μm or thinner. Adhesion between the layers appeared to be very good since delamination did not occur whether these PC/PMMA composites failed in a brittle or a ductile mode.

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KEY WORDS: coextrusion multilayering, polycarbonate (PC), poly(methyl methacrylate) (PMMA), ballistic impact, mode of failure, layer thickness.

INTRODUCTION

POLYCARBONATE (PC) IS widely used in many applications that require transparency as well as high impact resistance, including windshields, canopies, vision blocks, face shields, goggles and lenses. PC has excellent ballistic impact strength; however, it has poor chemical resistance and can scratch easily. Blending with other transparent engineering thermoplastic polymers or surface-modification with a transparent hard coating are two common approaches for remedying PC from its susceptibility to various service hazards.

Miscibility of the PC blends depends strongly upon the method of preparation. Single-phase morphology was observed when PC blends were prepared via solution blending; however, selection of proper solvents was critical to achieve a miscible blend [1–9]. In some cases, solvent induced crystallization of PC occurred which caused a loss of optical clarity. Immiscible blends usually resulted from melt processing, which displayed phase separation and yielded either a translucent or an opaque material [1,2,7–10]. Melt-blended PC with a copolyester, on the other hand, was shown completely miscible; these homogeneous mixtures exhibited optical clarity and enhanced chemical resistance, and their ballistic impact performance, however, was significantly reduced [11].

Hard coatings are needed for most commercial PC's; however, a hard coating capable of providing resistance to chemical resistance or abrasion hazard may sometimes cause an adverse effect on the impact performance of the coated PC. Optimization of coating thickness as well as coating chemistry is critical for the multi-functionalities requirements; this is particularly true for the coated systems that consist of a thin hard coating and a ductile substrate. The unique material characteristics of a ductile/brittle hybrid provided the motivation of this research for the development of model polymer-polymer composites with individual layer thickness on the order of micron or sub-micron scale [12]. A microlayer coextrusion technique [13–15], which allows for coextruded samples to be multiplied into hundreds or even thousands of layers via a set of unique die elements, was utilized for producing microlayers consisting of alternate layers of pure PC and pure poly(methyl methacrylate) (PMMA).

Extensive studies on mechanical properties and micro-deformation mechanisms were reported on coextruded PC/styrene-acrylonitrile (SAN) composites [13,16–18]. Results of the tensile and impact tests showed that a brittle-ductile transition occurred in the coextruded PC-SAN composites as the PC content increased; ductility increased as the total number of layers increased. Kohlman also observed that the fraction of brittle failure upon ballistic impact decreased as the PC content or the total number of layers increased in either PC/SAN or PC/copolyester composites [19].

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PMMA is selected for its optical, abrasion, and chemical barrier properties; however, PMMA is brittle upon impact. PC and PMMA are not miscible, and a single-phase morphology was only observed in the PC/PMMA blends prepared by solvent casting with tetrahydrofuron [1,2,7]. However, these mixtures became phase separated upon heating just above their glass transition temperature. Technical challenges for successful fabrication of coextruded PC/PMMA composites reside upon the ability to enhance the overall durability without the expense of the impact strength of PC.

This work examines the ballistic impact response of PC/PMMA microlayers which consist of different number of layers and various component compositions. The role of individual layer thickness on the mode of failure as well as on the overall ballistic strength is determined.

EXPERIMENTAL

Materials and Multilayer Processing

The PC was Calibre™ 300-15 from the Dow Chemical Company (Midland, MI) with a molecular weight of 27,000 g/mol, and the PMMA was V826-100 made by the Ato-Haas Company (Chicago, IL) with a molecular weight of 120,000 g/mol. PC/PMMA composites were fabricated using a unique coextrusion multiplying process available at Case Western Reserve University [13–15]. During the microlayer coextrusion, PC and PMMA melts were fed from two separate single screw extruders through a coextrusion block as parallel layers, followed by a series of die elements. Each die element doubles the number of layers as shown in Figure 1; it first splits the viscoelastic melts vertically (stage B), then compresses and spreads each half horizontally (stage C), and finally recombines them (stage D). Therefore, a coextruded multilayer sheet consisting of alternating PC and PMMA with $2^{(n+1)}$ layers can be fabricated using a set of n die ele-

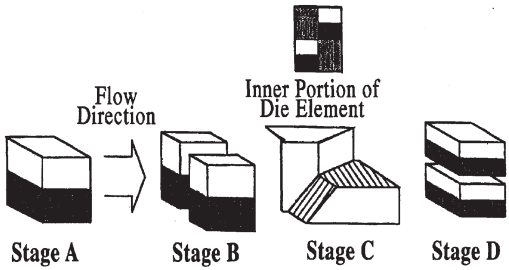

Figure 1. *Schematic of polymer melts passing through the first die element in the coextrusion multilayering process.*

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ments. The feed ratios of the two pure components into the two separate extruders pre-determined their composition. The average individual layer thickness was calculated based on the measured overall sheet thickness, the number of layers, and the pre-determined composition. The higher the volume content of one polymer, the larger the individual layer thickness of that respective component in the coextruded composites. In this study, two sets of PC/PMMA composites have been prepared. The number of layers associated with the first group are 256, 1024, and 2048, and the volume ratios between PC and PMMA vary from 20/80 to 80/20. The second set of PC/PMMA multilayers contain higher volume ratios of PC/PMMA, 80/20, 90/10 and 95/5, with the total number of layers being 2048 and 4096. The average sheet thickness of the first set is 1.14 mm, and that of the second set is 1.36 mm.

Ballistic Impact Test

Measurements of ballistic impact were carried out using a helium gas gun apparatus as shown in Figure 2. A PC/PMMA composite specimen of approximately 4 in. by 4 in. was mounted between two aluminum plates that had a 2-inch-diameter opening. These sample holder plates were firmly tightened, then placed and clamped in the center of an impact test apparatus. The projectile used for all ballistic testing was a 17 grain (1.1 gram weight), .22 caliber (0.22 inch diameter) fragment simulating projectile (FSP), and the projectile was propelled by using helium gas. Four light screens were used as triggers for timers to record the time-of-flight of the projectile to determine the velocity of the projectile before and after impact. Target is placed at midpoint between the second and third screens, and D_1, D_2, and

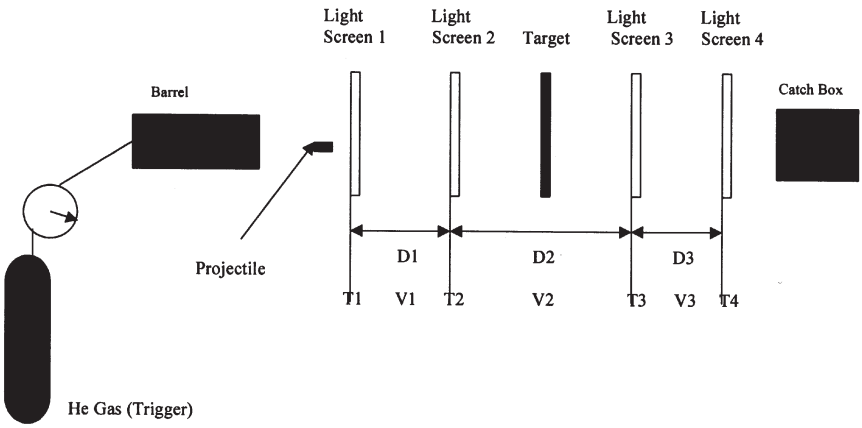

Figure 2. *Schematic of ballistic impact test setup.*

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D_3 denote distance between screens 1 and 2, 2 and 3, and 3 and 4. V_1 is the velocity measured between the first and second light screens, V_2 between second and third light screens, and V_3 between the third and fourth screens.

Striking velocity (V_s) at the front surface of the target was calculated from V_1 following an air drag correction; similarly, residual velocity (V_r) at the rear surface of the target was calculated from V_3. Air drag correction was made from the experimentally obtained third-degree polynomial fit between V_1 and V_2 for V_s and between V_2 and V_3 for V_r. Therefore, V_s and V_r were calculated as follows:

$$V_s = -3.34\times10^{-9}V_1^3 - 1.25\times10^{-6}V_1^2 + 0.983V_1 \quad (1)$$

$$V_r = -8.75\times10^{-8}V_3^3 + 6.81\times10^{-5}V_3^2 + 0.983V_3 \quad (2)$$

The range of impact velocity used throughout this work was between 120 to 160 m/s. Ballistic impact energy, which is a measure of kinetic energy absorbed or dissipated by the sample upon impact, is calculated following Equation (3) with an assumption that the mass (M) of the projectile is constant during its penetration into the target.

$$\text{Ballistic Impact Energy} = 1/2M(V_s^2 - V_r^2) \quad (3)$$

where V_s and V_r are the striking and residual velocities, respectively.

Photographs of ballistic impact test specimens were taken using a Kodak DCS 420 digital camera. Post processing of photographs was done using PhotoShop software to ensure resolution.

RESULTS AND DISCUSSION

Failure Mechanisms upon Ballistic Impact

Figure 3 compares the ballistic impact response of pure PC and coextruded 256-layer PC/PMMA composites. Ductile deformation occurred in the pure PC; however, all the composites with 35, 50, 65, and 80 vol.% exhibited brittle failure. Fracture surface of the pure PC revealed a circular pattern, which was not observed in any of the 256-layer composites. This ring characteristic was a result of the indent of the sample holder which had a circular opening upon the test specimen, indicating the extent of stress wave propagation during the ballistic impact.

Incorporation of 20 vol.% of PMMA in the 256-layer 80/20 PC/PMMA composite significantly reduced the impact strength of PC, indicating that the presence of a brittle PMMA phase strongly affects the ductility of PC. Therefore, multilayers with thinner layer thickness were fabricated through an increase of the

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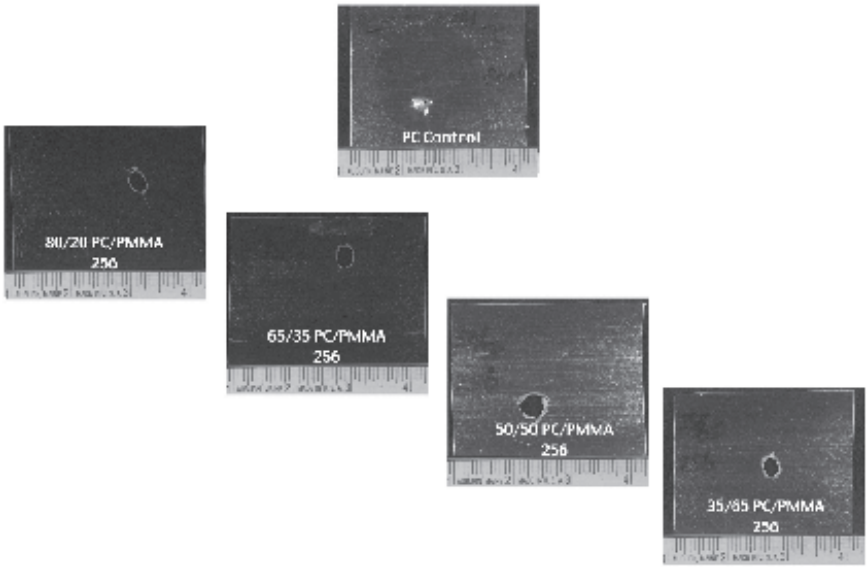

Figure 3. *Photographs of ballistic impact response of coextruded 256-layer PC/PMMA composites.*

total number of layers (i.e., 1024 and 2048 layers) to examine the compatibility of a ductile PC with a brittle PMMA. These coextruded composites consisted of 20, 35, 50, 65, and 80 vol.% of PMMA. Figure 4 shows the impact results of the 1024-layer composites; ballistic performance was apparently better in the 80/20 PC/PMMA composite in which deformation involving a larger damage area was observed on the fracture surface. Brittle failure, however, occurred in all the other 1024-layer composites with less than 80 vol.% of PC.

A significantly improved ballistic performance was evident in the 2048-layer 80/20 PC/PMMA composite; a ring pattern was present indicating that the extent of stress wave dissipation in the radial direction was significant during the ballistic penetration. This enhanced damage tolerance to the impact was not observed in the other 2048-layer composites that had less than 80 vol.% of PC. Figure 5 compares the ballistic response of 256-, 1024-, and 2048-layer 80/20 PC/PMMA composites; the more the total number of layers in these multilayers the better their ballistic impact performance. This clearly demonstrated that the layer thickness of PMMA is critical for determining the impact strength of these coextruded PC/PMMA composites.

Table 1 lists the average thickness of these coextruded composites, in which the calculated individual layer thickness of PC and PMMA is also included. The overall sheet thickness of the 256- and 1024-layer composites (about 1 to 1.3 mm) was

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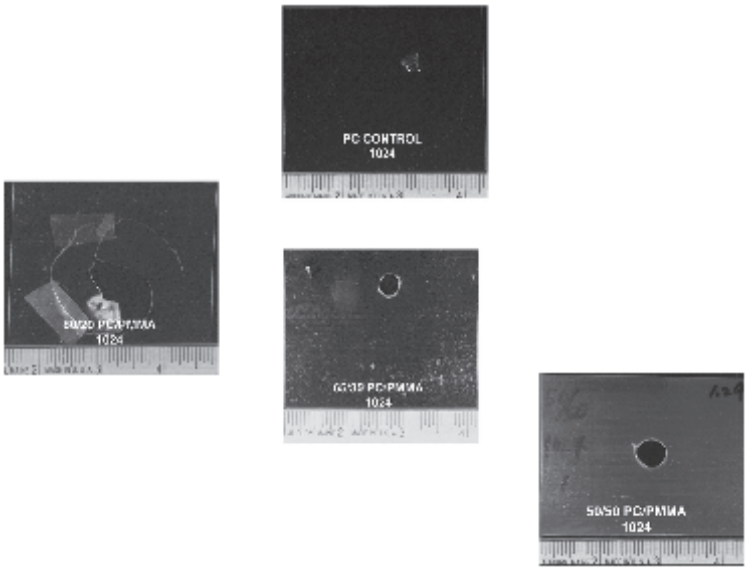

Figure 4. *Photographs of ballistic impact response of coextruded 1024-layer PC/PMMA composites.*

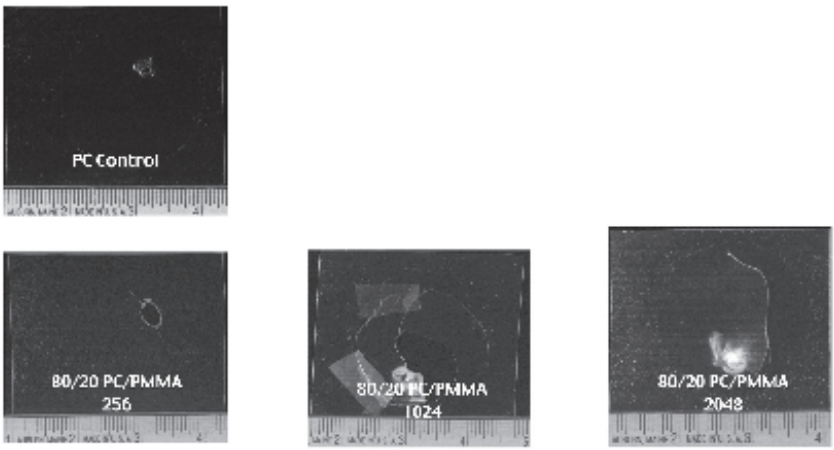

Figure 5. *Comparison of ballistic impact response of coextruded 256-, 1024-, and 2048-layer PC/PMMA composites with 80 vol.% of PC.*

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Table 1. Average sheet thickness of 256-, 1024-, and 2048-layer PC/PMMA multilayers and the calculated individual layer thickness of PC and PMMA for the corresponding composites.

	256-Layer		1024-Layer		2048-Layer	
PC/PMMA	Sheet Thickness (mm)	Layer Thickness (μm)	Sheet Thickness (mm)	Layer Thickness (μm)	Sheet Thickness (mm)	Layer Thickness (μm)
80/20	0.89	5.6/1.4	0.9	1.4/0.4	1.31	1.0/0.3
65/35	0.93	4.7/2.5	0.9	1.1/0.6	1.4	0.9/0.5
50/50	1.25	4.9/4.9	1.29	1.3/1.3	1.45	0.7/0.7
35/65	n.a.	n.a.	n.a.	n.a.	1.29	0.4/0.8
20/80	n.a.	n.a.	n.a.	n.a.	1.35	0.3/1.1

slightly thinner than that of the 2048-layer multilayers (about 1.3 to 1.5 mm). The individual layer thickness of one polymer decreases with a decrease in the volume content of the respective component. As the PMMA layer thickness reduced to about 0.4 μm or thinner, the damage zone size associated with the impact increased which was evidenced in the 1024- and 2048-layer 80/20 PC/PMMA composites. The latter displayed better impact strength in which the corresponding individual layer thickness of PMMA is about 0.3 μm.

In order to verify the role of PMMA layer thickness on the overall ballistic performance, a second set of composites were prepared with either the total number of layers increased to 4096-layer or an increase of the PC content to either 90 or 95 vol.%. Figure 6 summarizes the impact response obtained for the 1024-layer 80/20 PC/PMMA as well as 2048- and 4098-layer composites that consist of 5, 10, and 20 vol.% of PMMA. The 4096-layer 80/20 PC/PMMA composite exhibited much better ballistic strength compared to the corresponding 2048-layer multilayer; ductile deformation occurred in the former similar to that predominant in the pure PC. As the PC content further increased to 90 or 95 vol.%, composites consisting of either 2048 or 4096 layers displayed ductile response to the impact. A ring pattern was clearly seen in these multilayers which revealed that the extent of shock wave dissipation was across the entire 2-in-diameter area and not just limited to the immediate vicinity of impact. These ballistic test results further indicated that control of the brittle PMMA phase present in these multilayers is critical for their overall impact performance. Table 2 lists the calculated layer thickness obtained for the second set of PC/PMMA multilayers. Composites with individual layer thickness of PMMA being 0.14 μm or thinner exhibited ductile deformation upon impact; however, a mixed mode of failure occurred as the PMMA layer thickness reached approximately 0.3 to 0.4 μm. The latter appeared to be corresponding for a ductile/brittle transition evidenced in the Figure 7 for comparing the impact response of a series of 2048-layered composites. Fracture photographs shown in

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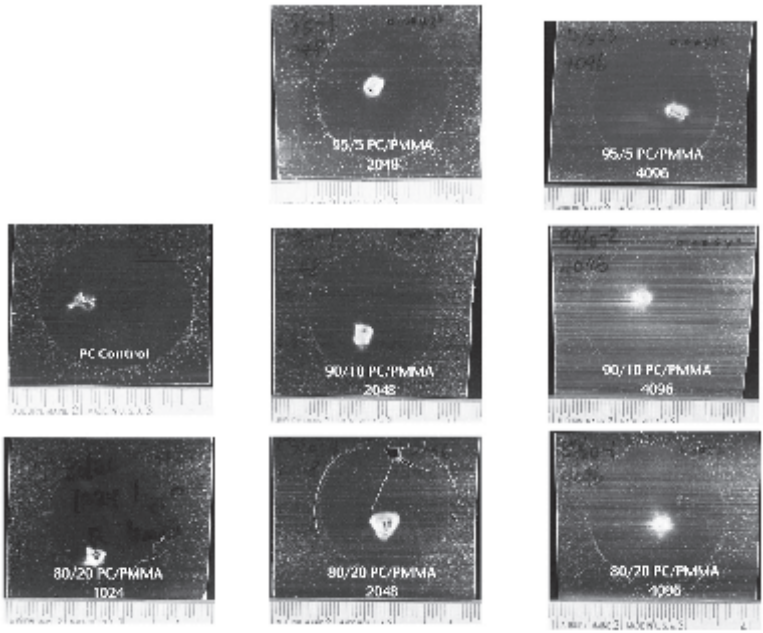

Figure 6. *Photographs of ballistic impact response of 1024-layer 80/20 PC/PMMA as well as 2048- and 4096-layer composites that contain 80, 90 and 95 vol.% of PC.*

Figure 8 further confirm the reproducibility of the mixed mode observed in the 2048-layer 80/20 PC/PMMA composites.

It is apparent that individual layer thickness of PMMA plays a critical role in determining the ballistic impact behavior of these coextruded PC/PMMA composites. Multilayers with PMMA layer thickness reached 0.5 mm always failed in a brittle mode regardless of their composition or configuration.

Table 2. Average sheet thickness of 256-, 1024-, and 2048-layer PC/PMMA multilayers and the calculated individual layer thickness of PC and PMMA for the corresponding composites.

	1024-Layer		2048-Layer		4096-Layer	
PC/PMMA	Sheet Thickness (mm)	Layer Thickness (μm)	Sheet Thickness (mm)	Layer Thickness (μm)	Sheet Thickness (mm)	Layer Thickness (μm)
95/5	n.a.	n.a.	1.1	1.0/0.05	1.26	0.6/0.03
90/10	n.a.	n.a.	1.14	1.2/0.14	1.62	0.7/0.08
80/20	0.9	1.4/0.4	1.4	1.1/0.3	1.4	0.5/0.14

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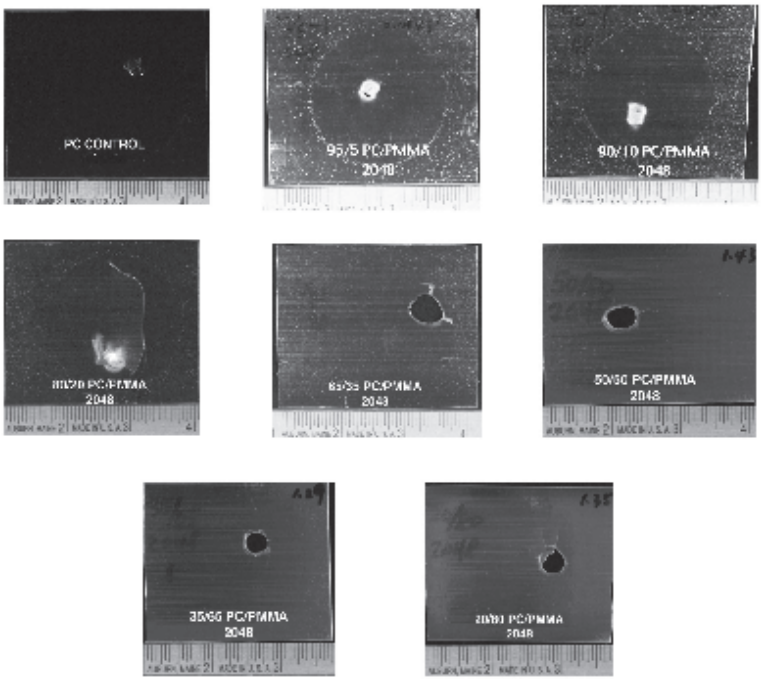

Figure 7. *Photographs of ballistic impact response of 2048-layer PC/PMMA composites.*

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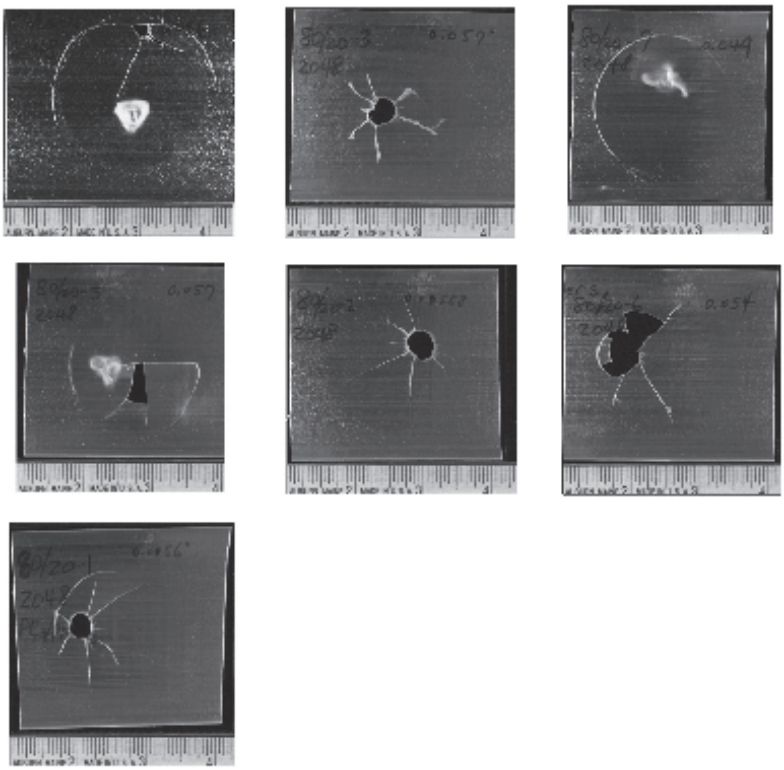

Figure 8. *Photographs showing a mixed mode of failure observed in 2048-layer 80/20 PC/PMMA composites.*

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Measurements of Ballistic Impact Energy

Kinetic energy absorbed or dissipated during the ballistic impact was calculated from the experimentally determined striking and residual velocities of the projectile; these ballistic impact energy values were then normalized by the thickness of each composite. Figure 9 shows the plot of normalized ballistic impact energy values vs. volume-percent of PC for the available PC/PMMA composites. Although data in Figure 8 were scattered, a strong dependence of the ballistic impact energy upon the PC content was clearly seen. For composites with less than 65 vol. % of PC (shown in the shaded region A), the determined values were relatively low and little improvement in the ballistic performance was seen as the PC content increased. The low ballistic impact energy values corresponded to a brittle mode of failure observed in the corresponding 256-, 1024- and 2048-layer PC/PMMA composites. A drastic increase in the ballistic impact energy occurred as the PC content increased to above 80 vol.%, particularly 90 and 95 vol.% (as shown in the shaded region B). Composites with these corresponding compositions exhibited

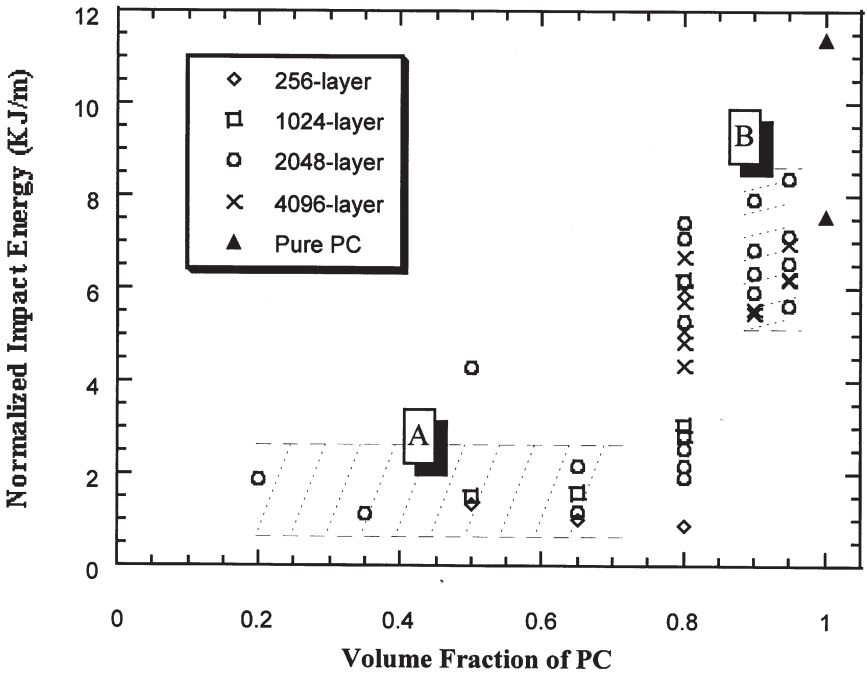

Figure 9. *Plot of normalized ballistic impact energy values as a function of PC content and total number of layers; shaded region A and region B denote that composites failed in a brittle and ductile mode respectively.*

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ductile deformation; however, a brittle-ductile transition appeared to be present with the PC content around 80 vol.%. The latter was dependent upon the total number of layers of these coextruded composites.

Figure 10 revealed the effect of the total number of layers on the ballistic performance of composites containing 80 vol.% of PC; it is clear that impact strength increased as the number of layers increased. The 4096-layer 80/20 PC/PMMA composite exhibited a ductile mode of failure; however, the 256-layer composite with the corresponding composition failed in a brittle mode. This indicated that increasing the PC content may improve the impact strength; however, the overall ballistic performance was dominated by the layer thickness of PMMA.

Effect of PMMA Layer Thickness

Figure 11 is a plot of normalized ballistic impact energy determined for all the composites as a function of individual layer thickness of PMMA of the corresponding composite. Ballistic strength increased significantly as the layer thickness of PMMA reduced to approximately 0.36 mm. The normalized impact energy

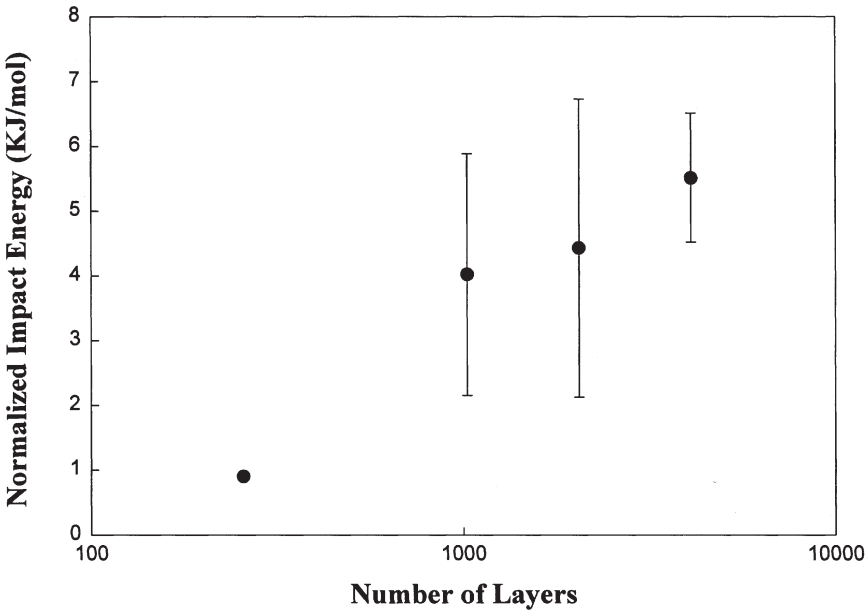

Figure 10. *Plot of normalized ballistic impact energy values obtained for 256-, 1024-, 2048-, and 4096-layer 80/20 PC/PMMA composites.*

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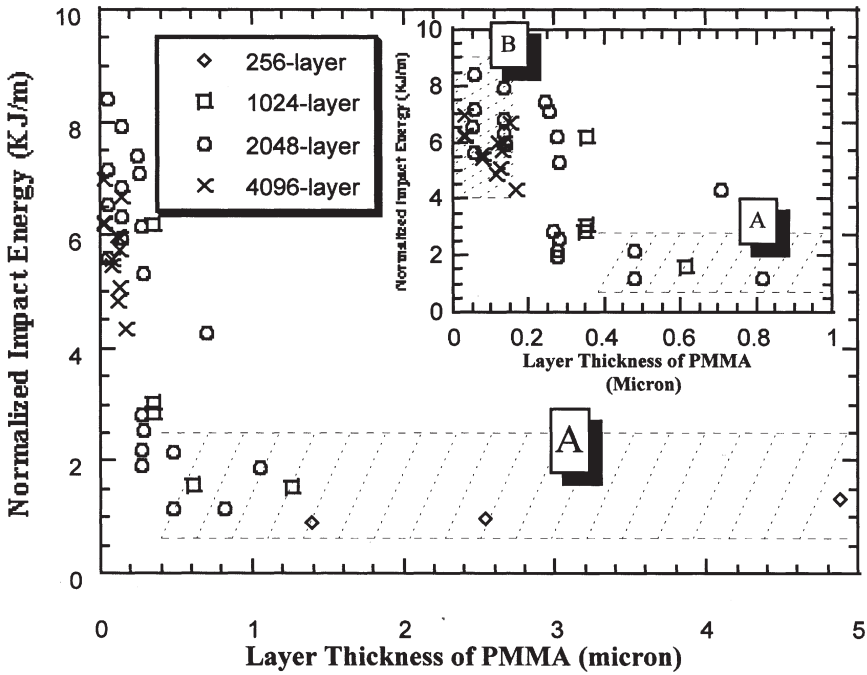

Figure 11. *Plot of normalized ballistic impact energy values versus PMMA layer thickness; the insert is an enlarged plot for PMMA layer thickness smaller than 1 μm; shaded region A and region B denote that composites failed in a brittle and ductile mode, respectively.*

was consistently higher as the PMMA layer thickness further reduced to 0.14 μm or thinner; this corresponded to ductile deformation observed in all the 4096-layer composites and the 2048-layer composites with 90 and 95 vol.% of PC. Impact performance decreased dramatically as the layer thickness of PMMA reached approximately 0.5 μm, and as a result brittle mode of failure occurred in these composites regardless of their composition and layer configuration.

CONCLUSION

Ballistic impact response of coextruded PC/PMMA composites with various compositions and layer configuration were investigated. The scaling effect of PMMA appeared to be critical; the determined impact energy values increased significantly as the layer thickness of PMMA reduced to approximately 0.36 μm. Furthermore, composites with PMMA layer thickness reduced to around 0.14 μm

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exhibited ductile mode of failure, similar to that observed in the pure PC. The damage zone associated with the ductile deformation was limited to the immediate vicinity of impact. A ring pattern was observed on the fracture surface of these composites, which indicated that the shock wave energy dissipation across the test specimen was significant during the projectile penetration. As a result, impact energy values obtained for these coextruded composites were consistently higher. A brittle mode of failure, however, was encountered in all the PC/PMMA composites as the thickness of the PMMA layers reached 0.5 μm or higher, regardless of their composition and configuration. Adhesion between the layers appeared to be very good since delamination did not occur whether these multilayers failed in either a ductile or a brittle mode. Results of this work clearly demonstrated a design rationale that proper control of layer thickness of the brittle component in a ductile/brittle hybrid is critical for their overall ballistic impact strength.

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