

**DEVELOPMENT OF SILICONE OR
FLUOROCARBON INTERLAYER MATERIALS FOR
LAMINATED GLASS AND LAMINATED PLASTIC**

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FOREWORD

This report was prepared by the Dow Corning Corporation, under USAF Contract No. AF 33(600)-23081. The contract was initiated under Project No. 7340, Rubber Plastic and Composite Materials, Task No. 73403, Transparent Materials, formerly RDO No. 616-12 SR-7p, and was administered under the direction of the Materials Laboratory, Directorate of Research, Wright Air Development Center, with Lt. R. M. Mandel acting as project engineer.

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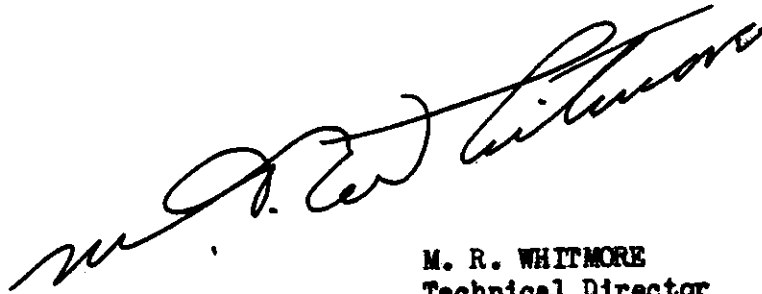
ABSTRACT

The development of a transparent interlayer material for laminated glass possessing thermal stability within the range of 400° to 500°F was the object of this research by the Dow Corning Corporation during the period of 15 April 1953 to 15 April 1954. Nine polysiloxane materials were introduced and evaluated for suitability as interlayers. A Type J interlayer was found which possessed tensile strength of 400 to 800 psi, 80 to 85% transmission, 4 to 10% haze and thermal stability of several hours at 450°F. This interlayer lends itself well to commercial fabrication techniques. The optical properties may require some improvement before this material will be completely satisfactory for aircraft glazing.

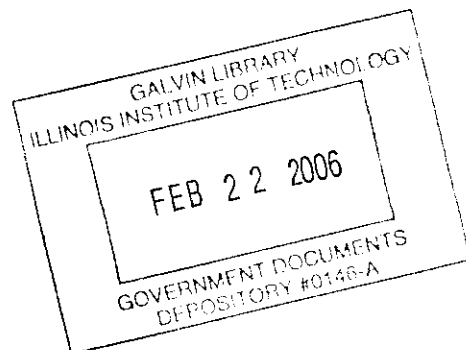
PUBLICATION REVIEW

This report has been reviewed and is approved.

FOR THE COMMANDER:



M. R. WHITMORE
Technical Director
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Contracts
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INTRODUCTION

Early in the 1930's the automobile industry began replacing plate glass windows in the new cars with plastic reinforced laminated glass. The purpose was to make the windows shatter-resistant and thus lessen the serious injuries due to broken glass. Plasticized cellulose nitrate was one of the first materials commercially used as an interlayer material. About 1935 plasticized cellulose acetate began to replace plasticized cellulose nitrate as the interlayer for auto safety glass, largely because of the instability of the nitrate to sunlight. By 1940 plasticized polyvinyl butyral was replacing both cellulose esters, primarily because of its superior stability, and adhesion even at low temperatures.

A very important application for laminated glass and laminated plastic windows is in the aircraft industry. Until the advent of jet propelled aircraft the butyral type interlayer met most requirements quite well. However, the high speeds encountered with jet planes necessitate many major changes in materials and design of the airplane. One of the factors leading to many changes is the high skin temperature developed by friction with the atmosphere. One of these changes will be the incorporation of heat stable shatter-resistant windows and canopies. At present there are no interlayer materials available that are operable at temperatures in excess of 400°F.

Silicones have created a name for being one of the most heat stable groups of polymeric compounds known today. Not only are these materials thermally stable, but they are also generally transparent. Silicones, therefore, represented a logical choice for potential interlayer development.

The objective of this program was to develop a silicone or fluorocarbon interlayer material for laminated glass and laminated polymethylalpha-chloroacrylate assemblies which possessed thermal stability at temperatures in the range of 400° to 500°F, optical clarity, adequate tensile strength, adhesion of surface sheets to the interlayer, and resistance to shattering under high velocity impact. Since no plastic surface sheets were available having thermal stability up to 400°F or above, the work under this contract was restricted to glass surface sheets.

The initial approach to the problem of developing a suitable thermally stable interlayer centered upon the evaluation of the available polysiloxane compounds which were considered potentially suitable for the application. As work progressed, special new compounds were developed by various Dow Corning research groups and their thermal stability was evaluated.

The interlayer materials studied were referred to as Type A, Type B..... In relation to these interlayer materials, the following specific items were investigated:

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1. Technique of conditioning the interlayer materials prior to forming laminates.
2. Technique of forming the laminates, including temperature conditioning.
3. Effect of interlayer thickness on the target properties.
4. The use of adhesives to improve the bond between the surface sheets and the interlayer materials.

This report is divided into three main sections. These are named General, Factual Data, and Summary. In Section I the subjects are Materials, Equipment, and Test Methods. In Section II each type of interlayer is discussed individually and data are presented. Section III summarizes the results collectively and points out the important findings of this work.

GENERAL

Part I Materials

The primary materials used in this work were polysiloxane interlayers and glass surface sheets of double strength sheet glass. Other materials used were polysiloxane adhesives, and a variety of additives.

Part II Equipment

The equipment used may be divided into processing equipment, and testing equipment.

A. Processing Equipment

Conventional two-roll rubber mills were used when necessary to work a particular interlayer material into an easy fabricating condition.

Steel chases were used for preparing laminates with high consistency interlayers. The chase enabled a laminate of desired uniform thickness to be prepared. A Silastic gasket was used in the chase cavity between the chase and the laminate to prevent run-off of interlayer material during the cross-linking process.

A special fluid polymer chase was designed for interlayers having a free flowing consistency prior to forming a laminate. Figure 1 shows this fluid polymer chase with the glass surface sheets in place. The bottom corners of the chase were hinged so that both sides of the chase could be moved. The top of the chase was held secure by two long bolts. Blue asbestos sheeting served as an edge seal to prevent the loss of the fluid interlayer during the cross-linking process. The surface sheets were easily aligned by placing a steel shim of the desired thickness between the surface sheets while they were being clamped in place. The shim was long enough to extend beyond the surface sheets so that it could be easily removed. The interlayer was poured between the glass surface sheets with the aid of an aluminum foil funnel.

Air circulating vented ovens were used to cure laminates held intact with the fluid polymer chase. These ovens were also used to give extended high temperature cures to laminates formed and precured in a press.

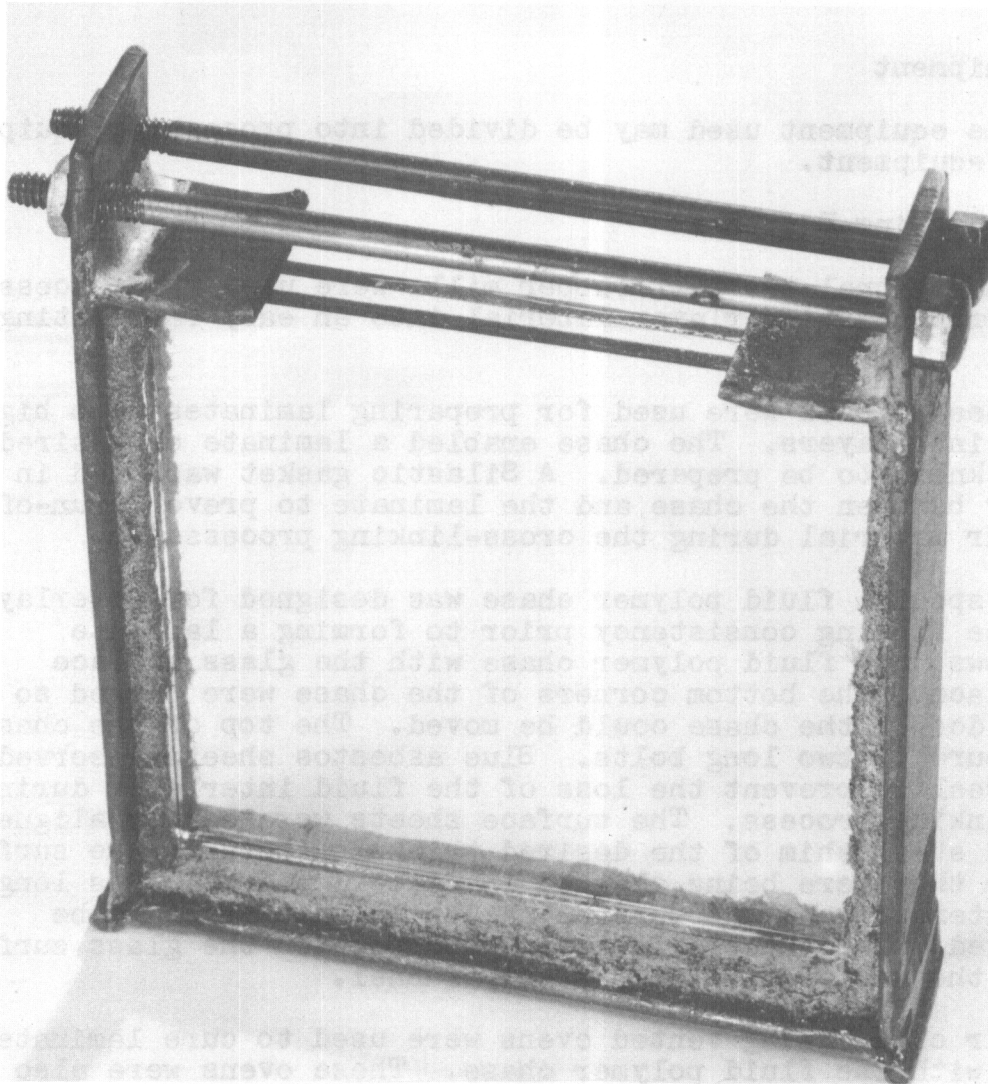


Figure 1

Fluid Polymer Chase

B. Testing Equipment

Thermal aging of laminates was done in vented air circulating ovens. Temperatures as high as 600°F were possible with these ovens.

A General Electric Spectrophotometer was used to obtain optical measurements of the laminates. These data were obtained as a service from the Dow Chemical Company.

A Scott L-6 tensile testing machine was used to obtain the tensile elongation properties of the interlayer materials. Adhesion measurements were also obtained with the L-6 testing machine. A Scott IP-4 tensile tester fitted with a high temperature box was used for obtaining tensile values at elevated temperatures.

A special gun trap was designed and built so that high velocity projectile impact tests could be performed on laminates. A .22 caliber Hornet rifle having a muzzle velocity of 2800 feet per second and an impact energy of approximately 750 foot pounds was used with the trap. Figure 2 shows the whole gun trap assembly. The gun and gun trap are labeled A and B respectively. The trap consists of a double walled steel construction throughout with insulation between the two walls. The bullet entrance hole may be seen in the door of the trap. A hole may also be seen in the top of the gun trap through which a steel ball may be dropped through for falling ball impact tests. The bracket, through which the one-half pound steel ball may be dropped, is labeled C. Letter D is a Brown Pyr-O-Vane temperature control-indicator for the gun trap. It controls over a temperature range of -100° to +600°F. The gun trap is heated by two heating elements which may be seen in Figure 3, Letter B. Letter A in Figure 3 shows the laminate rack in position for bullet impact. The bullet is deflected off a deflecting plate into a four inch bed of sand. The fan which circulates the air within the chamber may also be seen immediately behind the heating elements. For operation at low temperatures the gun trap is cooled by placing crushed dry ice in a wire basket, lettered B in Figure 4. The temperature is regulated at low temperatures by controlling the amount of circulation of air by the fan through the dry ice container. Also shown as letter A in Figure 4 is the laminate holder in position for falling ball impact tests.

Part III Test Methods

A. Thermal Aging

Thermal aging of laminates to determine their stability at elevated temperatures was accomplished in vented air circulating ovens. The general procedure followed was to determine at what temperature the laminates failed after various continuous periods at a particular temperature. The failure temperature was considered to be the temperature at which the interlayer or the adhesive bond between the interlayer and surface sheets developed optical flaws or underwent thermal degradation.

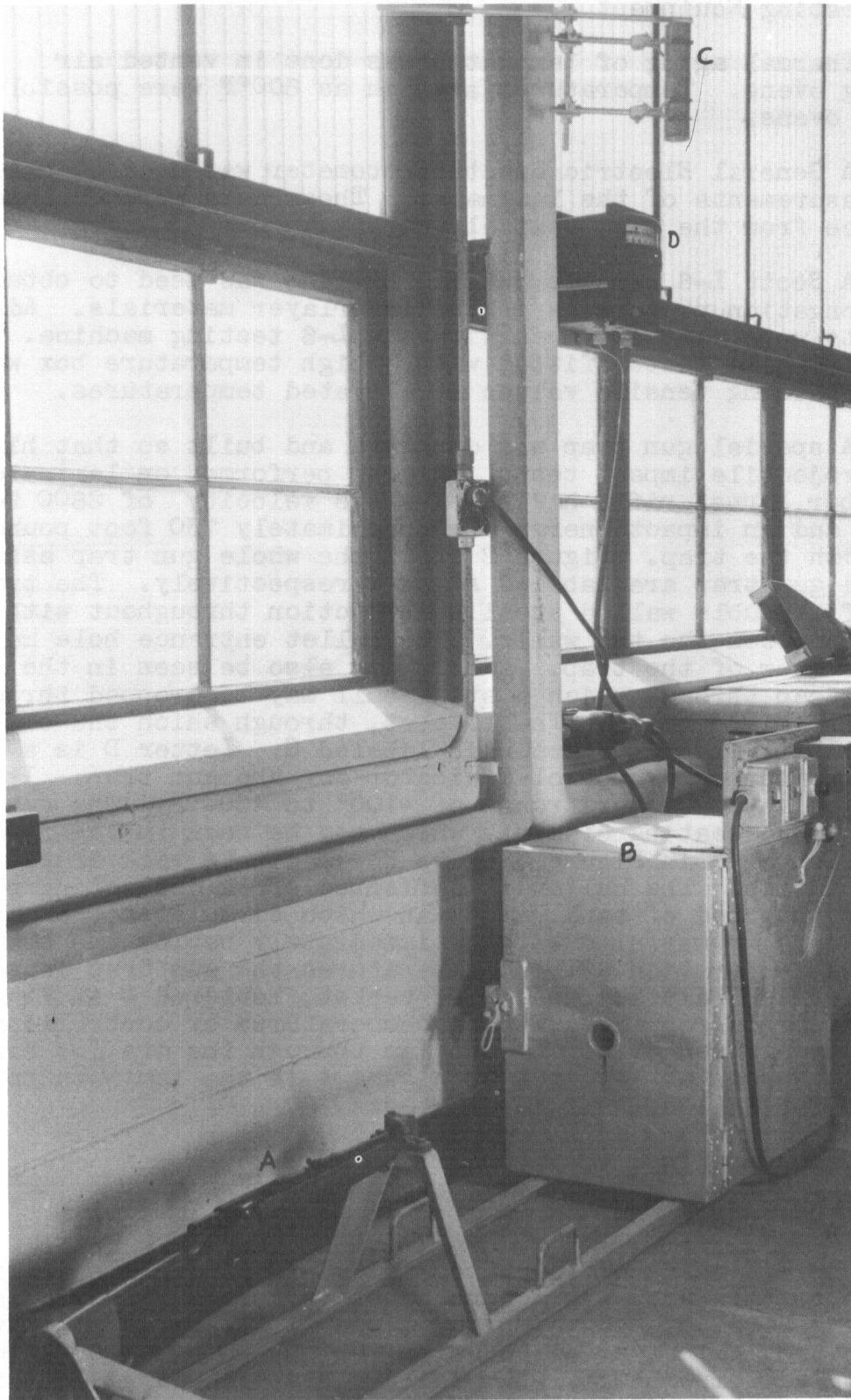


Figure 2
Gun Trap Assembly

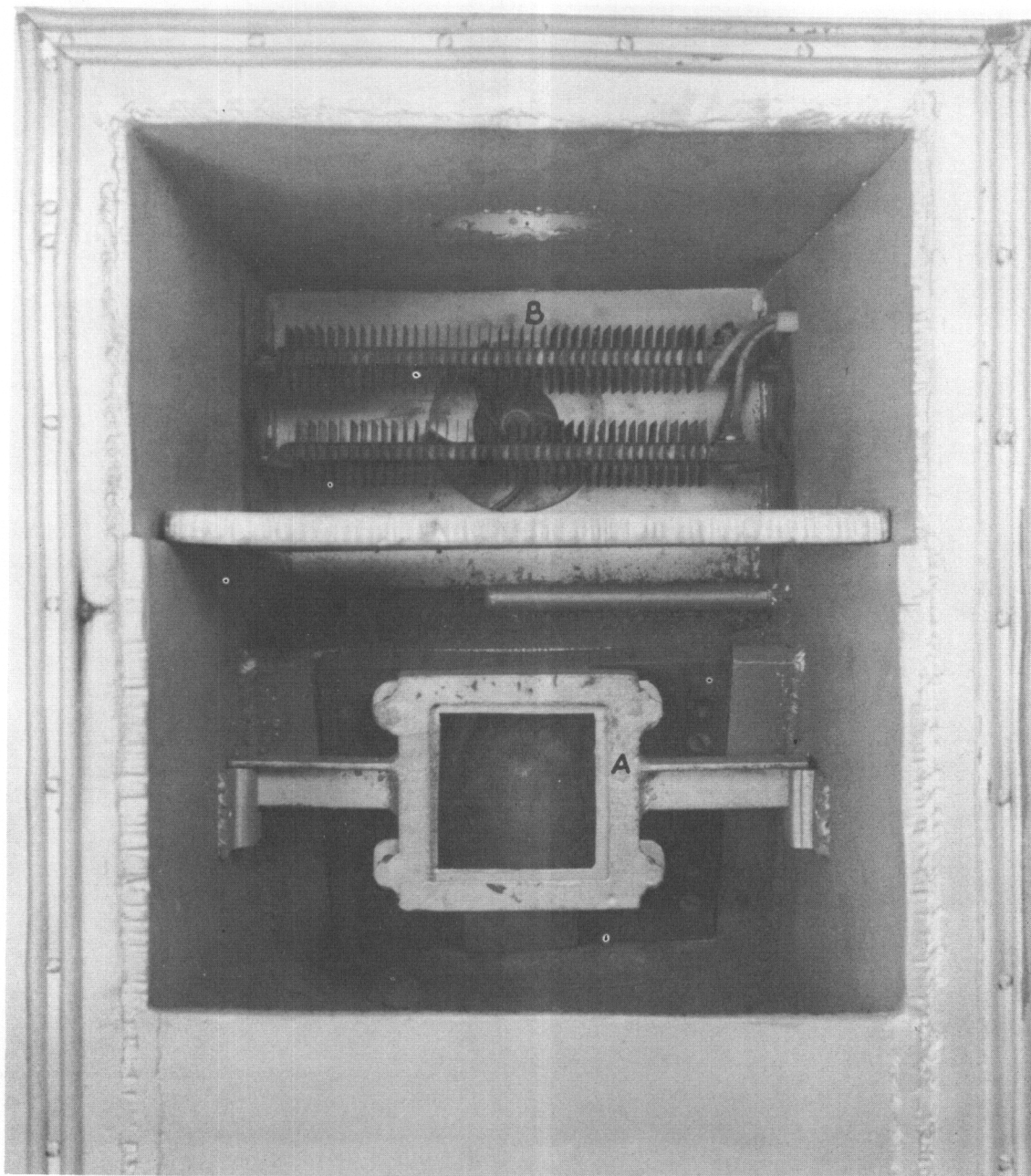


Figure 3

Gun Trap Interior Showing Laminate Rack Positioned for Bullet Impact

Gun Trap Interior Showing Laminate Rack Positioned for Bullet Impact

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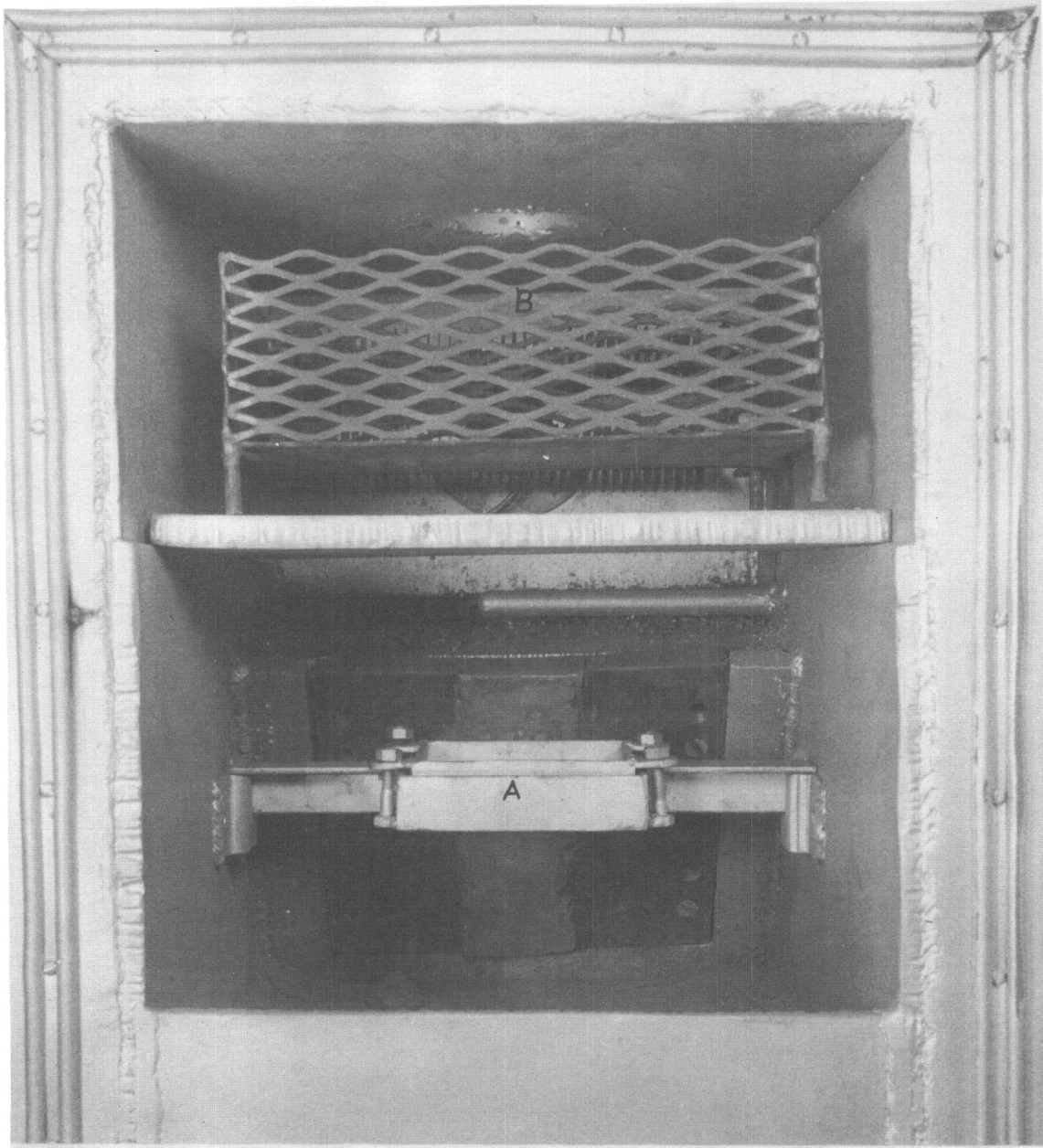


Figure 4

Gun Trap Interior Showing Laminate
Rack Positioned for Falling Ball Impact

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B. Optical Examination

The luminous transmittance and percent haze determinations were obtained as a service from the Dow Chemical Company Spectroscopy Laboratory. A General Electric Spectrophotometer was used for the optical measurements with I.C.I. illuminant C according to Federal Specification L-P 406b Method No. 3022.

C. High Velocity Impact

High velocity impact tests were made using a .22 caliber Hornet rifle described in Section I, Part IIB of this report. The test procedure was as follows: A four by four inch laminate was clamped in the laminate rack, A in Figure 3 and placed in position within the gun trap. The door was then closed and the high velocity projectile fired into the laminate. When test temperatures other than room temperature were used, a conditioning period of one hour at test temperature was used before the test was performed.

A method was developed to obtain a quantitative index of adhesion by comparing the entrance and exit craters of the experimental laminates with those of a glass-polyvinyl butyral laminate. The polyvinyl butyral standard was plasticized with 37.5 parts dibutyl sebacate and was one sixteenth inch in thickness. The entrance and exit craters for this laminate were 0.37 and 0.75 inches respectively. The crater diameters represented the area surrounding the perforation in the laminate that was completely void of glass. Figure 5 gives a sketch of a shot glass laminate. As adhesion was improved, more glass adhered to the interlayer and the crater diameters decreased. The exit crater values were considered more significant than the entrance craters.

D. Falling Ball Impact

A one half pound steel ball was used in this test. The gun trap was used for the falling ball test by placing the laminate rack in the position shown in Figure 4, letter A. The distance the ball fell was altered by adjusting the height of Bracket C in Figure 2. The laminates subjected to this falling ball test were qualitatively evaluated. The degree of delamination served as a qualitative measure of adhesion.

E. Tensile Test

Tensile tests at room temperature were performed with a Scott L-6 tensile testing machine using specimens cut with a standard rubber tensile die D. Procedure for test was followed as described in A.S.T.M. Designation D412-51T. This test method was used because the interlayer materials studied in this work were more like rubber than plastic.

The elevated temperature tensile properties were obtained using the Scott IP-4 tensile testing machine fitted with an air circulating oven. Samples were allowed 15 minutes to reach equilibrium at test temperature before testing. The load and elongation were plotted on a chart on the IP-4 machine.

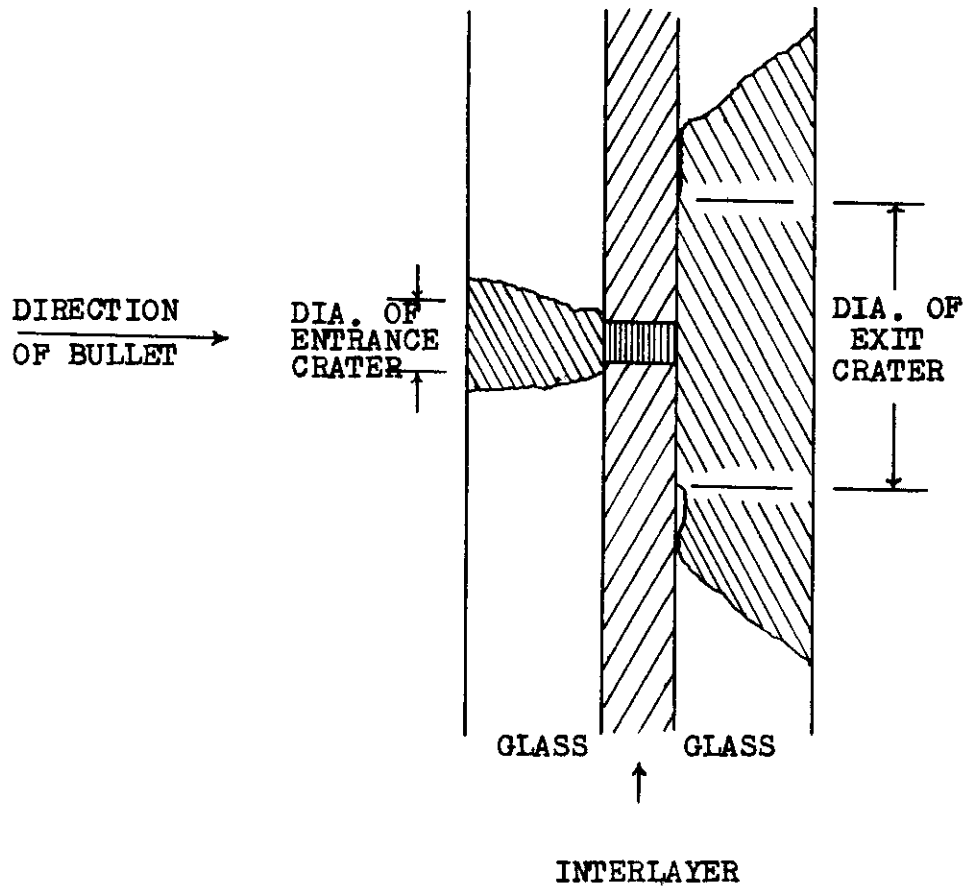


Figure 5

Sketch of Laminate Cross-section Showing Craters
Caused by High Velocity Impact of Bullet

F. Adhesion Test

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There were no specific methods prescribed for evaluating adhesion so the following three procedures were used in this work. The high velocity impact test gave an index of adhesion from the diameter of the entrance and exit craters. The delamination by falling ball impact gave a qualitative measure of adhesion. The third method used in this work for evaluating adhesion yielded a quantitative measure. This was accomplished by peeling strips of interlayer from the glass surface sheet at an angle of 90 degrees to the plane of the glass with the Scott L-6 tester. The test heads of the testing machine separated at a rate of 20 inches per minute. The adhesive strength was expressed as pounds per inch width of interlayer. For example, if an interlayer sample gave 20 pounds per inch adhesion, this would mean that a 20 pound force would be required to peel a strip 1 inch wide from the glass surface.

FACTUAL DATA

Part I Type A Interlayer

A. Description of Type A

Type A interlayer was a high molecular weight cross-linked polysiloxane possessing a high degree of tack. Although the polymer was cross-linked, flow could be accomplished by the application of heat and pressure.

B. Fabrication of Type A

Fabrication of Type A utilized the fact that it could be forced to flow by the application of heat and pressure. A desired quantity of Type A was placed between two glass plates and the laminate was then formed in a press by the application of heat and pressure. A steel chase was used to insure the desired uniform thickness throughout the laminate. A Silastic gasket was also used to assist in keeping the interlayer restrained between the glass plates.

C. Discussion of Data on Type A

The general properties of Type A were not fully evaluated because thermal stability above 300°F could not be achieved with this interlayer. Eighteen hours at 300°F without bubbling was the most severe test that Type A withstood. The tensile strength was around 25 psi or less. Many of the laminates prepared showed poor adhesion. The laminates were fairly good optically.

Typical data on laminates prepared using Type A interlayer are given in Table 1. A Type A laminate after several hours aging at 300°F is shown in Figure 6. The large bubbles seen in this laminate are typical of this interlayer material.

Table 1
Type A Laminates

<u>Reference Number</u>	<u>Interlayer Formation Condition</u>	<u>Heat Exposure</u>	<u>Interlayer Thickness in Inches</u>	<u>Special Remarks</u>
1. 1a	300°F, 2500 lb. press 5 min.	a) 77°F, 1 hr.	1/4	Bubbled. Edges of interlayer were sticky.
2. 4b	"	a) 300°F, 18 hrs.	1/4	No bubbling. Interlayer slightly hazy.
3. 5a	"	b) 480°F, 1 hr.		Bubbled.
4. 2b	"	a) 300°F, 18 hrs.	1/4	No bubbles, some delamination.
5. 2c	"	a) 77°F, 1-1/2 hrs. b) 300°F, 15 min. c) 392°F, 1-1/2 hrs.	1/4	Some delamination. Repressed to secure better adhesion. Large bubbles.
6. 6g	392°F, 2500 lbs. pressure 15 minutes.	a) 392°F, 1 hr.	1/4	Severe bubbling and flowing of interlayer.
7. 7f	"	392°F, 18 hrs.	1/8	Flowed on release
			1/16	Bubbled. Preheated 6 hrs. at 480°F.

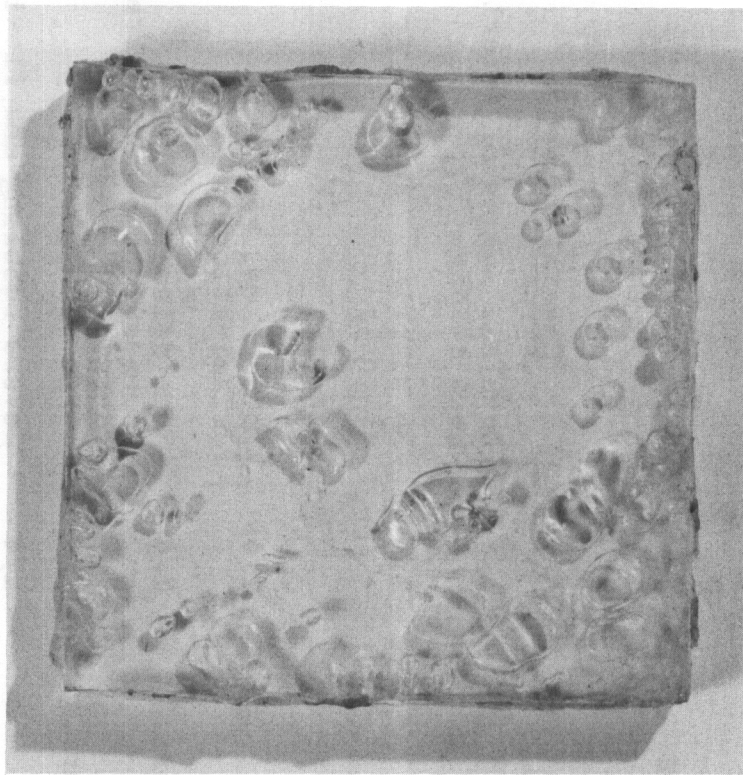


Figure 6

Typical Type A Laminate After
Several Hours at 300°F

Part II Type B Interlayer

A. Description of Type B

Type B interlayer was a polysiloxane which at the stage of preparing a laminate had a viscosity in the range of 10,000 poises.

B. Fabrication of Type B

The fabrication of Type B was complicated by its moderate viscosity coupled with the extended time at very high temperatures required to set the interlayer. When thin films were desired, laminates were fabricated by placing an adequate quantity of Type B between two glass plates, then pressing by hand until the desired film thickness was obtained. Silastic pressure sensitive tape (made from Dow Corning Corporation adhesives, XC-269 or XC-271) was wrapped around the edge of the laminate to confine the interlayer while it was being cured at elevated temperatures. For laminates with interlayers one sixteenth inches or greater in thickness, a chase and Silastic gasket was found suitable to obtain the desired uniform thickness and at the same time prevent run-off of the interlayer. Also, desirable preliminary setting was accomplished by heating at 392°F for 15 minutes or longer in a press under pressure. The laminates did not flow nearly as much after preliminary setting which greatly simplified the wrapping of the laminates with Silastic pressure sensitive tape to prepare them for further cure.

C. Discussion of Data on Type B

The physical properties of Type B interlayer were encouraging from the aspect of thermal stability. Sample 8b in Table 2 demonstrated the excellent thermal stability by withstanding 71 hours aging at 480°F without becoming discolored or developing bubbles. A Type B laminate which was cured 66 hours at 392°F, gave 82.8% transmission and 3.9% haze. This laminate had an interlayer thickness of one-sixteenth inches. A big physical drawback to Type B interlayer was its tendency to be brittle which led to poor shatter-resistance. For example, a laminate was shattered when a one-half pound steel ball was dropped four feet on the laminate. In another test, upon impact of a high velocity bullet (2800 ft/sec.) the laminate disintegrated.

Table 2 gives data on a number of Type B laminates. Laminate 3d, which had been cured 17 hours at 480°F, is shown in Figure 7. The excellent optical properties of this laminate may be seen.

Table 2
Type B Laminates

Reference Number B-126-2-	Interlayer Formation Condition	Heat Exposure	Interlayer Thickness in Inches	Special Remarks
1. 3a	Hand pressed between glass plates	a) 480°F 17-1/2 hrs.	Thin Film	Clear, Tough, excellent adhesion, no bubbles.
2. 3b	"	a) 480°F 23 hrs.	1/8	No bubbles. Interlayer firm, brittle.
3. 3c	"	a) 392°F 22 hrs.	1/8	Clear, but crumbly.
4. 3d	"	a) 480°F 17 hrs.	Thin Film	Clear, no bubbles, excellent adhesion. A duplicate plate broke into 5 large pieces under the impact of a 155 gm. weight dropped 4 feet.
5. 6b	392°F, 2500 lb. pressure 15 minutes	480°F 21 hrs.	1/8	Yellow and hazy. No bubbles.
6. 6c	"	480°F 23-1/2 hrs.	1/8	No bubbles. Some delamination
7. 6d	"	480°F 51 hrs.	1/8	Outer edge darkened.
8. 6e	"	392°F 64 hrs.	1/8	Slightly yellow. Clear. No bubbles. Good adhesion.
9. 7a	"	392°F 16 hrs.	1/8	Clear.
10. 7d	"	392°F 16 hrs.	1/8	No bubbles. Clear
11. 7g	392°F, 2500 lb. pressure 25 minutes	a) 392°F 66 hrs. b) 480°F 47 hrs.	1/16	Clear. No bubbles Clear. No bubbles Interlayer brittle.
12. 8b	392°F, 2500 lb. pressure 20 minutes	a) 392°F 24 hrs. b) 480°F 71 hrs.	1/16	Clear. No bubbles. Brittle Clear. No bubbles. Preheated 15 minutes at 300°F.

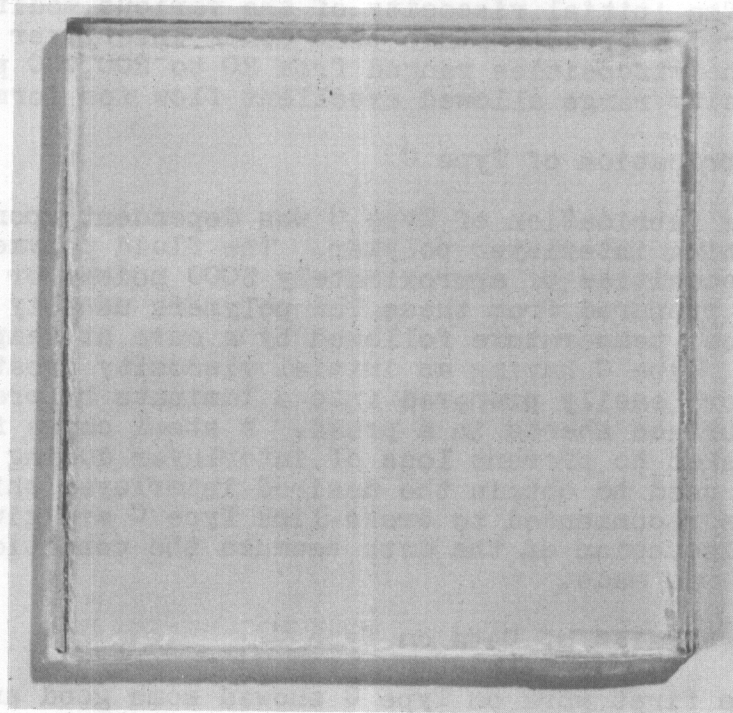


Figure 7

Type B Laminate After
17 Hours at 480°F

Part III Type C Interlayer

A. Description of Type C

Type C interlayer material was primarily a linear polysiloxane. The initial viscosity of the various modifications of Type C covered a broader range than other interlayer materials studied. The viscosities ranged from 20 to 200,000 poises. This whole viscosity range allowed excellent flow for forming laminates.

B. Fabrication of Type C

The fabrication of Type C was dependent upon the viscosity of the starting interlayer polymer. The fluid polymer chase worked well for viscosities of approximately 5000 poises or less. The interlayers prepared from these low polymers usually were partially set up at room temperature followed by a cure at temperatures of 300°F or greater. Type C having an initial viscosity greater than 5000 poise was more easily prepared into a laminate by pressing between two glass surface sheets in a press. A steel chase fitted with a Silastic gasket to prevent loss of interlayer during the setting up process was used to obtain the desired interlayer thickness. The temperatures recommended to cross-link Type C are given in the following discussion of the data because the conditions were altered as progress was made.

C. Discussion of Data on Type C

The first work on Type C showed some good and some poor possibilities for this interlayer. These results are given in Table 3. The results indicated that it was possible to prepare a laminate which had good adhesion, good optical properties, and good thermal stability for at least 48 hours at 480°F. This was true if a low viscosity polymer was used. However, the resulting interlayers of these early samples were much too thin. (In particular, note samples 1 and 2 in Table 3).

The results of higher viscosity polymers were not as encouraging. Numbers 3 through 6 in Table 3 demonstrate the typical early results for this higher viscosity Type C. The samples exhibited good optical properties, but the thermal stability and adhesion were both poor.

Numbers 7 and 8 in Table 3 were attempts to lessen bubbling by adding stabilizing ingredients to Type C. Higher laminating temperatures were also employed. No improvement of Type C was achieved in these trials.

Table 3

Type C Laminates

<u>Reference Number</u>	<u>Interlayer Formation Condition</u>	<u>Heat Exposure</u>	<u>Interlayer Thickness in inches</u>	<u>Comments</u>
1. 123-4b	Hand pressed between glass plates	480°F, 48 hrs.	Thin	Good adhesion. Optically good
2. 123-10B	Hand pressed between glass plates	392°F, 2 hrs. 480°F, 4 hrs.	Thin	No bubbling. Run-off bad No bubbling.
3. 123-5a	350°F, 2500 lb. pressure 15 minutes	300°F, 1 hr.	1/4	Bubbled. Adhesion poor, some haze.
4. 123-5A	300°F, 2500 lb. pressure 15 minutes	300°F, 1 hr. 480°F, 15 min.	1/4	Bubbled. Bubbled badly.
5. 123-6C	" "	300°F, 1 hr.	1/4	Bubbled.
6. 123-7A	" "	300°F, 15 min.	1/8	Bubbled. Preheated 3 hours at 300°F.
7. 123-10c	392°F, 2500 lb. pressure 30 minutes	392°F, 5 min.	1/8	Preheated 6 hrs. at 300°F. Bubbled badly.
8. 123-10d	" "	480°F, 1 hr.	1/8	Preheated 6 hrs. Bubbled badly.

Continued

An investigation was carried out to study the effect of changing the degree of cross-linking in Type C. This was done by comparing two analogous series. One of the series contained an initial polymer of lower viscosity than the other series. Each sample was given a cure of 120 hours at room temperature followed by 192 hours at 300°F. The cures were made in open aluminum dishes. At the end of the heat treatment, all of the samples of both series had gelled. The gels were perfectly clear, colorless, and bubble free. The adhesion of each gel to the dish was excellent. It was further noted that the hardness of the gels increased with the amount of crosslinks to a point at which the sample became cheesy. In this way the optimum amount of cross-linking was determined. The samples prepared from the higher viscosity material gave tougher gels than those from the lower viscosity polymer. This investigation yielded valuable groundwork for the further development of Type C.

The effect of initial gelling conditions of Type C on the final laminate when using low viscosity polymer is given in Table 4.

It was found that room temperature gelation yielded a more stable laminate than if heat was used to hasten the initial gelation. Sample 1 in Table 4 was subjected to the high velocity impact test and exhibited encouraging results. The entrance and exit craters were 0.5 and 0.63 inches respectively.

Up to this point the work showed that the major difficulty with Type C was its tendency to bubble between 320° and 356°F. This bubbling was due mainly to entrapped air and reaction products. From the study of gelation rates it was found that for the lower molecular weight polymers, initial gelation at room temperature was the most successful. On the other hand, for the higher molecular weight polymers, it was found to be better to use 30 minutes at 300°F as the initial gelation conditions. Another approach to eliminating bubbles was to evacuate the polysiloxane before fabrication of the laminate. This method was tried with slight success. Table 5 gives some of the typical results obtained during the course of this study.

During the seventh and eighth months of work, Type C was modified to yield laminates which possessed stability above 400°F. This latest version of Type C lended itself to fabrication better than some of previous Type C material because of its high viscosity. The fabrication method for this new version of Type C was to first place the proper amount of interlayer between glass surface sheets. These were then placed in a chase fitted with a Silastic gasket and put in a press at 194°F under 2000 psi pressure for one hour. The temperature was increased to 266°F for one hour and then to 320°F for two hours after which it was released cold. The laminates were then placed directly into a 400°F oven without fear of bubbling, delamination, or discoloration. One such sample was subjected to 480°F for 38 hours without visible change.

Table 4

Effect of Initial Gelling
Conditions of Type C

<u>Reference Number</u>	<u>Gelling Conditions Prior to Laminating</u>	<u>Laminating Conditions</u>	<u>Interlayer thickness in inches</u>	<u>Cure</u>	<u>Comments</u>
1. 126-11-12A	Room Temp. 72 hrs.	Pressed at 80°F	1/32	300°F 48 hrs. No bubbles; firm	Entrance Crater 0.5" Exit Crater 0.63"
2. 126-11-12B	212°F 16 hrs.	Pressed at 80°F	1/8	300°F 24 hrs. Bubbled	None
3. 126-11-12D	300°F 2 hrs.	Pressed at 80°F	1/8	300°F 16 hrs. Bubbled	None
4. 126-11-12E	(See Col. 3)	Poured between glass plates in fluid polymer chase 230°F - 18 hrs. 300°F - 144 hrs.	1/8	(See Columns 3 and 6)	Center portion of interlayer not gelled. Entire laminate destroyed.
5. 126-11-13B	Preheated 149°C	Poured between glass plates in fluid polymer chase. 212°F - 2 hrs. Heated slowly to 392°F. Maintained 19 hrs.		(See Column 3) 5 large bubbles after 392°F cure	None

Table 5

Type C Laminates

<u>Reference Number</u>	<u>Laminating Conditions</u>	<u>Heat Exposure</u>	<u>Interlayer Thickness in Inches</u>	<u>Special Remarks</u>
1. 123-27B	Room temp., 1500 lbs. pressure., Gelled room temp. 72 hrs.	300°F, 1 hr.	1/8	Entrapped air bubbles. Adhesion good, strength fair
2. 123-30A	300°F, 1500 lbs. pressure, 30 min.	300°F, 2 hrs. 392°F, 1 hr. 392°F, 18 hrs.	1/8	No bubbling. Many small bubbles. No increase in bubbling. Good adhesion. Clear.
3. 123-30C	Room temp., 1500 lbs. pressure	None	1/8	Entrapped air bubbles.
4. 123-31B	300°F, 1500 lbs. pressure, 25 min.	300°F, 48 hrs. 392°F, 18 hrs.	1/8	No bubbling. Partially gelled.
5. 123-31C	Room temp., 1000 lbs. pressure	None	1/8	Several large bubbles. Good adhesion. Evacuated polymer 10mm pressure, 2 hrs. before forming laminate
6. 123-32A	Room temp., 1000 lbs. pressure Gelled room temp., 18 hrs.	None	1/8	Entrapped air bubbles. Evacuated interlayer material 5mm press., 3 hrs.
7. 123-33A	257°F, 1500 lbs. pressure, 5 min.	300°F, 24 hrs.	1/16	Good adhesion, fair strength
8. 123-33B	257°F, 1500 lbs. pressure, 5 min.	300°F, 72 hrs.	1/16	Bubbled badly. Interior of laminates depolymerized
9. 123-33C	257°F, 1500 lbs. pressure, 5 min.	300°F, 48 hrs.	1/16	Entrapped air bubbles. No further bubbling after heating. Adhesion fair.

A series of moldings of Type C was made to observe the change in tensile strength with time and temperature. Because of the adhering power of this interlayer, it was extremely difficult to prepare samples suitable for cutting into tensile test bars. Up to two days were required for the production of any one sample which could be released sufficiently well from the molding plates for testing purposes. Table 6 contains typical examples of these results, all of which fell within the same range.

Table 6
Tensile Strength of Type C

<u>Cure</u>	<u>Tensile Strength psi</u>	<u>Elongation %</u>
300°F, 10 min.	28	415
392°F, 18 hrs.	22	315
392°F, 48 hrs.	21	280

Part IV Type D Interlayer

A. Description of Type D

Type D interlayer material was a linear polysiloxane. The viscosity of this interlayer prior to laminating was approximately 100,000 poises and thus required pressure to create flow. The cross-linking of Type D was accomplished at elevated temperatures.

B. Fabrication of Type D

A portion of Type D was placed between two glass surface sheets and pressed to the desired thickness with the aid of a press and chase. An oven cure at temperatures up to 480°F was required to permanently set the interlayer.

C. Discussion of Data on Type D

The first laminate prepared from Type D was a one inch square hand pressed laminate. The interlayer in this laminate was a thin film. This laminate was subjected to 480°F for 48 hours without bubbling. The optical properties were good, but the adhesion was only fair.

Type D presented a problem in adhesion, so several additives were tried to improve this characteristic. These additives caused the polymer to have less heat stability. The laminates bubbled at 300°F and darkened on further cure.

A Type D laminate is shown in Figure 8 with several small bubbles visible.

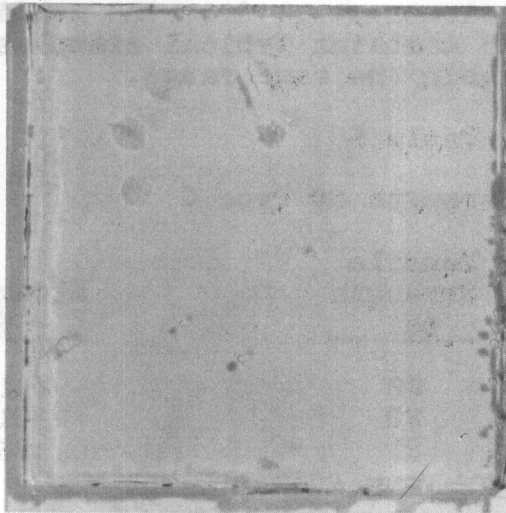


Figure 8

Type D Laminate

These problems of insufficient adhesion, tendency to bubble and darken at high temperatures, and the requirement of high temperatures to set the interlayer discouraged further work on Type D.

Part V Type E Interlayer

A. Description of Type E

Type E interlayer is a linear polysiloxane. During the stage of forming a laminate this interlayer had a viscosity in the range of 100,000 to 200,000 poises. Three hundred degrees Fahrenheit were required to crosslink Type E.

B. Fabrication of Type E

Pressure and heat greatly aided in the fabrication of Type E. This meant that a press with heated platens was well suited for this interlayer in the same manner as for some of the previously described high viscosity linear polysiloxanes such as Type C and Type D. This involved placing the desired amount of Type E between two glass surface sheets. These were placed within a steel chase fitted with a Silastic gasket to obtain the correct laminate thickness, and then pressure and heat were applied. At least 10 minutes at 300°F were used in setting the interlayer.

C. Discussion of Data on Type E

The first laminate prepared using Type E gave very discouraging results. The laminate was pressed at 2500 psi pressure for 15 minutes at 300°F. The laminate was heated 1 hour at 392°F causing slight bubbling. The laminate at this point was hazy and had poor adhesion. It was then heated for 1 hour at 480°F. This extreme heating caused severe bubbling and an increase in haze. This particular sample is shown in Figure 9 and is listed in Table 7 as sample number one. Sample number two in Table 7 had the glass surface sheets coated with a titanate to improve adhesion. The titanate caused additional discoloration with no apparent improvement in adhesion.

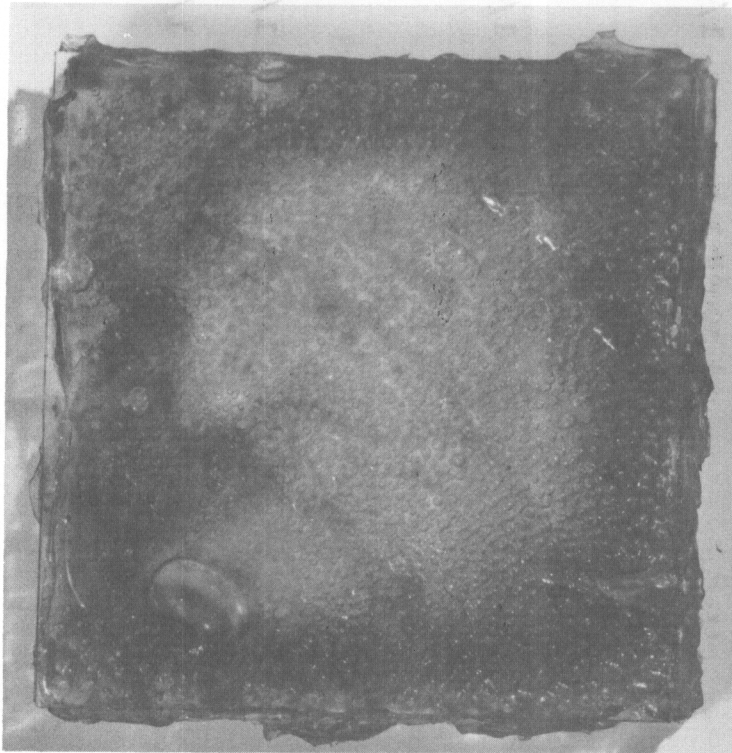


Figure 9

First Type E Laminate

Table 7

Type E Laminates

<u>Reference Number</u>	<u>Interlayer Formation Condition</u>	<u>Heat Exposure</u>	<u>Interlayer Thickness in Inches</u>	<u>Special Remarks</u>
1. 123-2a	300°F, 2500 pressure 15 minutes	a) 392°F 1 hr. b) 480°F 1 hr.	1/4	Slight bubbling. Hazy poor adhesion. Slight increase in haze. Severe bubbling. (See Fig. 9)
2. 123-3a	" "	a) 300°F 1 hr. b) 392°F 1 hr.	1/4	Coated glass with titanate adhesive. Some haze. Fair adhesion. Adhesive oxidized on heating, discoloring laminate. Delaminated.
3. 123-4D	" "	300°F 1 hr. 480°F 1 hr. 480°F 18 hrs.	1/8	No bubbling. Pre-heated 5 hours at 300°F. No bubbling. Slight haze. Poor Adhesion. Bubbled badly.
4. 123-6B	" "	300°F 1 hr. 392°F 1 hr.	1/8	No bubbling. Pre-heated 4 hours at 300°F. No bubbling. Slight increase in haze.
5. 123-7C	" "	300°F 1 hr. 392°F 1 hr. 437°F 1 hr. 437°F 72 hrs.	1/8	No bubbling. Pre-heated 5 hours at 300°F. No bubbling. Slight increase in haze. Severe bubbling.
6. 123-8A	" "	480°F 72 hrs.	1/8	Swelled and bubbled. Pre-heated 4 hours at 300°F.

Table 7 Continued

<u>Reference Number</u>	<u>Interlayer Formation Condition</u>	<u>Heat Exposure</u>	<u>Interlayer Thickness in Inches</u>	<u>Special Remarks</u>
7. 123-9A	300°F, 2500 lb. pressure 15 minutes	300°F 2 hrs. 392°F 2 hrs. 480°F 1 hr.	1/8	No bubbling. Pre-heated 5 hours at 300°F. No bubbling. No bubbling. Slight discoloration.
8. 126-II-10a	374°-392°C, 400 lb. Pressure, 15 minutes	480°F 16 hrs.	1/8	Preheated 2 hrs. at 480°F Interlayer split and delaminated due to overcure.
9. 126-II-10b	392°F, 2500 lb. pressure 10 minutes	None (poor adhesion on release)	1/8	Preheated 1 hr. at 480°F
10. 126-II-10c	392°F, 2500 lb. pressure 10 minutes	480°F 2 hrs.	1/8	Preheated 15 min. at 480°F. Poor adhesion. Bubbled.
11. 126-II-8d	Hand pressed 77°F	480°F 18 hrs.	1/16	Bubbled.
12. 126-II-9a	374°-392°F, 2500 lb. pressure, 15 minutes	392°F 2-1/2 hrs.	1/8	Clear. Some bubbling. Material devolatilized 16 hrs. before formation of laminate.
13. 126-II-9b	Cold pressed, 4000 lb. pressure, 10 minutes (Poor Adhesion) Repressed at 2500 lb. 383°F. 5 minutes	480°F 6 hrs.	1/16	Bubbled and flowed.

Contrails

It was found that the interlayer thickness could be decreased from one quarter inch to one eighth inch without harming the shatter resistance and at the same time making an improvement in the optical properties. Sample 123-6B having a cure of 1 hour at 300°F plus 1 hour at 392°F and a thickness of one quarter inch gave 58.2% transmission and 24.2% haze. Sample 123-9H having a cure of 2 hours at 300°F plus 2 hours at 392°F and a thickness of one eighth inch gave 69.0% transmission and 17.5% haze.

Preheating Type E at 300°F for three to five hours improved the thermal stability of the laminates. Samples 3 through 7 in Table 7 represent this work.

Three series of experiments were conducted in order to improve this material. Numbers 8-10 of Table 7 were precured for various time intervals in order to remove any low molecular weight volatiles. In each case the precure caused the interlayer to set up to a point where it could no longer be easily pressed or adhered to the glass.

Numbers 11-13 of Table 7 represent changes in formulation designed to increase the stability. All three resulted in excessive bubbling above 392°F.

In Table 8 an attempt was made to determine the feasibility of thermally stabilizing Type E interlayer. Four basic formulations were used. The material was molded at 300°F, and a strip was heated at 480°F to observe changes in stability. The stability of the strips of interlayer increased down the table but haze also increased.

Table 8
Stabilization of Type E

	<u>Sample</u>	<u>Heat Exposure</u>	<u>Remarks</u>
1.	126-I-6b	480°F 8 hrs.	Bubbled
2.	126-I-5d	480°F 50 hrs.	Bubbled
3.	126-I-6c	480°F 11 days	Decomposed
4.	126-I-5b	480°F 32 hrs.	No bubbles, Material hazy

Samples 1 and 2 in Table 9 demonstrated that Type E withstood varied continuous heating for better than 500 hours at temperatures up to 437°F. Slight delamination occurred after 3 to 4 days at 300°F. A slight increase in haze developed after 3 to 4 days when heated in the range of 338° to 356°F. The laminates withstood 203 hours at 374° and 392°F with no ill effects. Depolymerization after 72 hours at 437°F was noted.

Adhesion of Type E was constantly a problem in the work discussed thus far. Several glass treatments, samples 3 through 6 in Table 9, were tried to improve adhesion. Comparisons were based on the dimensions of craters formed by the high velocity bullet test. At this stage of development the adhesion of Type E was not as good as that of polyvinyl butyral laminate of equal thickness on the basis of high speed projectile results at room temperature. The entrance

Contrails

crater for Type E was 1.5 inches in diameter against 0.37 inches for polyvinyl butyral while the exit crater was 1.75 inches in diameter against 0.75 inches for polyvinyl butyral. A photograph, Figure 10, was taken of the exit crater of sample 3 Table 9.

Further studies were carried out to improve the adhesion of Type E. Two methods of approach were used to improve adhesion. The glass surface sheets were treated and the results are listed in Table 10. The glass treatments were performed by preparing solutions in isopropanol of ethyl silicate, methylhydrogendichlorosilane, methyltrichlorosilane, trimethylchlorosilane, and vinyltrichlorosilane in varying strengths. The glass was first washed with isopropyl alcohol to remove foreign matter, then dipped in a solution of one of the above silicone compounds. The glass was allowed to dry at room temperature and then baked at a specified temperature and time. Laminates were then prepared with this treated glass by pressing at 2500 psi pressure at 300°F for 10 minutes. The glass was intentionally cracked and a cracked portion was removed to note adhesion. No success was attained using this approach, as all specimens separated readily by hand.

Adhesion comparisons were also made after several additives had been incorporated in Type E. These additives were two experimental silicone compounds and tertiary butyl hydroperoxide. The laminates were prepared as other Type E laminates have previously been formed. The laminates were intentionally cracked as for glass-treated laminates. The glass was separated from the interlayer by hand, noting the adhesion. The additives used in this series did not improve the adhesion. The results are given in Table 11.

In continuation of the silane treatment of glass, silicone-pressure-sensitive adhesives XC-269 and XC-271 were used to coat the glass. The glass was cleaned with organic solvents, dipped in the adhesive, and baked. No success was attained using this method. These silicone adhesives were added to the Type E polymer and good adhesion resulted. However, severe bubbling resulted at elevated temperatures. (See sample 2, Table 12).

Some success in improving the adhesion was obtained by the addition of 0.5 parts of ethyl silicate to the Type E interlayer. In order to complete the adhesive action of ethyl silicate, a cure of 18 hours at 392°F was necessary. Bullet impact crater dimensions were reduced, indicating better adhesion. The room temperature falling-ball test likewise showed encouraging results. (See samples 4 and 5, Table 12; and Figures 11 and 12).

It is of interest to compare Figure 11 with the former Figure 10 to see the improvement in adhesion by the addition of ethyl silicate to Type E. Polyvinyl butyral samples are also included for comparison.

The pre-heat treatment of Type E polymer improved the thermal stability. However, the preheated polymer and ethyl silicate laminates were only about 70 percent reproducible as they occasionally bubbled after 18 to 20 hours at 392°F.

Table 9
Type E Laminates

<u>Reference Number</u>	<u>Interlayer Formation Condition</u>	<u>Heat Exposure</u>	<u>Interlayer Thickness in Inches</u>	<u>Special Remarks</u>
1. 123-16C	300°F, 2500lbs. pressure 10 minutes	300°F-120 hrs. 338°F- 50 hrs. 356°F-100 hrs. 374°F-103 hrs. 392°F-100 hrs. 437°F- 72 hrs. <u>Total 545 hrs.</u>	1/8	Slight delamination Slight haze increase Beginning to bubble
2. 123-17A	" "	248°F- 72 hrs. 320°F- 24 hrs. 338°F- 50 hrs. 356°F-100 hrs. 374°F-103 hrs. 392°F-100 hrs. 437°F- 72 hrs. <u>Total 521 hrs.</u>	1/8	Slight delamination Slight haze increase Beginning to bubble
3. 123-3B	" "	none	1/4	Bullet crater diameter Entrance- 1.5 inches Exit - 1.5 inches (See Fig. 10)
4. 123-21B	" "	none	1/8	Treated glass with ethyl silicate. Heated glass 300°F, 2 hrs. Bullet crater diameter Entrance- 1.5 inches Exit - 1.75 inches
5. 123-22A	" "	none	1/8	Treated glass with Dow Corning 1107 and baked glass 300°F, 2 hrs. Bullet crater diameter. Entrance- 1.5 inches Exit - 1.75 inches
6. 123-22B	" "	none	1/8	Treated glass with experimental resin and baked glass 300°F, 2 hrs. Bullet crater Entrance- 1.5 inches Exit - 1.75 inches

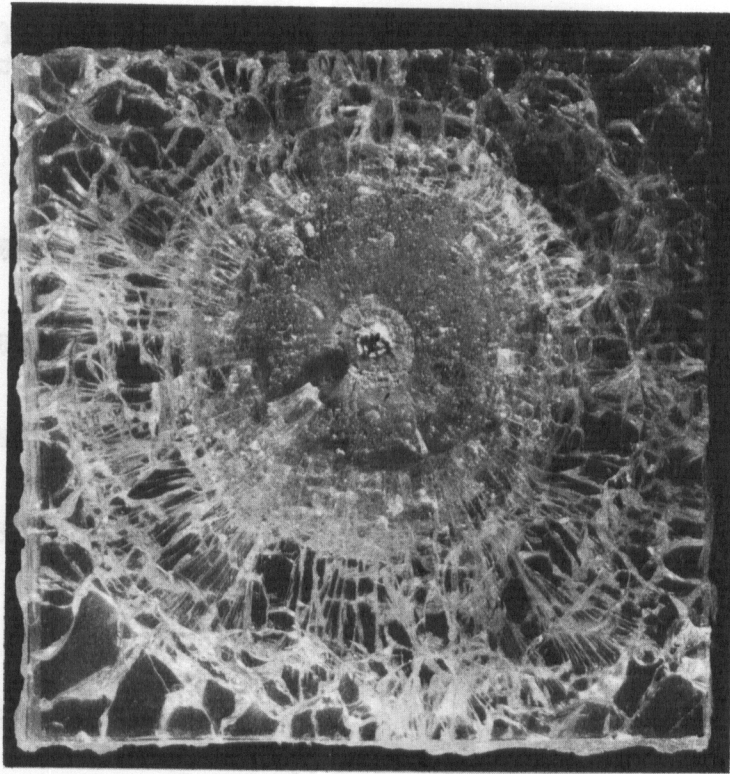


Figure 10

Exit Crater of Type E

Laminate No. 3 in Table 9

Contracts
Table 10

Glass Treatment to Improve Adhesion of Type E
Isopropanol used as Solvent in Solutions

<u>No. Sample</u>	<u>Glass Treatment and Exposure</u>	<u>Adhesion</u>
1. 123-22C	10% ethyl silicate Heated 2 hrs./480°F	Poor
2. 123-22D	1% MeHSiCl ₂ Heated 2 hrs./480°F	Poor
3. 123-23A	1% MeSiCl ₃ Heated 4 hrs./392°F	Poor
4. 123-23B	1% Me ₂ SiCl Heated 4 hrs./392°F	Poor
5. 123-24B	1% V ₁ SiCl ₃ 18 hrs./392°F	Poor
6. 123-25A	10% V ₁ SiCl ₃ Heated 4 hrs./392°F	Poor
7. 123-25D	10% MeSiCl ₃ Heated 18 hrs./392°F	Poor

Table 11

Adhesion Tests of Type E Containing Additives

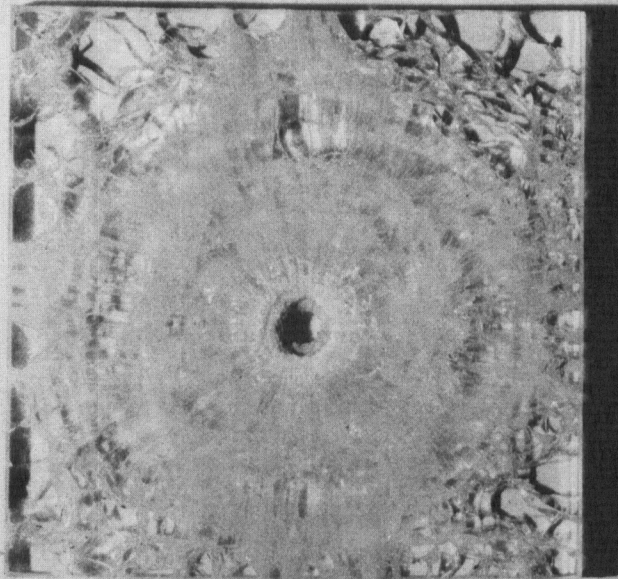
<u>No. Sample</u>	<u>Additive to Type E</u>	<u>Adhesion</u>
1. 123-24D	2 pts. experimental silicone No. 1	Poor
2. 123-25B	15 pts. tertiary butyl hydroperoxide	Poor
3. 123-25C	5 pts. experimental silicone No. 2	Poor
4. 123-26D	50 pts. experimental silicone No. 2 plus 50 pts. experimental silicone No. 1	Poor
5. 123-27A	10 pts. experimental silicone No. 2 plus 10 pts. experimental silicone No. 1	Poor

Table 12

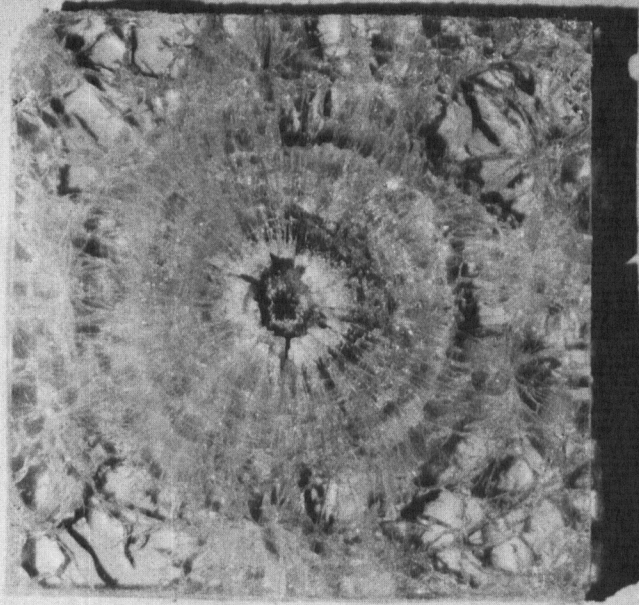
Type E Laminates

<u>Reference Number</u>	<u>Laminating Conditions</u>	<u>Heat Exposure</u>	<u>Interlayer Thickness in Inches</u>	<u>Special Remarks</u>
1. 123-28B	300°F, 1500 lbs. pressure, 10 min.	300°F, 2 hrs.	1/8	Coated glass with XC-271 adhesive. Slight improvement of adhesion.
2. 123-29A	300°F, 1500 lbs. pressure, 10 min.	300°F, 2 hrs. 392°F, 1 hr.	1/8	No bubbling. Bubbled badly. Adhesion good. Added adhesive, XC-271, to interlayer.
3. 123-33C	300°F, 1500 lbs. pressure, 30 min.	None	1/8	Treated glass with condensed polysiloxane. No improvement in adhesion.
4. 123-35A	300°F, 1500 lbs. pressure, 10 min.	392°F, 20 hrs.	1/16	Added 0.5 pt. ethyl silicate. Some improvement in adhesion. No bubbling (See falling-ball test, Figure 12).
5. 123-36A	300°F, 1500 lbs. pressure, 10 min.	392°F, 20 hrs.	1/16	Added 0.5 pt. ethyl silicate. Some improvement of adhesion. crater diam. Entrance - 0.5 in. Exit - 1.25 in. (See Figure 11)
6. 123-28C	300°F, 1500 lbs. pressure, 10 min.	300°F, 12 hrs.	1/8	Coated glass with XC-269 adhesive. Slight improvement in adhesion.

BULLET-IMPACT TEST



POLYVINYL BUTYRAL



TYPE E

Figure 11

High Velocity Impact Comparison of a Polyvinyl Butyral Laminate with Type E Laminate No. 5 Table 12

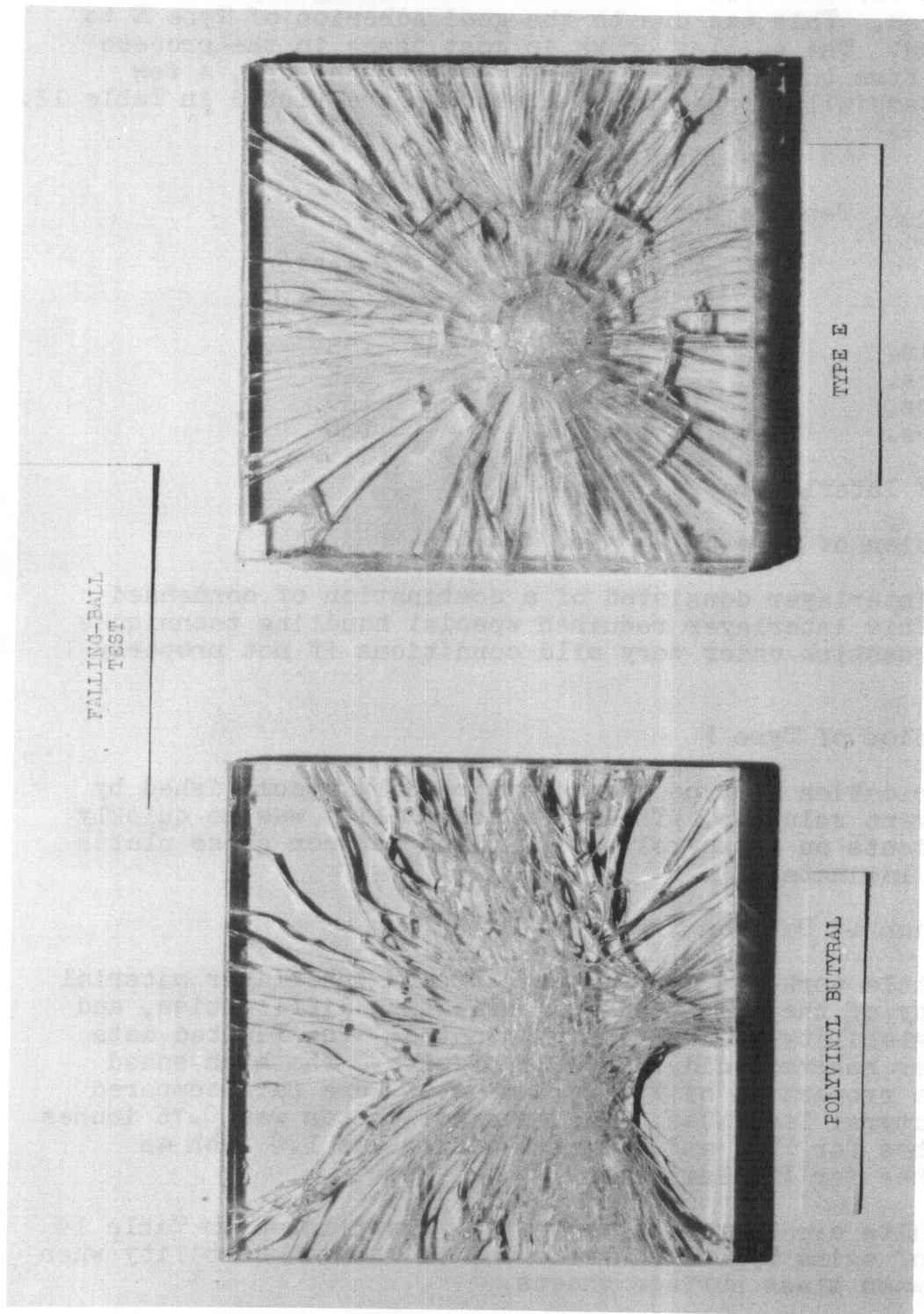


Figure 12
Falling Ball Impact Comparison of Polyvinyl Butyral Laminate with Type E Laminate No. 4 Table 12. Samples were tested with a one half pound steel ball dropped 5 feet at 80°F

Most of the target properties were approached quite well with Type E. One exception was the tensile strength. The preparation of Type E samples for the evaluation of tensile strength proved to be a difficult problem. This was due to the good adhesion of Type E to the molding plates. The samples broke in most cases in the process of removing them from the molding plates. After much work, a few samples were successfully prepared. The results are listed in Table 13.

Table 13

Tensile Strength of Type E

<u>Cure</u>	<u>Tensile psi</u>	<u>Elongation %</u>
300°F, 10 min.	26	175
300°F, 6 hrs.	26	185
392°F, 18 hrs.	17	160
392°F, 48 hrs.	17	160

Part VI Type F Interlayer

A. Description of Type F

Type F interlayer consisted of a combination of condensed polysiloxanes. This interlayer required special handling techniques since it would crosslink under very mild conditions if not properly controlled.

B. Fabrication of Type F

The fabrication of Type F was most readily accomplished by casting from solvent solution. The other possibility was to quickly blend the ingredients on a two-roll mill, place between glass plates and press into a laminate.

C. Discussion of Data on Type F

Very little work was done on this Type F interlayer material due to variability of the ingredients, fabrication difficulties, and lack of thermal stability of the final laminates. The limited data on this interlayer however, did show some promise. The high speed projectile impact properties of Type F laminates were fair compared with polyvinyl butyral laminates. The entrance crater was 0.75 inches against 0.37 inches for PVB, and the exit crater was 1.0 inch as against 0.75 inches for PVB laminates.

The results are listed in Table 14. Also listed in Table 14 are the results of aging Type F to evaluate its thermal stability when not confined between glass surface sheets.

Contrails

The result of a high velocity impact on a Type F laminate is shown in Figure 13.

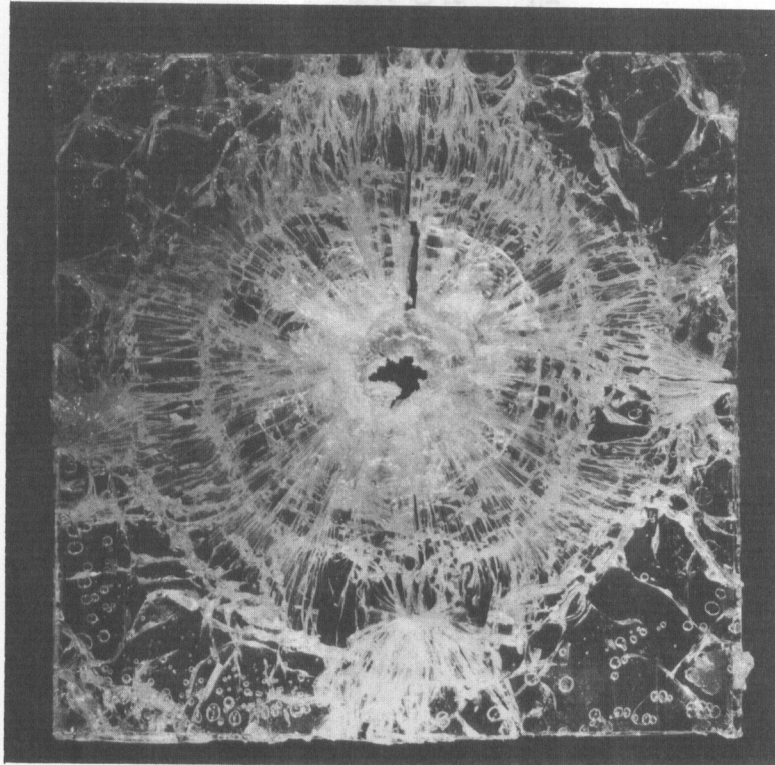


Figure 13

Type F Laminate Exit Crater
From High Velocity Impact

Contrails

Table 14

Type F Interlayer

<u>Reference Number</u>	<u>Interlayer Formation Condition</u>	<u>Heat Exposure</u>	<u>Interlayer Thickness</u>	<u>Special Remarks</u>
1. 123-18C	Hand pressed. Autoclaved 100 lbs. steam, 10 min.	none	Thin	Bullet crater diameter Entrance- 0.75 inches Exit - 1.00 inches Good optically, weak interlayer
2. 127-43C	Interlayer material dissolved in methylene chloride. Removed solvent, 24 hrs. at room temperature. Open glass sample	122°F, 24 hrs. 300°F, 24 hrs. 392°F, 72 hrs. 480°F, 24 hrs.		No effects No effects No effects No effects Fairly tough, elastic material. Adhesion to glass poor. Good optically.

Part VII Type G Interlayer

A. Description of Type G

Type G interlayer consisted of condensed polysiloxanes. At the laminate forming stage it was normally a rather viscous fluid. However, it could be partially gelled if desired by heating.

B. Fabrication of Type G

The fluid polymer phase worked ideally with Type G in preparing laminates. Specifically, this entailed pouring Type G between the two glass surface sheets and heating the assembly to gel the interlayer.

C. Discussion of Data on Type G

Listed in Table 15 are the results of several Type G laminates. The Type G interlayer seemed very tough at room temperature and possessed excellent optical properties. The major limiting characteristic was its tendency to be thermoplastic. This problem did not appear to have a suitable answer.

Figure 14 shows a Type G laminate after being subjected to high velocity impact. The exit crater is shown for sample number 4 in Table 15.

Table 15

Type G Interlayer

<u>Reference Number</u>	<u>Interlayer Formation Condition</u>	<u>Heat Exposure</u>	<u>Interlayer Thickness in Inches</u>	<u>Special Remarks</u>
1. 123-16B	Formed in beaker. Viscous fluid polysiloxane	392°F, 30 min. 392°F, 2 hrs. 480°F, 18 hrs.	---	Slight gel. Slight gel. Fluid. Thermoplastic at 300°F Tough at room temperature.
2. 123-17B	Mixed polysiloxane and catalyst in beaker. Poured in special chase. Heated 300°F, 2 hours, and 392°F, 18 hours	none	Thin	Run-off bad Optical properties good. Thermoplastic at 300°F. Tough
3. 123-18B	Same procedure as 123-17B	392°F, 18 hrs.	Thin	Slightly thermoplastic Yellowish tinge. Clear tough interlayer.
4. 123-19A	Allowed to gel in beaker 410°F, 5 hrs. Pressed gel between glass plates at 498°F, 10 minutes	none	1/8	Yellowish-tinge. Clear. Thermoplastic above 410°F. Bullet crater diameter Entrance- 1.25 inches Exit - 1.5 inches Air bubbles formed after 24 hours at room temperature
5. 123-20B	" "	300°F, 2 hrs.	1/8	No bubbling. Thermoplastic. Optically clear. Bullet crater diameter. Entrance- 1.18 inches Exit - 1.40 inches

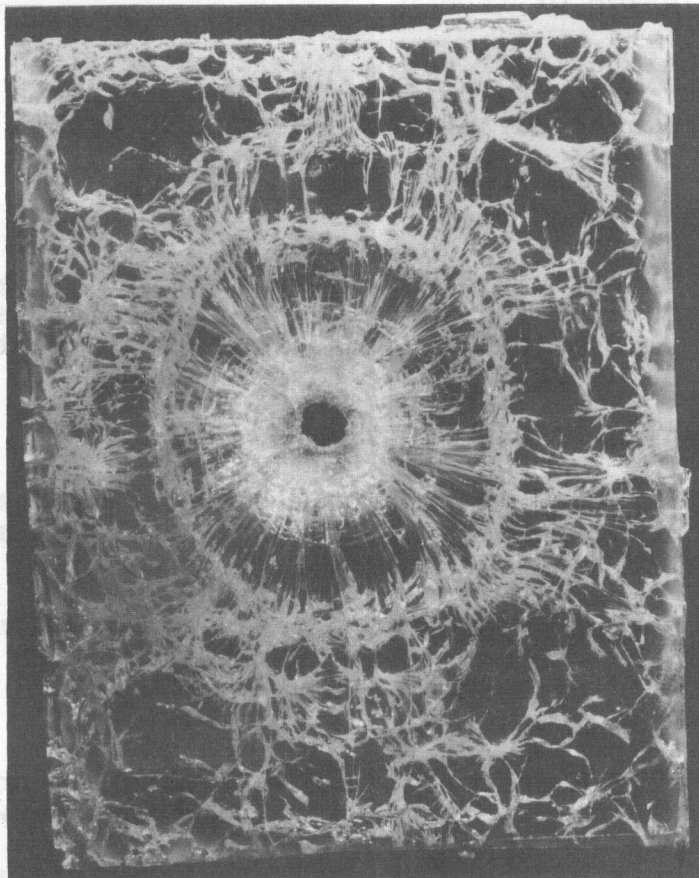


Figure 14

Type G Laminate Exit
Crater from High Velocity Impact

Part VIII Type H Interlayer

A. Description of Type H

Type H interlayer was a free flowing fluid of several hundred poises viscosity prior to being cross-linked into a suitable interlayer. Type H was introduced during the seventh and eighth months of contract work.

B. Fabrication of Type H

The fluid polymer phase described earlier was necessary to prepare laminates with Type H. The curing cycle used for Type H was slow, beginning at 194°F to obtain initial gelation followed by an increase of about 27°F per hour up to 392°F.

C. Discussion of Data on Type H

The first samples of Type H had good optical properties and adhesion, but on heating above 392°F exhibited a tendency to produce large cracks in the interlayer.

Further work eliminated the crack formation problem and also simplified the curing cycle. The curing cycle was simplified to 1 hour at 230°F followed by 1 hour at 300°F, and then finished with 16 hours at 400°F.

Haze and transmission data were obtained on laminate 126-II-22E. Percent transmission was 86.8. Percent haze was 1.8.

As in most of the previous type interlayers discussed, the problem of low tensile strength was present along with an additional problem of poor low temperature properties. An extensive investigation was carried out on variations of Type H in an effort to find improvement in these two property limitations. The findings indicated that it was impossible to obtain improvements in the low temperature characteristics of Type H and at the same time maintain adequate optical clarity. In addition, the tensile properties did not show improvement. In the light of these facts further investigation of Type H was discontinued.

Part IX Type J Interlayer

A. Description of Type J

During the ninth and tenth months of contract work a new interlayer material was introduced which possessed a combination of properties which were the most promising of the interlayers studied. This interlayer material was called Type J and basically consisted of a polysiloxane polymer filled with a treated silica.

Type J possessed tensile strength of several hundred pounds per square inch and thermal stability above 400°F. Because of its physical strength Type J offered for the first time the possibility of forming laminates using a tough, strong, free sheet of a polysiloxane interlayer material. This form would be compatible with current commercial fabrication techniques. Type J offered two question marks, one with regard to optical properties, the other with regard to adhesion.

B. Fabrication of Type J

The fabrication of Type J was altered during the progressive development of this interlayer. At the start of work with Type J, the preparation of laminates was identical with the technique used for Type E. Briefly, interlayer material was placed between glass surface sheets under heat and pressure while in a chase, and released cold after the interlayer set up. After tensile properties were improved, the fabrication was altered to the use of prefabricated sheets which could then be laminated between glass surface sheets with the use of suitable adhesives.

In most cases the adhesives were applied by brush to hot-chromic-acid-cleaned-glass surface sheets followed by a 10 minute heating at 122°F before forming the laminate. A ten minute press period at 300°F was used to adhere the surface sheets securely to the interlayer. The pressures used varied up to about 500 pounds per square inch.

C. Discussion of Data on Type J

In the initial stages of development of Type J, effort was concentrated on the improvement of tensile strength. Early Type J tensile strengths ranged from 400 to 600 pounds per square inch. The data presented in Table 16 show the general increase in tensile strength as improvements were made on Type J. Tensile values of 800 to 1100 psi after exposures of 24 hours at 480°F were not uncommon on the improved versions of Type J.

Table 16
Properties of Type J Laminates

Sample No.	Thermal Stability of laminate after 24 hrs.	Tensile Properties			Optical Properties		
		10 min./300°F psi	24 hrs./480°F psi	% El.	10 min./300°F % Trans.	24 hrs./480°F % Trans.	% Haze
1. 42B	- - - - -	---	---	---	Very Poor		
2. 41C	Delamination. No bubbles. 392°F.	655	290	100	80.8		7.2
3. 43A	- - - - -	---	---	---	Very Poor		
4. 45B	- - - - -	470	430	160	----		----
5. 46C	Delamination. Some increase in haze 480°F	---	---	---	77.7	10.3	----
6. 46A	- - - - -	396	316	200	----		----
7. 46B	Some increase in haze. Craze upon cooling. 480°F.	---	Poor thermal stability	----	----		----
8. 48B	- - - - -	475	400	100	79.0	13.1	
9. 49A	Delaminated. No bubbling. Little increase in haze. 480°F.	390	350	200			75.5
10. 52A	Delamination No bubbling. 480°F.	708	650	150			
11. 49B	- - - - -	---	---	---	80.2	10.8	
12. 52B	Slight brownish tinge. No bubbling. Delamination. 480°F.	708	640	180	----		----

Table 16 (cont'd.)

Sample No.	Thermal Stability of laminate after 24 hrs.	Tensile Properties				Optical Properties			
		10 min./300°F		24 hrs./480°F		10 min./300°F		24 hrs./480°F	
		psi	% El.	psi	% El.	% Trans.	% Haze	% Trans.	% Haze
13. 50B	- - - - -	-----	-----	-----	-----	69.8	15.8		
14. 50C	Interlayer very weak and brittle. Brownish tinge and crazed. 480°F.	760	410	645	170				
15. 54A	- - - - -	825	450	630	140	83.2	5.0		
16. 53A	- - - - -	1012	575	870	170	Very milky			
17. 54B	- - - - -	1120	490	710	120	Very milky			
18. 55B	- - - - -	405	290	486	95	Very Poor	---	---	---
19. 55C	- - - - -	290	275	372	90	Very Poor	---	---	---
20. 58A	- - - - -	1220	660	1165	460	81.9	5.6		
21. 56B	Delaminated. Some haze. 392°F.	1145	585	1160	460	82.0	5.7		
22. 58B	- - - - -	870	530	940	340	80.7	6.9		
23. 59A	- - - - -	810	490	1047	410				
24. 59B	- - - - -	1070	620	1050	450				
25. 60A	- - - - -	1542	700	986	275				
26. 60B	Delaminated. No bubbles. 300°F.	1220	645	1195	450				

Table 16 (cont'd.)

Sample No.	Thermal Stability of laminate after 24 hrs.	Tensile Properties			Optical Properties			
		10 min. / 300°F psi	% El.	24 hrs. / 480°F psi	% El.	10 min. / 300°F % Trans.	24 hrs. / 480°F % Trans.	% Haze
27. 61A	Delaminated. No bubbles. Haze. 392°F.	1210	670	817	310			
28. 61B	- - - - -	1170	675	1018	381			
29. 66C	- - - - -	990	600	1000	470	83.2	7.9	
30. 68C	- - - - -	1110	630	1176	450	81.4	13.2	
31. 126- 25F		1650	900	-----	---	84.7	6.3	

Contrails

After substantial improvement was achieved in tensile strength, the emphasis for improvement was directed towards optical and adhesion properties. Some of the early samples were too poor to consider for measuring transmission and haze. Some of the laminates tested gave quite poor results such as No. 13 in Table 16 which had 69.8% transmission and 15.8% haze. Some of the better results of the first two months work on Type J gave a range of 5 to 6.9% haze and 80 to 83% transmission. However, it was noted at this stage of development that Type J laminates offered quite good vision despite haze of at least 5 percent and transmission of the order of 80 percent. This was particularly apparent when viewing distant objects. As a means of capturing this fact, two photographs were taken to simulate the human eye looking through a Type J laminate as compared with a polyvinylbutyral laminate. In Figure 15, the two laminates (Type J-123-58B on the left, polyvinylbutyral on the right) were positioned one foot from the camera lens and the newspaper located eight feet from the camera lens. The camera was focused on the newsprint in this case. This photograph represented quite well the visional impairment when viewing objects through a typical Type J laminate at distances ranging from several feet to several miles. Figure 16 gives the condition where the camera was focused on the same laminates as used in Figure 15. The camera-to-laminate distance was 2 feet, with the printed matter lying on the table next to the vertically positioned laminates. By focusing on the laminate rather than at a distant point, the haze and transmission deficiencies of Type J were more apparent.

It was found for the Type J laminates discussed thus far that the optical characteristics exhibited a temperature sensitivity. By visual observation it was noted that haze increased at temperatures above 400°F, but returned to the original condition when the temperature was reduced to room temperature.

The temperature sensitivity of the optical properties vastly improved during the final two months of work. In order to accomplish this, it was necessary to sacrifice some tensile strength. Tensile strengths on the limited number of samples prepared of this latest revised Type J have ranged from 400 to 800 pounds per square inch.

During the ninth and tenth months adhesives were not given concentrated effort because Type J was only in the process of being evaluated. During this period it was found to be very advantageous to clean the glass surface sheets with hot chromic acid prior to lamination with Type J. It was found that vinyl-triacetoxysilane or ethyl acrylate treatments did not improve adhesion with Type J. Due to the fact that Type C possessed good adhesion to glass, sheets of Type J were coated with Type C prior to lamination. Little or no success was achieved with Type C as an adhesive.

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TWENTY CENTS

STATE DEMOCRATS ELEVATE DESAPIO TO NATIONAL POST

Tammany Leader Designated
for National Committee as
Only Three Fail to Vote

HE URGES PARTY ACCORD

Successor of Flynn Pledges
Doing Best for Victory
in November Elections

By JAMES A. HAGERTY

The Democratic State Commit-
tee yesterday chose Carmine G.
Desapio, the leader of Tammany
Hall, as Democratic National
Committeeman to succeed the
late Edward J. Flynn.

Technically, the committee
nominated Mr. Desapio for this
party office, as the nomination
must be ratified by the National
Committee. This ratification will
come automatically at the next
meeting of the committee.

The nomination, made at a
largely attended meeting in the
grand ballroom of the Biltmore
Hotel, was virtually unanimous
with only three members of the
State Committee being recorded
as "not voting."

By his nomination, Mr. Desapio
becomes the first member of
Tammany to be chosen for the
National Committee
jury.

The meeting
by Richard H.
at 12:30 p.m.



The New York Times by Herald Staff
CONGRATULATE NEW COMMITTEEMAN: Carmine G. DeSapio, left, receiving the good
wishes of Mrs. William Goud, member of Democratic National Committee and Richard Baich,
Democratic state chairman, after yesterday's election in grand ballroom of Biltmore Hotel.

Lehman Links McCarthy And Dewey as 'Smear' Team FOR A. F. I. BATTLE

TEAMSTERS GIRD

PRESIDENT TO ASK Atomic Talks End in Berlin POWER THIS WEEK TO GIVE ATOM DATA

No Statemate Seen After Dulles, Molotov
Meet Second Time on Procedural Phase
Diplomatic Aides Will Carry On

Message to Seek Sharing of
Weapons News With Allies
and to Spur Peaceful Uses

By JOHN D. MORRIS

Special to The New York Times
THOMASVILLE, Ga., Feb. 13
President Eisenhower will send
Congress a special message next
week detailing his proposals for
immediate revision of the Atomic
Energy Act.

Specifically, he will seek au-
thority to give friendly countries
some information on the use and
effects of nuclear weapons and to
make available to private inter-
ests in the United States data
needed for development of atomic
power for industrial use.

Plans for the special message
were announced by James C.
Hagerty, Presidential press sec-
retary, while President Eisen-
hower was out shooting quail on
the South Georgia estate of
George M. Humphrey, Secretary
of the Treasury.

The President arose early for
a 7:30 breakfast of scrambled
eggs and smoked sausage with
Mr. Humphrey and their two
hunting companions, Clifford Rob-
erts and W. Alton Jones of New
York. Mr. Roberts, a retired
stock broker, is board chairman
of the Augusta National Golf
Club, where the President spends
frequent holidays. Mr. Jones is

By WALTER SULLIVAN
Special to The New York Times
BERLIN, Feb. 13.—John Foster Dulles, Washington, and Charles F.
Dulles, United States Secretary of State, met with Mr.
of Sclavo, and Yurchukov, Mr. Sclavo to Russia, met with Mr.
Molotov, Soviet Foreign Minister, Molotov in Moscow. These chan-
day on President Eisenhower's the Foreign Ministers go home.
atomic energy pool, but discuss appears likely. In the event
sions will continue through other committee talks. The Big Four
diplomatic channels.

United States quarters empha-
sized that termination of the said by Western sources to be
Berlin talks on this subject was raised to objections to a propo-
no indication of a statement. They that the conference end but
have insisted throughout that next week-end
these discussions concerned pro-
cedural matters and did not de-
into the substance of the problem, by the ministers today. It pr
cedural matters and did not de-
forty minutes. The two previous the foreign ministers at 3
meetings here were similarly P. M. tomorrow on the Austr
brief, in each case they came question. This presumably
immediately before or after a
session of the Big Four foreign
ministers.

No details of the negotiations
have been made public. Before
the Berlin conference, Georgi N.
Zarubbin, Soviet Ambassador to
the United States, held two talks
with Mr. Dulles on the subject in
Continued on Page 5, Column 2

French in Indo-China Seek No Korean or U. S. Troop

Type J Laminate on Left and PVB Laminate on Right Simulating
Focusing the Eye on a Distant Object Through the Laminate

Figure 15

Contrails

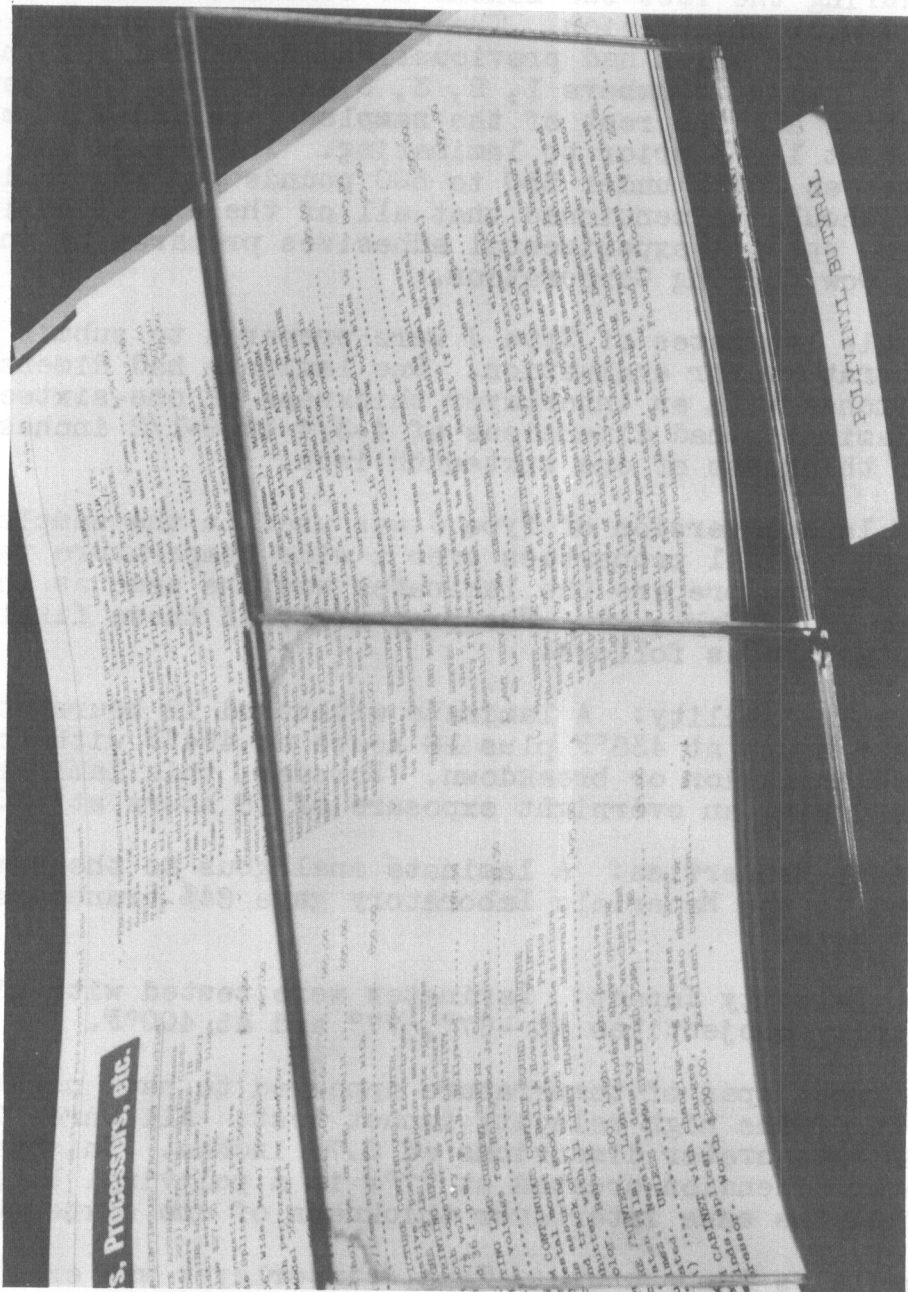


Figure 16

Type J Laminate on Left and PVB Laminate on Right Simulating Focusing the Eye on the Laminate

Adhesion is of paramount importance for the success of a laminate. Consequently, much effort was directed toward adhesion improvement during the last two months of contract work. Table 17 summarizes the work on adhesion. The adhesives were brushed on the glass surface sheets which had previously been cleaned with hot chromic acid solution. Numbers 1, 2, 3, 5 and 6 were then laminated with Type J while all the rest of the samples received a 10 minute heating period at 122°F prior to laminating. The laminating conditions were 10 minutes at 300°F under 250 to 500 pounds per square inch pressure. It should be mentioned that all of the adhesives listed in Table 17 are special experimental adhesives prepared by one of the groups at the Dow Corning Corporation.

Sample laminates of Type J were prepared to submit to the Materials Laboratory for evaluation. One laminate had dimensions of 18 by 18 inches with an interlayer thickness of one-sixteenth inch. Five laminates had dimensions of 4-3/4 by 8-1/2 inches with an interlayer thickness of one sixteenth inch.

The latest version of Type J was used in the samples to insure the best optical properties over a wide temperature range. The adhesive used to prepare the laminates was the same as that used in Number 18 of Table 17. The properties of these final Type J laminates are as follows:

1. Thermal stability: A laminate withstood 18 hours at 400°F plus 6 hours at 436°F plus 18 hours at 454°F with no signs of delamination or breakdown. However, this laminate broke down during an overnight exposure of 18 hours at 480°F.
2. Optical Properties: A laminate analogous to the samples sent to the Materials Laboratory gave 84% transmission and 6.7% haze.
3. High Velocity Impact: Laminates were tested with high velocity projectiles at -67°, 77° and at 400°F.

The broad span of temperature appeared to have no effect on the results of the high velocity impact test. All three laminates had identical exit crater dimensions of 0.75 inches. This was also the exit crater dimension created at 77°F in a polyvinyl butyral laminate having the same interlayer thickness of one sixteenth inch.

4. Tensile vs Temperature: Type J interlayer was examined over a wide temperature range to determine the effect on tensile strength.

Contrails

Table 17

Adhesive Tests with Type J

<u>Reference No. 126-2-</u>	<u>Adhesion</u>	<u>Peel Strength lbs./inch of width</u>	<u>Remarks</u>
1. 23E	Good	----	Adhesive darkened at 392°F
2. 24A	Good	----	Adhesive turned white
3. 24B	Fair	----	Whitened at 300°F
4. 25B	None	----	-----
5. 25C	Good	10.0	1 plate laminate
6. 25D	Good	----	2 plate laminate
7. 26A	None	----	Adhesive thermoplastic
8. 26B	None	----	Adhesive thermoplastic
9. 26C	None	----	Adhesive thermoplastic
10. 26D	Poor	----	Delaminated at 392°F
11. 26E	Fair	----	No delamination at 392°F
12. 26F	Fair	----	No delamination at 392°F
13. 26G	Fair	4.4	No delamination at 392°F
14. 26H	Fair	5.0	No delamination at 437°F
15. 26J	Good	10.0	Cured 1 hour at 392°F
16. 27E	Fair	5.6	Aged 1 hour at 392°F
	Good	8.8	Aged 2 hours at 392°F
	Good	10.8	Aged 17 hours at 392°F
17. 27H	Fair	5.6	-----
18. 28A	Very good	26.5	Cured 25 min. at 300°F
	Very good	>28.0	Aged 18 hours at 392°F Interlayer broke before adhesive bond.

Contrails

Samples were allowed 15 minutes to reach equilibrium at test temperature before testing. Previous high temperature test data showed that a 15 minute conditioning period was adequate to reach equilibrium for the size test samples used.

The results are given in Figure 17. The tensile strength of the particular sample tested was 483 psi at 77°F and was reduced to 264 psi at 450°F. This degree of drop-off could be expected for other samples of Type J, but the room temperature tensile strength might be higher or slightly lower. The tensile values at room temperature have ranged from 400 to 800 psi for different batches of the final modification of Type J.

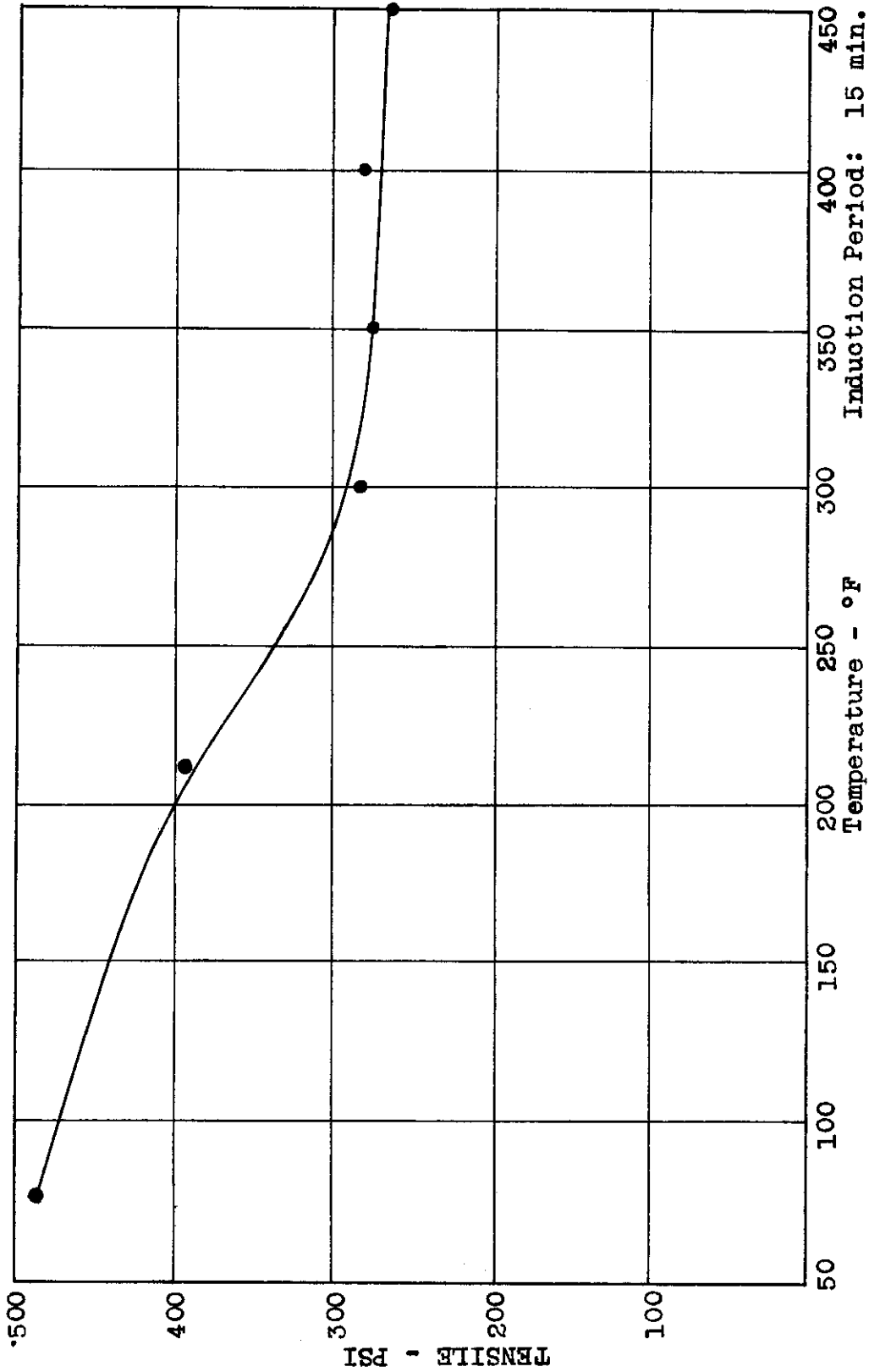


Figure 17
Type J Tensile Strength
vs Temperature
Induction Period: 15 min.

Summary

Originally, five types of polysiloxane materials were chosen to be evaluated. These potential interlayer materials were designated Types A through E. Preliminary tests showed the following characteristics:

<u>Type</u>	<u>Optical</u>	<u>Thermal</u>	<u>Adhesion</u>	<u>Tensile Strength</u>	<u>Handling</u>
A	Very good	Poor	Fair	Fair	Fair
B	Very good	Very good	Good	Poor	Fair
C	Good	Very good	Good	Fair	Poor
D	Very good	Good	Fair	Good	Poor
E	Fair	Fair	Poor	Fair	Good

After further work, Type A was dropped because of the poor thermal properties. Type B was discontinued because of its poor strength. Type D was eliminated because of its tendency to darken on heating and because its other properties were parallel to those of Type C. This left only Types C and E of the original five.

In addition to concentrating efforts on Types C and E, a search for new materials was continued. One of these materials, Type F, was investigated and showed some promise, but had to be sidetracked due to material inconsistencies.

A Type G was studied, but its thermoplastic character could not be improved, so work on this interlayer was discontinued.

By the end of the first six months of work, Type E had been improved to the point where it was the best interlayer material. Six month samples of Type E laminates were given to the Materials Laboratory for evaluation.

Shortly thereafter, a substantial improvement was made on Type C which resulted in slightly better thermal stability and better adhesive strength than Type E. Other properties were about equal to those of Type E.

During the seventh and eighth months of work Type H was introduced, but was eliminated because of poor low temperature characteristics and no advantage in tensile strength over Type C or E.

The ninth month of work produced Type J. Type J possessed tensile strengths of 400 to 800 psi, 80 to 85% transmission, 4 to 10% haze and thermal stability at 454°F. Type J offered the possibility of using commercial fabrication techniques. At the very end of the contract work, an adhesive was introduced to use in laminating Type J which gave adhesive peel strengths greater than 20 pounds per inch width. A laminate prepared from Type J and this adhesive withstood 18 hours at 400°F plus 6 hours at 436°F plus 18 hours at 454°F with no signs of delamination or breakdown. Given in Table 18 is a comparison

Contrails

of the properties of PVB with Type J. Properties are given of glass laminates as well as the interlayers themselves. The comparison points out the fact that the optical properties of Type J interlayer may require some improvement before this material will be completely satisfactory for aircraft glazing.

A new contract will be started shortly to evaluate Type J laminates with the objective of improving the optical and adhesion properties while retaining the present tensile strength and thermal stability.

Comparison of PVB with Type J

(a) Glass Laminates

<u>Property</u>	<u>PVB¹</u>	<u>Silicone-Type J</u>
Thermal Stability- No degradation of any sort	300°F for about 1.25 hrs. ²	454°F for greater than 18 hours
Adhesion	Excellent ²	Good
Haze	1.2% ²	4-10%
Light Transmission	91%	80-85%
High Velocity Impact	0.75 inch dia. exit crater at 77°F	0.75 inch dia. exit crater at -67°, +77°, +400°F

(b) Interlayer Alone

Tensile Strength	2000 psi ²	400-800 psi
H ₂ O Absorption ³	2.47% by wt.	0.32% by wt.

1. PVB with 37.5 parts by weight of DBS.
2. Values given by Lt. R. Mandel, Transparent Materials Section, Materials Laboratory, Wright Air Development Center.
3. Determined According to Federal Specification L-P-406b, Method 7031.