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EFFECT OF ADHESIVE ON THE IMPACT RESISTANCE OF  
LAMINATED PLASTICS FOR WINDSHIELD APPLICATIONS

ARMY MATERIALS AND MECHANICS RESEARCH CENTER

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**EFFECT OF ADHESIVE ON THE IMPACT RESISTANCE OF  
LAMINATED PLASTICS FOR WINDSHIELD APPLICATIONS**

Technical Report by  
*JOYCE L. ILLINGER and ROBERT W. LEWIS*

August 1973

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ABSTRACT

Clamped acrylic-polycarbonate laminates (polycarbonate as backup) show improved resistance to penetration over that shown by either material alone. This resistance is dependent upon relative amounts of each material. Use of thermal bonding techniques to form a laminate resulted in a significant decrease in the ballistic resistance of these laminates. Use of a brittle epoxy adhesive to bond the materials gave similar results. The brittle adhesive induces spallation of the polycarbonate causing significantly lower impact resistance and changes the mode of failure from ductile to brittle.

Several flexible, transparent, segmented polyurethane adhesives were synthesized with systematic variation in proportions of monomer (diisocyanate and chain extender) and structure of the soft segments. Glass transition temperatures showed small differences with compositions. Ballistic resistance of laminates formed using these adhesives was improved by approximately forty percent over that of the clamped laminate. The change in relative amount of the hard segment (diisocyanate and chain extender) had no statistically significant effect upon the impact behavior. At constant molecular weight of the soft segment, variation of the structure also showed no effect.

Several proprietary flexible film adhesives were also tested. Strength of adhesive bonding was varied by a factor of two through changes in laminating temperature. However, no correlation between bond strength and ballistic performance was found. These film adhesives formed laminates slightly better than those using polyurethane adhesives. Thus, laminates using transparent flexible adhesives with sufficient bond strength to prevent delamination upon impact show ballistic resistance up to fifty percent better than Lexan polycarbonate which is currently considered the best commercially available transparent impact-resistant polymeric material.

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## INTRODUCTION

Currently Plexiglas is the transparent polymeric impact-resistant material in widespread use for aircraft applications. Lexan polycarbonate with slightly higher impact resistance and a ductile mode of failure is replacing Plexiglas in some applications. However, there are some problems due to poor abrasion and poor solvent resistance. It is desirable to further increase the impact behavior of this type of material. This can be accomplished through three principal techniques: (1) toughening polymers by the inclusion of a rubbery material forming a copolymer (graft, block, random); (2) by the introduction of preferred orientation; and (3) by lamination of dissimilar materials.<sup>1</sup>

The synergism in ballistic performance due to lamination of dissimilar material has been shown for several systems including glass/plastic and plastic/plastic laminates.<sup>2</sup> Since Lexan and Plexiglas are very different polymeric materials it was not unreasonable to assume that laminates of these two materials would show enhanced ballistic resistance. As preliminary studies bore this out, an extensive investigation of the polymethyl methacrylate/polycarbonate (PMMA/PC) without and with an adhesive interlayer was carried out.

## RESULTS AND DISCUSSION

### No Bonding, No Interlayer

The ballistic performance of clamped Plexiglas/Lexan laminates was investigated over a range of relative compositions with results shown in Figure 1. At the extreme left is the ballistic resistance of 100% PMMA with a  $V_{50}$  of 1040 ft/sec. At the extreme right is 100% polycarbonate with a  $V_{50}$  of 1100 ft/sec.

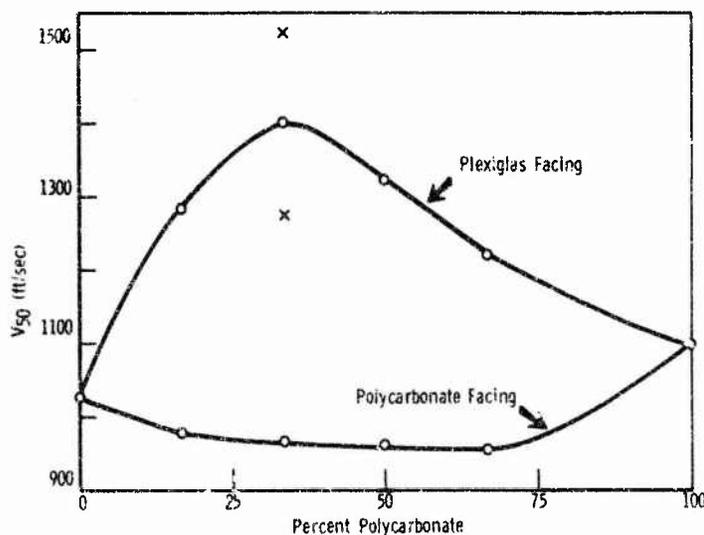


Figure 1. Ballistic Behavior of Plexiglas - Polycarbonate Laminates (17-Grain Fragment Simulators, 38 oz/sq ft)

19-066-1679/AMC-71

<sup>1</sup>ROYLANCE, M. E., and LEWIS R. W. *Development of Transparent Polymers for Armor*. Army Materials and Mechanics Research Center, AMMRC TR 72-23, July 1972.

<sup>2</sup>LEWIS, R. W., and PARSONS, G. R. *Ballistic Performance of Transparent Materials for Eye Protection (U)*. Army Materials and Mechanics Research Center, AMMRC TR 72-36, November 1972 (Confidential Report).

With PMMA facing the impact (upper curve), the optimum ballistic resistance of the combinations tested occurred at a 2:1 weight ratio of PMMA to PC ( $V_{50} = 1400$  ft/sec) in good agreement with the optimum ratios for glass to plastic. With the ductile PC facing the impact (the lower curve), however, there was a reduction in ballistic resistance compared to that of the homogeneous materials over the range of compositions investigated.

Under ballistic impact, with the 22-caliber 17-grain fragment-simulating projectile (FSP), PMMA by itself behaves in a brittle fashion and undergoes cracking and spallation (Figure 2a). Polycarbonate, on the other hand, is ductile, does not spall, and fails in shear (Figure 2b). Both materials, however, have essentially the same  $V_{50}$  (PMMA  $V_{50} \sim 1040$ , PC  $V_{50} \sim 1100$  ft/sec, Figure 1). In the laminated system, though, the brittle PMMA, when it is facing the impact, spreads out the impact over a wider area than a ductile material, while the ductile PC then serves both to absorb the impact and to prevent the brittle material from spalling (Figure 2c), thereby allowing more extensive damage and raising the PMMA's resistance to penetration. With the ductile layer facing, however, the load is not spread out as efficiently, and there is nothing to prevent the brittle material from spalling (Figure 3); hence less energy is absorbed during the penetration process. Consequently, the more efficient combination ballistically is for the more brittle PMMA to face the impact.

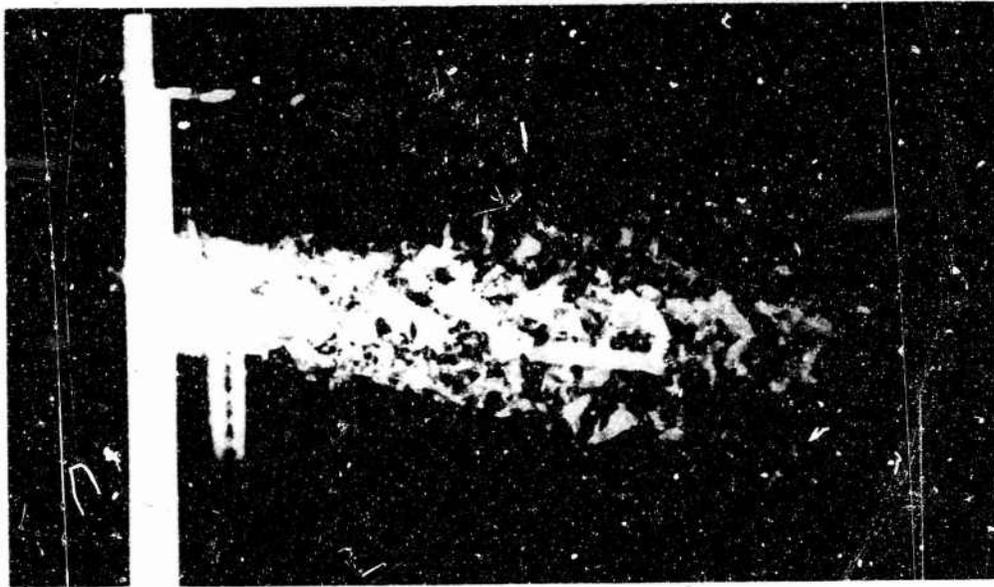
#### Thermal Bonding, No Interlayer

In order for this laminated system to be useful for aircraft applications it would be necessary to bond the materials together, either thermally or by the incorporation of an adhesive interlayer. A thermally bonded sample would have the advantage of maintaining optical clarity without the problems associated with applying transparent adhesives to polycarbonate. Accordingly, a piece of 1/4" PMMA (Lucite molding powder) and 1/8" Lexan were placed in a 6" x 6" mold. The mold was put into an 18" x 18" press and heated under an 18-ton load to 150 C. The press was held at a temperature in the vicinity of 150 C for thirty minutes and then cooled to room temperature.

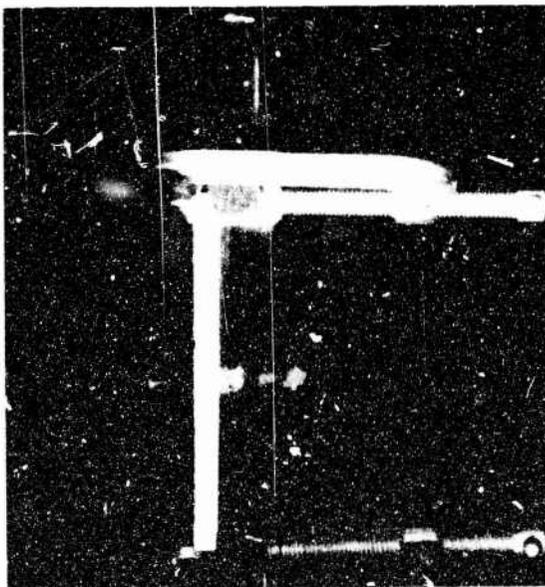
The resultant sample was ballistically evaluated and found to be substantially inferior to the nonbonded sample ( $V_{50} = 1110$  ft/sec at 36.9 oz/sq ft versus  $V_{50} = 1400$  ft/sec for the nonbonded sample from Figure 1). It is interesting to note that unlike the situation when the samples are clamped together (Figure 2c), here the polycarbonate is failing in a brittle fashion and spalling under ballistic impact (Figure 4). It is this spallation that results in the poorer ballistic resistance of the thermally bonded samples.

To verify that this degradation was not due to the thermal treatment, per se, however, Lucite and Lexan were individually subjected to the same thermal treatment as the laminate and ballistically evaluated. The results are given in Table I.

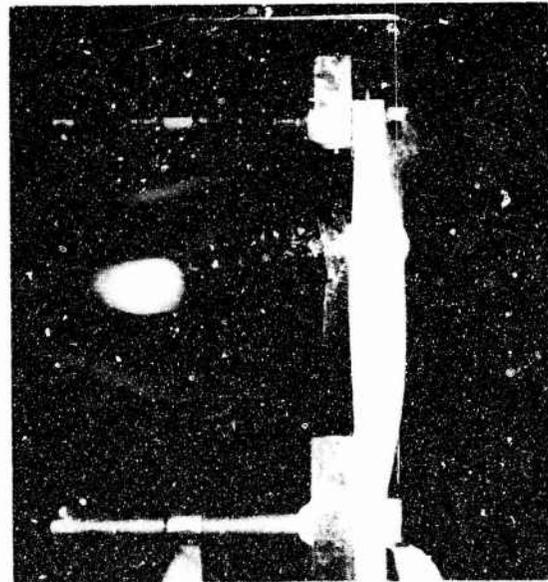
It is evident from these data that the thermal treatment was not responsible for the degradation in ballistic resistance. Rather, it is apparently due to the absence of an interlayer. The interlayer, which may be either an adhesive or an air gap (as in the case of the clamped samples), prevents the polycarbonate from spalling.



a. 1/4" Plexiglas



b. 1/4" Lexan



c. Plex/Lex Laminate  
Plexiglas is Facing Impact

Note Front Spall from Plexiglas but no Rear Spall from Lexan

Figure 2. Multiflash photographs of Plexiglas, Lexan, and Plex/Lex Laminate Under Ballistic Impact. Edges of Samples are Shown, Missiles are Moving from Left to Right.

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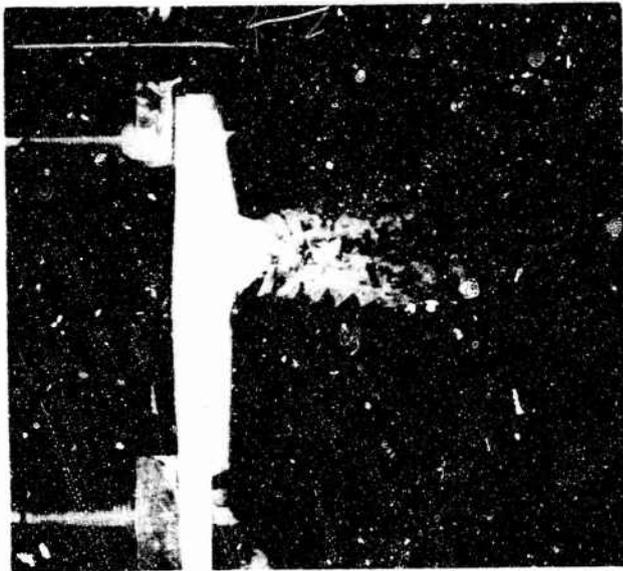


Figure 3. Multiflash Photograph of Plex/Lex Laminate Under Ballistic Impact. Edge of Sample is Shown, Lexan is Facing Impact, Missile is Moving from Left to Right. Note Rear Spall from Plexiglas.  
19-066-159/AMC-72

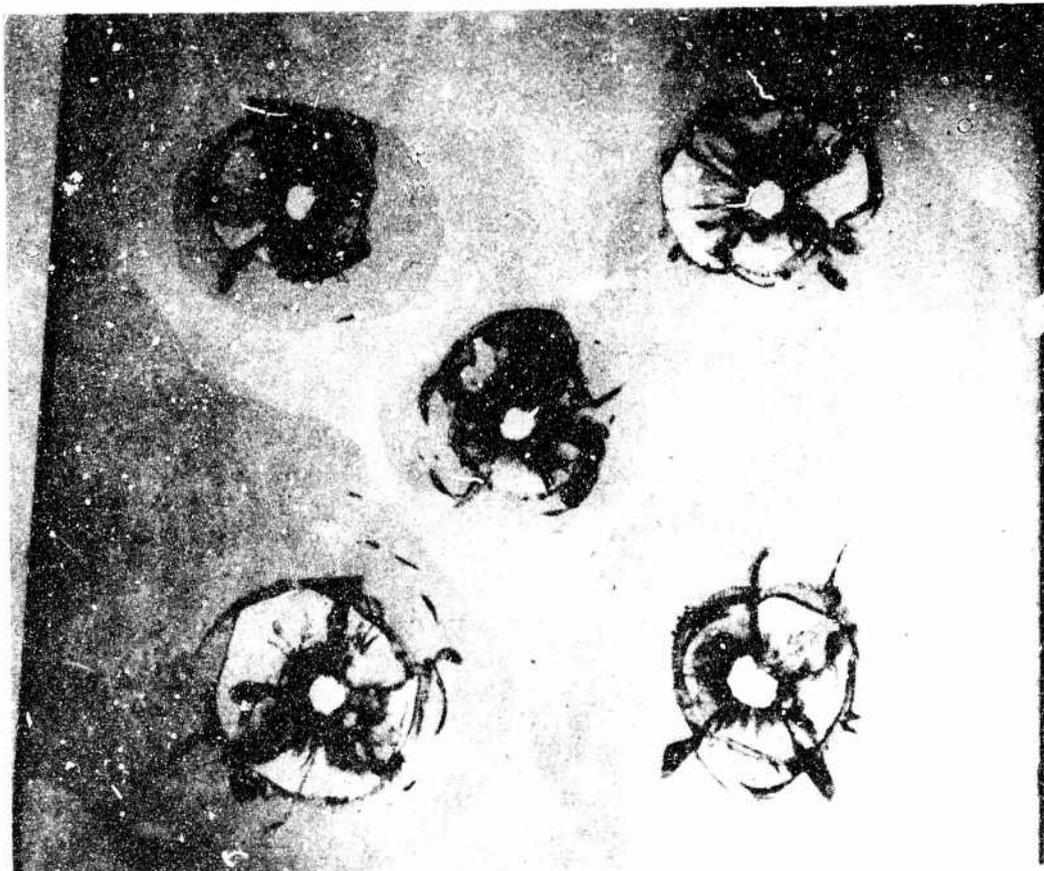


Figure 4. Thermally Bonded Plex/Lex Laminate, Rear View. Note Lexan Back-Up has Spalled Under Impact  
19-066-155/AMC-72

Table I. BALLISTIC BEHAVIOR OF PMMA AND LEXAN\*

Material Description	Areal Density (oz/sq ft)	V <sub>50</sub> (ft/sec)	Penetration (ft/sec)	
			High Partial	Low Complete
Lucite PMMA Mounting Powder	36.2	1096 (4 shot)	1071	1122
Plex 55 PMMA (control)	37.4	1040 (8 shot)	1015	1072
Lexan Press Molded	24.5	900 (6 shot)	915	900
Lexan (control)	25.4	905 (6 shot)	908	899

\*17-grain FSP, 0° obliquity

The next step, then, was to investigate the effects of various adhesive interlayers.

#### Adhesive Bonding, Brittle Interlayer

Several brittle interlayers were evaluated. Typical of these was a commercially available epoxy adhesive, EPON 828 cured with 13 pph DEH24. A 12" x 12" laminate (2 PMMA: 1 PC) was fabricated using this adhesive system and tested ballistically. A complete penetration was obtained at 1291 ft/sec with a 6"-diameter area of delamination. Damage to the facing Plexiglas was minimal, but the polycarbonate cracked and also spalled, unlike its normal ductile performance.

In an effort to explain the reason for this degradation, a 12" x 12" sample of 1/8" Lexan was coated with a 15-mil-thick layer of the EPON 828 adhesive. The ballistic resistance is given in Table II together with that of a control piece of Lexan.

There occurred about a 10% decrease in V<sub>50</sub> but a much larger overlay between partial and complete penetrations. In addition, the adhesive-coated Lexan spalled badly, quite unlike the normal ductile impact behavior (see Figures 5 and 6). Some of the adhesive was then removed from the Lexan, and the sample was reimpacted. The ductile behavior returned indicating that the adhesive had not chemically affected the polycarbonate. Apparently, then, even a thin layer of brittle material is sufficient to affect the ductility and thus to lower the ballistic resistance of polycarbonate itself and of a laminate containing polycarbonate.

The poor ballistic behavior obtained with the brittle epoxies made it obvious that a more flexible adhesive was required.

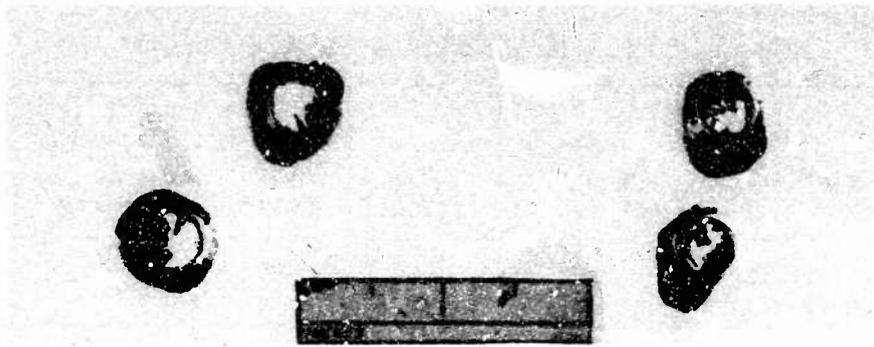


Figure 5. Uncoated Lexan Subjected to Ballistic Impact  
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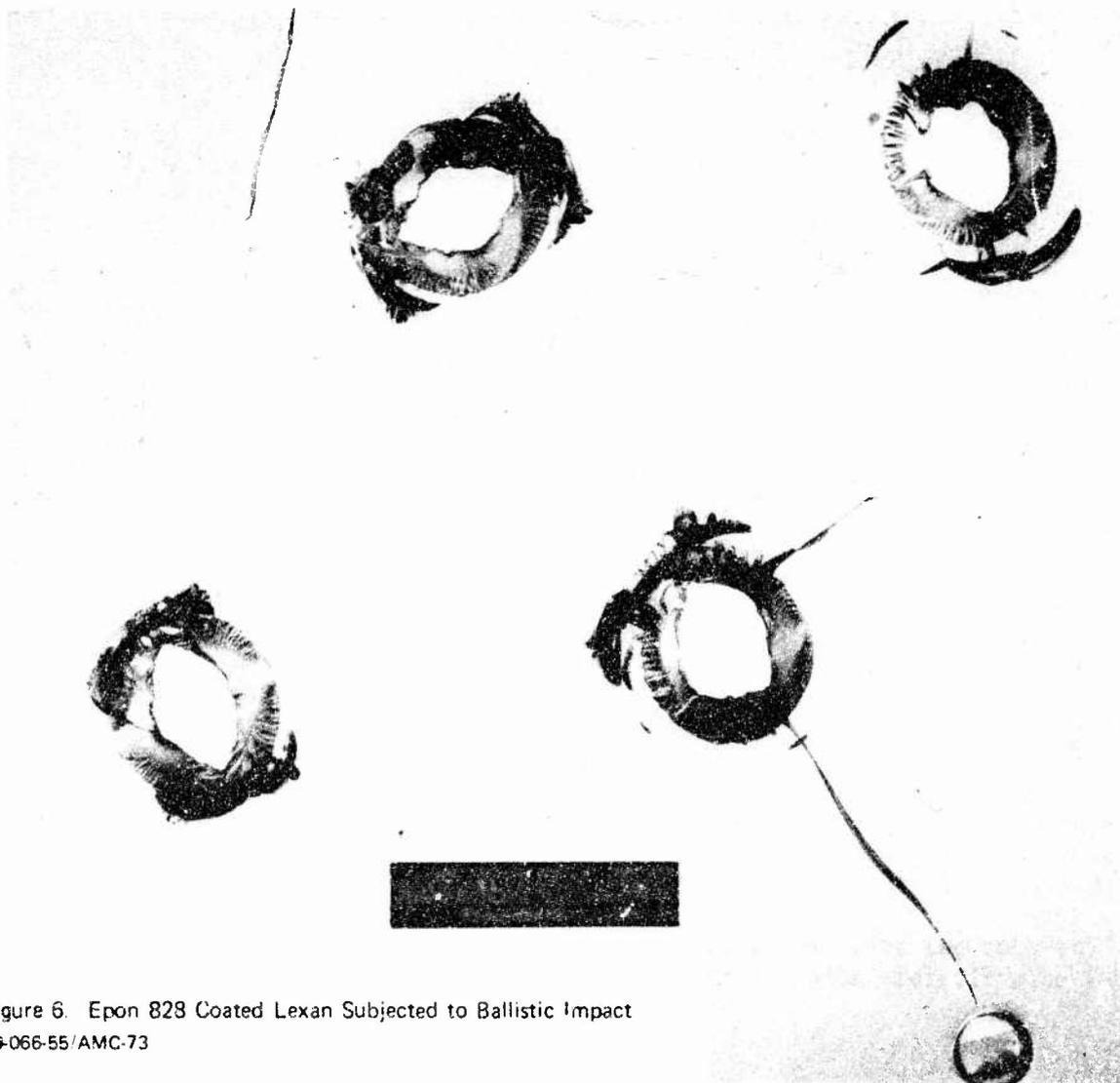


Figure 6. Epon 828 Coated Lexan Subjected to Ballistic Impact  
19-066-55/AMC-73

Table II. BALLISTIC BEHAVIOR OF LEXAN\*

	Lexan with EPON 828 Coating	Control Lexan (No Coating)
V <sub>50</sub> (ft/sec)	682 (10 shot)	767 (8 shot)
Penetration (ft/sec)		
High Partial	714	771
Low Complete	639	753
Areal Density (oz/sq ft)	14.2 (12.8 Lexan + 1.4 EPON 328)	12.8

\*17-grain FSP, 0° obliquity, coating facing impact

### Adhesive Bonding, Flexible Interlayer

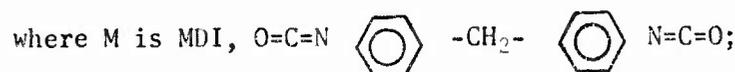
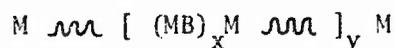
#### a. Survey

Several commercially available flexible adhesives were evaluated. The results are given in Table III.

It is apparent that these flexible adhesives are substantially better ballistically than the epoxies and are all superior to the samples with no adhesive interlayer shown in Figure 1. However, each adhesive has a serious drawback. The Silastics are generally white or milky white and not optically acceptable, and the experimental clear Silastic delaminated after one shot. The Uralane 5716 cast-in-place polyurethane does not give very good adhesion, and total delamination occurs with one impact. The Desmocoll 400, a linear aliphatic polyurethane, is theoretically transparent but actually turned out to be white and optically unacceptable. Since the polyurethanes appeared to give good ballistic results a program was initiated to prepare systematically varied materials.

#### b. Synthesis and Composition

A standard two-step synthesis was used to prepare three series of adhesive polymers. The general structure may be represented as



B is 1, 4 butanediol, HO CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>OH; and

$\text{---} \text{---}$  is a polymeric glycol, HO(CH<sub>2</sub>CH<sub>2</sub>O)<sub>a</sub> (CH<sub>2</sub>CHO)<sub>b</sub> (CH<sub>2</sub>CH<sub>2</sub>O)<sub>a</sub> H  
|  
CH<sub>3</sub>

Table III. BALLISTIC BEHAVIOR OF PLEX/LEX LAMINATES WITH COMMERCIAL ADHESIVES\*

Adhesive	Areal Density (oz/sq ft)	V <sub>50</sub> (ft/sec)	Penetration (ft/sec)	
			High Partial	Low Complete
Silastic - Si Rubber (milky white)	39.5	1496 (4 shot)	1487	1493
Silastic - Si Rubber† (milky white)	38.0		1255	1427
Silastic - Si Rubber (experimental clear)	39.8		1340 (samples delaminated after 1 shot)	
Silastic - Si Rubber (white opaque)	40.3		1538	
Uralane 5716 Polyurethane (brownish-transparent)	39.7		1431 (samples delaminated after 1 shot)	1504
Desmocoll 400 Polyurethane (white)	39.8	1429 (4 shot)	1437	1437

\*17-grain FSP, 0° obliquity, 1/4" Plexiglas laminated to 1/8" Lexan

†Double laminate - 1/8" Plex/1/16" Lexan/1/8" Plex/1/16" Lexan

Table IV shows the variation in composition and structure of the available polymeric glycols.

All three series had y held constant. Series I was made using L35 as the glycol and varying x from 0 to 1.0 at intervals of 0.2. Series II was made holding x constant at 0.4 and making polymers using each of the seven polymeric glycols for the soft segment. Series III was similar to series II except that x = 0.5.

Two additional polymers were evaluated, both of composition 3.2 MDI: 2 Butenediol: 1 Glycol. One had a soft segment of molecular weight 1000 (a = 1, b = 17) and the other a soft segment of molecular weight 2000 (a = 11, b = 17).

#### c. Description and Characterization

The first series of polymers ranged from extremely sticky with very little elastomeric behavior to slightly tacky with very little permanent set, Table V.

Table IV. GLYCOL COMPOSITION AND STRUCTURE

Glycol	Composition	a	b
C1540	100% PEG	0	35
L35	50/50 PPG/PEG	11	17
L44	60/40 PPG/PEG	10	23
L43	70/30 PPG/PEG	6	23
L42	80/20 PPG/PEG	3	22
L61	90/10 PPG/PEG	2	33
P2010	100% PPG	34	0

From  $x = 0.0$  to  $x = 0.6$  the polymers were transparent becoming translucent at  $x = 0.8$  and  $1.0$ . The molecular weights of all but  $x = 0$  are  $\sim 40,000$  so that the gradual change in tackiness, elastomeric properties, and transparency must be due to change in structure due to increasing butanediol content. Differential scanning calorimetry was performed to determine  $T_g$  to see if there was any change with structure which might be related to ballistic performance.  $T_g$  shows a monotonic increase with increasing butanediol content (Figure 7) over a 15 C temperature range. Leveling off of  $T_g$  occurs above the butanediol content which remains transparent.

Table VI summarizes the properties of series II and III.

It appears that increasing amounts of polypropylene glycol in soft segments of around 2000 molecular weight leads to translucency at lower butanediol content.

$T_g$  behavior of these series showed no meaningful change.

Both samples with high MDI content were opaque. The polymer with the lower molecular weight soft segment showed some embrittlement as compared to the other.

Table V. VARIATION OF HARD SEGMENT

MDI	Butanediol	Glycol	Description	$\bar{M}_n$
1.05	0	1	sticky, gradual flow, set, transparent	30,000
1.261	0.2	1	sticky, set, slightly elastomeric, transparent	39,000
1.471	0.4	1	tacky, less set, elastomeric, transparent	40,000
1.576	0.5	1	less tacky, still some set, elastomeric, transparent	40,000
1.681	0.6	1	slightly tacky, very little set, elastomer, transparent	41,000
1.891	0.8	1	little tack, very little set, elastomer, translucent	42,000
2.1	1	1	nontacky, no rapid set, elastomer, translucent	45,000

d. Ballistic Resistance of Laminates

Laminates from 1/4" Plexiglas 55, 10 mil adhesive, 1/8" Lexan were fabricated at press conditions of 4000-lb ram force over a 4"-diameter ram at 95 C for 40 minutes residence time. The results of ballistic evaluation are shown in Table VII.

As long as the materials remain transparent the  $V_{50}$ 's of the laminates are essentially the same for this polymeric glycol. At sufficiently high butanediol content which results in loss of transparency, the  $V_{50}$  drops. The 1.0 butanediol content polymer was nontacky, indicating some embrittlement. The lowered

Table VI. PROPERTIES OF SERIES II AND III

Glycol	Series II 1.471 MDI: 0.4 Bd: 1 glycol	Series III 1.576 MDI: 0.4 Bd: 1 glycol
100% PEG	tacky, opaque in bulk (transparent in laminate)	tacky, opaque in bulk (transparent in laminate)
50/50 PPG/PEG	tacky, transparent	tacky, transparent
60/40 PPG/PEG	tacky, transparent	tacky, translucent
70/30 PPG/PEG	sticky to tacky, transparent	tacky, transparent
80/20 PPG/PEG	sticky, transparent	sticky to tacky, transparent
90/10 PPG/PEG	sticky, translucent	sticky, translucent
100% PPG	very sticky, translucent	sticky, translucent

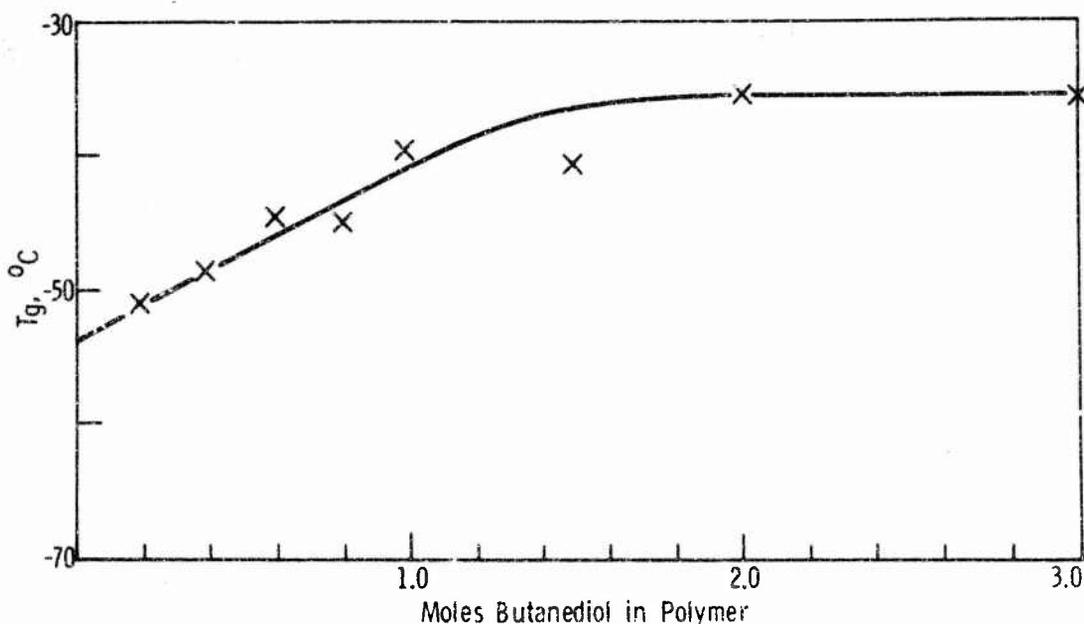


Figure 7. Glass Transition Temperature Versus Butanediol Content

ballistic performance is in agreement qualitatively with earlier observations on the effect of brittle adhesives. Very small areas of delamination were observed with the synthesized transparent polymers in contrast to those commercially available. These adhesives showed definite improvement ballistically over the earlier survey.

As seen in Table VIII the structure of the polymeric glycol (molecular weight 2000) at constant MDI and butanediol composition had no significant effect on the ballistic performance of the laminates. These polymers were all transparent at the 10-mil thickness of the adhesive layer used for laminate evaluation.

At another composition, i.e., 3.2 MDI: 2 Butanediol: 1 Polymeric Glycol, a  $V_{50}$  of 1300 ft/sec was determined for a polymer with glycol 90/10 PPG/PEG 1000 molecular weight versus a  $V_{50}$  of 1550 ft/sec for a polymer with glycol 50/50 PPG/PEG 2000 molecular weight. Since changing the ratio of PPG showed no effect on ballistic behavior we conclude that halving the length of the glycol has drastically affected the performance of the laminate.

Table VII. EFFECT OF BUTANEDIOL CONTENT ON BALLISTIC PERFORMANCE\*

Polymer Series I Butanediol Content	$V_{50}$ (ft/sec)	Optical Properties
0.0	1560	clear
.2	1570, 1600+	clear
.4	1510	clear
.5	1550, 1580+	clear
.6	1560	clear
1.0	1410	translucent

\*17-grain FSP, 0° obliquity, Plexiglas facing,  
8 shot  $V_{50}$

+Determination of  $V_{50}$  for duplicate polymers

Table VIII. EFFECT OF VARIATION OF POLYMERIC GLYCOL SERIES II AND III

Glycol	$V_{50}$ (ft/sec)*	
	Series II 1.47 MDI: 0.4 Bd: 1 glycol	Series III 1.576 MDI: 0.5 Bd: 1 glycol
100% PEG	1580	1584
50/50 PPG/PEG	1510	1550, 1580+
60/40 PPG/PEG	--	1585
70/30 PPG/PEG	1586	1584
80/20 PPG/PEG	1626	1581
90/10 PPG/PEG	1592	1602
100% PPG	--	1580

\*17-grain FSP, 0° obliquity, Plexiglas facing

+Determination of  $V_{50}$  for duplicate polymers

### e. Effect of Adhesive Bond Strength

An industrial proprietary adhesive to bond polycarbonate to glass and other plastics was evaluated in the Plexiglas/Lexan system. Two configurations, referred to as Type A and Type B, were investigated to determine the effect of bond strength upon ballistic performance. The strength of the adhesive bond was markedly dependent upon laminating temperature. Samples were fabricated using 10-mil Type B and 5-mil Type A adhesive at four different temperatures. These samples were evaluated ballistically and by the cross-lap tensile adhesion test ASTM D1344. This test uses a 1-square-inch cross section of adhesive between crossed adherends. The value of the peak load is a measure of the strength of the adhesive bond. The results of the adhesion test are shown in Figure 8.

It can be seen that over the range of temperatures investigated there is a twofold increase in the maximum load, while the Type A adhesive has twice the tensile peak load as Type B, in qualitative agreement with peel tests using modified ASTM D903. The ballistic behavior is shown in Table IX.

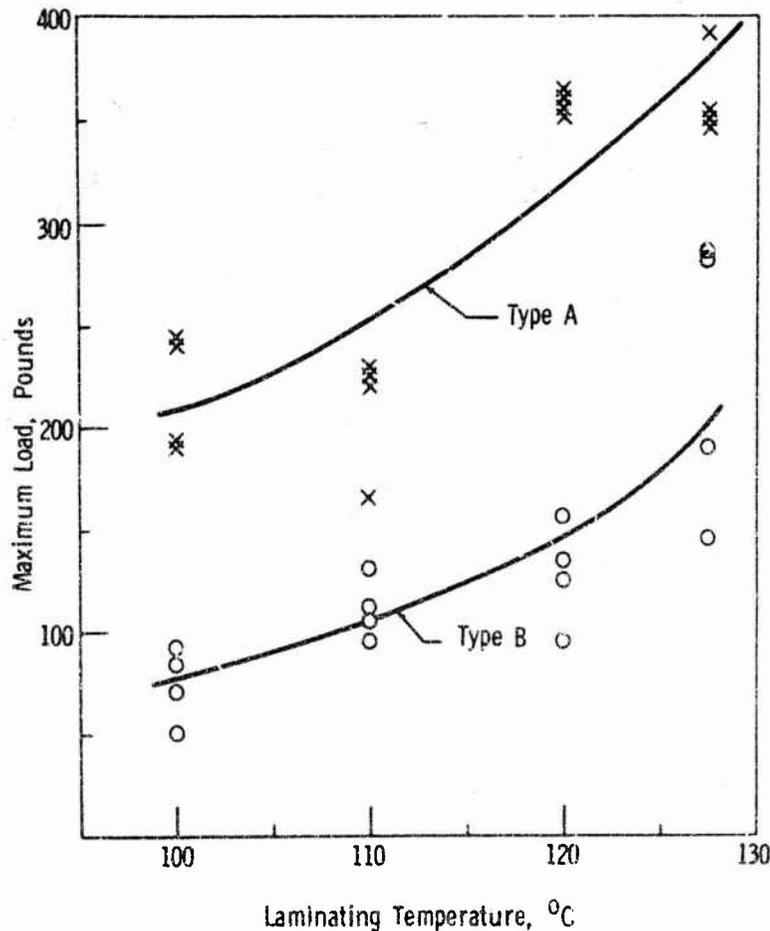


Figure 8. Adhesive Strength Versus Laminating Temperature

Table IX. BALLISTIC\* PERFORMANCE  
VERSUS LAMINATING TEMPERATURE

Type of Adhesive	Laminating Temp (°C)	4 shot V <sub>50</sub> (ft/sec)
Type A 5 mil	100	1625
	110	1626
	120	1645
	130	1635
Type B 10 mil	100	1605
	110	1600
	120	1613
	130	1630

\*Data obtained on 1/8" Plex 55 laminated to 1/8" Lexan, Type A adhesive = 275 psi, Type B adhesive = 500 to 600 psi pressure, 17-grain FSP, 0° obliquity.

Thus, it is evident that for the Plex/Lex system the strength of the adhesive bond does not affect the ballistic performance. Apparently, considering the marked effect of type of adhesive, it is the mechanical properties of the adhesive, per se, and not its adhesiveness, so long as delamination does not occur, that is the important parameter in determining the ballistic response.

#### ACKNOWLEDGMENT

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