

**RESEARCH, DEVELOPMENT, AND FABRICATION
OF SILOXANE-ACRYLATE INTERLAYER MATERIAL**

B. DAVID HALPERN

WILLIAM PROSK

LEROY TIBERY

JOHN ISLER, JR.

WOLF KARO

MONOMER-POLYMER

THE BORDEN COMPANY - CHEMICAL DIVISION

DECEMBER 1955

MATERIALS LABORATORY

CONTRACT AF 33(616)-2528

PROJECT No. 7340

WRIGHT AIR DEVELOPMENT CENTER
AIR RESEARCH AND DEVELOPMENT COMMAND
UNITED STATES AIR FORCE
WRIGHT-PATTERSON AIR FORCE BASE, OHIO

Contrails

FOREWORD

This report was prepared by Monomer-Polymer, The Borden Company - Chemical Division, under USAF Contract No. AF 33(616)-2528. This contract was initiated under Project No. 7340, "Rubber, Plastics, and Composite Materials", Task No. 73403, "Transparent Materials", formerly RDO No. 616-12, "Transparent Materials" and was administered under the direction of the Materials Laboratory, Directorate of Research, Wright Air Development Center, with Lt. Don Schmidt acting as project engineer.

This report covers period of work from June 1954 to June 1955.

The cooperation of the following companies and personnel is gratefully acknowledged: Mr. J. M. Joyner, The Proves Research Laboratories, Farrell-Birmingham Company, Ansonia, Connecticut; Mr. Luther Yaeger, Bjorksten Laboratories, Madison, Wisconsin; Mr. W. H. Bamford and Dr. J. T. Scheuer, Rex Corporation, West Acton, Massachusetts; Mr. L. A. Keim, Pittsburgh Plate Glass Company, Pittsburgh, Pennsylvania; and Dr. J. D. Ryan and Mr. P. T. Mattinoe, Libbey-Owens-Ford Company, Toledo, Ohio.

The help of Lt. R. Mandel, Lt. N. M. Wilson, II, and Capt. F. J. Wilcox of the Materials Laboratory is also gratefully acknowledged.

WADC TR 55-296

Contracts

ABSTRACT

The object of this research, development, and fabrication program was for the production of interlayer sheet material. This interlayer material consisting of a copolymer of 99 parts of ethyl acrylate and one part of vinyl polysiloxane was developed under Air Force Contract 33(600)-22723 and is described in WADC TR 54-57.

This developmental work was undertaken between 15 June 1954 and 15 June 1955.

Attempts to mill and calender the interlayer material were unsuccessful. This necessitated the production of interlayer sheets by casting between glass plates. Thermal polymerization was substituted for ultraviolet polymerization for quality production purposes. Sheets produced by this method, however, were optically imperfect.

An adhesive was required for laminating the interlayer sheet to glass or plastic. Residual traces of the parting agent, however, prevented good lamination.

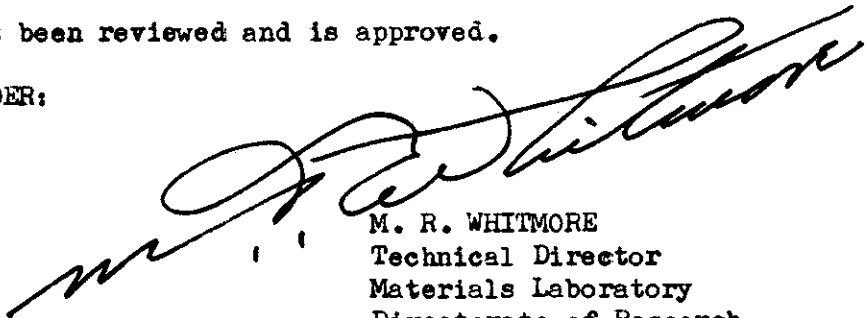
Seven 0.200" by 37" by 42" interlayer sheets varying in quality were submitted.

Recommendations are given for methods of improving the optical quality of future interlayer sheets.

PUBLICATION REVIEW

This report has been reviewed and is approved.

FOR THE COMMANDER:



M. R. WHITMORE
Technical Director
Materials Laboratory
Directorate of Research

Contrails

TABLE OF CONTENTS

	<u>Page No.</u>
I. INTRODUCTION	1
II. DISCUSSION	2
A. Theory	2
B. Milling and Calendering Experiments	4
C. Preparation of Vinyl Polysiloxane	5
D. Preparation of Casting Syrups	5
E. Parting Agents	6
F. Experimental Cells	7
G. Physical Properties of Interlayers	9
H. Lamination Experiments	9
I. Large Cell Production	10
III. SUMMARY AND CONCLUSIONS	13
IV. BIBLIOGRAPHY	15
V. APPENDIX	16
Release Agents	17
Correspondence	18
Tables	23

Contracts

LIST OF TABLES

<u>TABLE NO.</u>	<u>TITLE</u>	<u>PAGE NO.</u>
I.	Bulk Polymerizations of Ethyl Acrylate to Syrup	23
II.	Experimental Cast Interlayers	24
III.	Large Interlayers, Preparation	28
IV.	Large Interlayers, Characteristics	29
V.	Further Cast Interlayers	30
VI.	Comparison of Physical Properties of WADC 54-57 Interlayers With Those Produced Under This Contract	38

Contrails

Contracts

I. INTRODUCTION

The purpose of this contract was to obtain sufficient ethyl acrylate-polyvinyl siloxane interlayer sheet material for research test work on laminated plastic and glass. This material was to conform to the material known as Bjorksten A-1054, developed by the Bjorksten Research Laboratories, Madison, Wisconsin, under Air Force Contract AF 33(600)-22723.

It was requested that a screening investigation be performed at the outset of the contract to determine whether (1) casting the sheets in glass cells or (2) bulk polymerization of the prepolymer, with subsequent calendaring into sheets, was the most feasible method of preparing the prescribed interlayer.

Fifty interlayers 36 inches wide, 0.020 ± 0.005 inches and having a minimum length of 36 inches was originally requested. However, because the amount of research and development work was much greater than originally anticipated, change order no. C1(55-994) requested that 5 sheets of 0.200" by 37" x 42" ethyl acrylate-polyvinyl siloxane interlayer material be delivered in place of the fifty thinner sheets with the contract price remaining the same.

II. DISCUSSION

A. Theory

Since the desired interlayer contains 99% ethyl acrylate, knowledge of ways to polymerize this monomer to bubble-free, optically clear and undistorted product would be useful.

Bulk polymerization of acrylic esters on a commercial scale has been almost exclusively in the production of cast sheets of methyl polymethacrylate (3) (17). The physical properties of methyl polymethacrylate are vastly different than those of polyethylacrylate so all techniques effective for one may not be applicable to the other. Methyl polymethacrylate is a fairly hard, brittle, tack-free polymer having high tensile strength while polyethylacrylate is a soft, rubbery, tacky polymer having low tensile strength. However, some of the problems of polymerization are common to each monomer.

The two principal difficulties in achieving a good quality sheet are adjustment for the 20% shrinkage in thickness of the interlayer that takes place during polymerization, and adequate control of the highly exothermic heat of reaction in the early stages (3), especially after the material has reached the gel stage. Other difficulties are in preventing "fisheyes", "pinholes", "popcorn", "bubbles", "waves", dirt, color, cracks, optical distortion, haze, sticking to the mold, and in obtaining the required physical properties such as tensile strength, thermal stability, low temperature flexibility, and flex resistance, and ability to be adhered to glass (1, 2, 3, 5).

The greatest problem to solve is the one caused by shrinkage of monomer during polymerization. Numerous procedures have been patented in an attempt to overcome this difficulty (3)(4). WADC 54-57 (1) stresses slow polymerization under diffused light as essential. Curing can be overdone however, since another source states that any cells (of methyl polymethacrylate) cured 30 or more days contain "lakes" or "sinks". "Lakes" and "sinks" are uneven depressions away from the cell wall that show up as optical imperfections.

Most of the cells leading to the results reported in WADC 54-57 were 6" x 6" in size while only 8-10 cells were as large as 30" x 30" (15). A compressible gasket was essential for producing optically clear cells (3). This gasket was either laminated cork, Hycar rubber, polyethylene or Tygon tubing (15). With the latter, some discoloration and swelling occurred at the outer two inches of the sheets but this was trimmed away (15). Interlayers produced in WADC 54-57 (1) had many of the same optical defects (5) encountered in this research.

Contrails

Two differences between the material reported in WADC 54-57 (1) and that required for completion of this contract are in the size of the sheet and the requirement that the sheet be stripped from the cell walls. The bigger the sheet, the more difficult it is to make it optically perfect. With small cells, lakes may be eliminated by applying sufficient pressure at the cell edges, if the gasket is flexible enough. With larger cells, pressure applied at the edges cannot be uniformly distributed to the center of the cell. Uniformity of cell mix, photo illumination and heat transfer, are also more difficult to achieve in the larger cells.

The vinyl polysiloxane pretreatment used to help in eliminating "lake" formation by improving adhesion between interlayer and glass (15) used cannot be used if the glass is to be readily separated. The parting agent added makes separation easy but makes "lake" formation more likely and makes good lamination more difficult.

With some monomers the formation of a porous granular insoluble polymer having a physical appearance similar to popcorn has been observed (3). Lack of oxygen, water contamination, iron contamination and storage at too high a temperature are known to contribute to the formation of popcorn polymer (3). It can be prevented if sufficient amounts of catalyst or chain transfer agent are present (3).

Extraneous polymers are found to accelerate the polymerization (3). Since, even when refrigerated, ethyl acrylate is subject to deterioration (3) in quality in about a week, it is advisable to test for the presence of polymer (3) if monomer is not used as soon as prepared. (Some dry, inhibitor free monomer stored at 16-22° C. for 1 month in steel gave a negative test for polymer.)

Bulk polymerization time is difficult to estimate for another reason. Oxygen, even in the relatively low concentrations found in commercial nitrogen, has an inhibiting effect (3). However, excess air (oxygen) can lead to the formation of peroxides which catalyze the polymerization. Differences in activity of the peroxide catalyst can also affect induction time.

Color in the cell is due to impurities. Waves are believed to be caused by uneven polymerization. Haze may be caused by traces of water. Desired physical properties are a result of polymerization techniques.

Contrails

"Fisheyes", "pimples", "pinholes" and "dirt" are optical imperfections that appear mostly on the surface of the interlayers. They cannot be readily seen when the interlayer is between glass, but are undesirable since they represent areas of non-homogeneity. The cause of "fisheyes" is not clear. They are formed in spite of all efforts at cleanliness in preparing the casting cells, and despite extreme care in applying an even film of parting agent. They appear to be caused by local polymerization to a cross-linked polymer which is not homogeneously distributed throughout the remaining sheet. Dust, polymer "seed" particles, prematurely cross-linked vinyl polysiloxane, the nature of the polymerization cycle, or other factors may also contribute to "fisheye" formation.

"Fisheyes" appear to be more prevalent in cells prepared from syrup or monomer over two days old (from date of distillation) even though the monomer was kept refrigerated, and in cells containing PVS which had been prepared by evaporation of the solvent not only at reduced pressure but also at a temperature near 100° C. for a prolonged period of time.

B. Milling and Calendering Experiments

It was originally believed that a continuous 20 mil sheet of interlayer material might be formed by milling and calendering starting with fully polymerized ethyl acrylate. To test this hypothesis, several runs were conducted at the Research Laboratory of Farrell-Birmingham at Ansonia, Conn. The starting materials were suspension-polymerized ethyl acrylate and butyl acrylate. These materials closely resembled Bjorksten A-1054 interlayer material except that the latter contained 1% vinyl polysiloxane cross-linking agent.

The results of all the runs were negative; it was impossible to obtain a uniform sheet from the acrylate rubbers off either the mill or calender. The rubbers had a great deal of "nerve" and resilience which prevented the formation of a homogeneous sheet from the mill even after prolonged working at various roll speeds and temperatures. The rubbers showed great adhesion to metal which compounded the problem. The difficulty in milling was partly attributable to the fact that the acrylate rubbers were slightly cross-linked as evidenced by gel formation. This is thought to occur by chain transfer involving an alpha hydrogen on the acrylate moiety.

In the case where 1% siloxane would be present the degree of cross-linking would be even greater.

It was regarded as inevitable that flat sheets would have to be prepared most practically by casting in cells.

C. Preparation of Vinyl Polysiloxane

Linde X-31 resin was the source of the vinyl polysiloxane. The siloxane powder was recovered from the X-31 toluene solution by distilling off the solvent at 70-100° C. under the 27" vacuum obtained by a water aspirator. Technical Report WADC 54-57 (1) does not give the time-temperature-pressure at which vinyl polysiloxane was recovered, but a bulletin of Linde Air Products states that the resin may be obtained "by vacuum desolvation at 110° C. and 27" vacuum". However, a more recent communication (19) from them states that "desolvation above 60° C. tends to cross-link the resin in a manner as to decrease its solvent solubility". They produce a material designated as Linde Y-1004 which is Linde X-31 silicone which has been desolvated, i.e., freed of solvent, to about 99% solids by heating below 60° C. (water bath) at reduced pressure (3 to 5 mm.) for about 5 hours. Presumably, the shorter the heat exposure the less cross-linking will take place. Since Y-1004 has the least history, it is expected that it would have the least amount of cross-linking. The 1% of nonsolids in Y-1004 might affect the physical properties of the interlayer so this point should be investigated before large cells are produced.

D. Preparation of Casting Syrups

The pre-polymerization of monomer to a thin syrup has been recommended for the production of interlayers (1). Preparations of syrup were made using diacetyl as an ultraviolet absorption catalyst as described in WADC 54-57, and shown in Table I. However, cells containing diacetyl require an extended ultraviolet radiation cure to decompose residual diacetyl catalyst, and even then are not as clear as cells made without this material.

To increase the production rate of cells, better methods of syrup production were studied. Much work was done on thermal polymerization of syrup with various amounts of different thermal catalysts. No method was completely satisfactory when applied to large batches. The runs were characterized by indeterminate induction periods followed by vigorous reactions. Apparently the first trace of polymer catalyzes the reaction, hence, future experiments should be run with "seeded" monomer.

Syrups prepared from colorless benzoin using ultraviolet irradiation developed some color during the cell curing process.

Tests at Monomer-Polymer indicated that monomer prepared interlayers were as strong and heat stable as samples of interlayer material produced in WADC 54-57 from syrup (1). Other, later tests at Wright Air Field were not in agreement with these results.

Contrails

Syrup produced cells have other advantages. Casting syrups avoids long induction periods in the mold (3), allows polymerization to proceed at a low enough temperature to prevent "run-away" polymerization, reduces the dissolved gas content with consequent bubble formation (3), and reduces the danger of leakage from the mold.

Further studies established a quick, reproducible method of syrup production without the use of added catalysts. The essentials of this technique are:

- a. Displacement of dissolved air with CO₂ bubbled into the solution.
- b. Intense ultraviolet radiation (300 watt - 1" away).
- c. Good agitation (120 rpm) to prevent polymer build-up near the light source (blades scrape the sides of the flask).
- d. Seeding with syrup from a previous batch.
- e. Control of temperature between 32-41° C.
- f. Stopping the reaction with air and cooling at a given viscosity (by observing power requirements for constant rpm).

This method was used to prepare syrup used in cells that were colorless.

E. Parting Agents

Cells could not be separated from glass without the use of a parting agent. This parting agent can be another film such as Mylar, Kel-F, or polyethylene or a release agent. A list of these parting agents and their manufacturers is given in the Appendix.

Some of these materials stained the interlayer to varying degrees. Aerosol OT, Aerosol AY and Stearic Acid appeared to cause very little staining. The Aerosol OT was applied to the glass plates from a 10% gel in distilled water but can be applied from other solvents to facilitate drying. Stearic acid was added to the monomer mixture (0.1%) and appeared promising. Aerosol AY is somewhat similar to Aerosol OT but forms water clear solutions which are easy to prepare. Unless otherwise specified, tabulated cells had Aerosol OT as the release agent.

Contrails

F. Experimental Cells

The cells were composed of two glass plates with a "dam" construction of two general types. One type was formed by using spacers between the plates until a glue soaked parchment tape around the edges dried. The spacers were removed as the cell was filled with monomer or syrup. After charging, the top edge was sealed and the cell placed at a 15° angle with the horizontal. When the gel stage was reached, the tape was slit to permit the glass to follow the shrinkage of the interlayer.

The second type utilized a compressible tubing which was compressed with "C" clamps spaced 1" apart around the edges of the glass. Rubber or felt washers prevented injury to the glass from the metal clamps.

Monomer was distilled at atmospheric pressure with the first and last fractions being discarded. (Phenothiazine was added to the still pot as a polymerization inhibitor.) Each fraction was checked for absence of hydroquinone. In many instances, the refractive index and density were also obtained, but the small variations permitted no correlation with polymerization behavior. In general, distilled monomer for the small cells was stored in dark glass bottles while that for the larger cells was stored in new steel cans in an outside air-cooled box.

Early experimental cells were produced to duplicate previous work, and to determine the most effective parting agent, and cell gasket. Later cells were made to test various syrups, determine an optimum cure cycle, produce material for testing, produce material for lamination experiments, and improve casting and polymerizing techniques to eliminate optical imperfections.

Tables II, III, IV and V are representative of the cells produced under this contract. The problems and the ways they differ from those of WADC 54-57 (1) are generally discussed under "II. Discussion, A. Theory" of this report.

The procedure described in WADC 54-57 (1) whereby pre-polymer syrup is produced by irradiating monomer containing diacetyl requires that the final curing of the interlayer be by ultraviolet irradiation in order to decompose the residual yellow diacetyl. This method is not as readily adapted to volume production in existing plants and equipment as are thermal methods and cells produced were no more optically perfect than thermal cells.

Contrails

Although induction time of freshly distilled monomers is more predictable than older monomer (possibly because of the conversion of dissolved air to peroxides, or the formation of a few "seeds"), some cold storage of monomer is required if several large cells are to be polymerized simultaneously. Other than affecting the polymerization time, this is not considered to affect the physical properties of the interlayer adversely since a.) more seeds are present in WADC 54-57 syrup produced interlayers and b.) 0.01% benzoyl peroxide was present in interlayers having good physical properties. Water to the extent of 1% added to the monomer (cell 213) resulted in an opaque cell and cells which contained haze may have acquired this condition because of insufficient drying of cells or monomer before distillation or casting. Monomer dried a minimum of 48 hours over Drierite (anhydrous calcium sulfate) produced cells that were not always optically perfect (some still contained lakes, waves and pinholes) but were optically clear (provided the polymerization did not "run away").

The experimental cells established the following additional points:

1. Most of the cell imperfections (except "lakes" and "fish-eyes") are eliminated with slow polymerization with a monomer-resistant compressible gasket until a gel stage is reached. Polymerization at 40° C. with 0.01% benzoyl peroxide added to air saturated monomer or syrup (or up to 47° C. with good heat transfer) is a satisfactory technique for preventing "run-away" reactions.

2. Rubber and vinyl chloride tubing are not suitable gaskets unless protected from leaching with a protective film such as "Mylar".

The cause of "fisheyes" was not clearly established. They were present in monomer as well as in syrup cells and were originally thought to be caused by dust since they appear to be only on the surface of the interlayer. Since "fisheyes" are areas of non-homogeneity and are not detectable until the glass is stripped from the cells, they might also be present in the interlayer. There is some evidence that indicates that "fisheyes" may be caused by vinyl polysiloxane cross-linking with itself.

The problem requiring the greatest effort was "lake" formation or uneven shrinkage of the interlayer from the glass plates during polymerization. This shrinkage was determined to take place a few hours after a "gel stage" was reached. It was established that these "sinks" were not caused by either PVS, improperly applied parting agent, or improperly cleaned glass plates.

Cell 228 (parting agent Aerosol AY) described in Table V was lake free at the end of the cure cycle. It should be noted that lakes were originally present but were made to disappear by application of additional pressure with the cell still in the oven during the final cure. On larger cells, uniform pressure at the center of the cell cannot be applied by clamps at the edges. Attempts to do so result in cell bulges and cracking of the glass plates.

It is expected that larger cells could be made lake free if sufficient pressure could be applied in such a way that good heat transfer would not be prevented. The contract expired before this approach could be pursued.

G. Physical Properties of Interlayers

The results of testing two 15" x 12" thermally polymerized interlayers (0.01% benzoyl peroxide, no diacetyl) at Wright Air Development Center are shown in the Appendix. Results indicate that the submitted samples are inferior to WADC 54-57 produced material (1)(5).

Comparison tests shown in Table VI indicate that thermally polymerized sheets are at least equal in measured physical properties to the tested samples of WADC 54-57 material. Because of the inherent difficulties involved in comparing physical properties obtained at one laboratory with those obtained at another (or at two different times) it is recommended that future tests be run with controls consisting of WADC 54-57 produced material.

H. Lamination Experiments

Lamination experiments (cells 135, 137, 144, 147, 149) at Libbey-Owens-Ford, described in the Appendix, established that successful laminates would require an adhesive to overcome the release properties caused by residual parting agents.

The results of bonding experiments of the Pittsburgh Plate Glass Company (cells 162, 163, 164) are shown in the Appendix. The results do not appear too promising at first glance. However, a telephone call established that only four adhesives were tested. These tested adhesives are not normally tested at 400° F. so the reported failures are not too significant. None of the four adhesives tested was a silicone type. It was suggested that products such as (a) General Electric adhesive No. 81517, which is a newly developed water white silicone polymer adhesive useful over the range -100° F. to 500° F., (b) the "universal"-type glass to resin bonding agent discovered by the Naval Ordnance Laboratory (condensation product of resorcinol and allyltrichlorosilane), and (c) adhesive XA-4034 developed under AF 33(600)-27185, "Silicone Interlayer Material Program" should be tried in future bonding experiments.

Contrails

I. Large Cell Production

Tables III and IV summarize the seven largest cells produced in an effort to meet contract requirements. All contained 0.01% benzoyl peroxide and 1% PVS.

A. Cell 151

The plate glass sheets were 48" x 44" x 1/4". The glass plates were first washed with a 5% aqueous solution of Aerosol OT, then polished with Monsanto lampblack and finally, rinsed with water. After washing, a thin layer of 10% Aerosol OT was applied to the glass plate surfaces which could contact the casting solution. The glass plates were laid up parallel with three steel rods 42" x 1/4" x 1/4" as spacers.

The cell was sealed by taping up the periphery with three successive layers of parchment paper soaked in animal glue (Arabel CFG-100).

After the parchment paper dried, it was covered with Scotch masking tape.

The 20 pound charge consisted of 98.99% distilled ethyl acrylate monomer, 1% PVS, and 0.01% benzoyl peroxide.

The cell was set into a forced air draft steam-heated oven, inclined several degrees above the horizontal. The liquid level was within 4 inches of the cell top.

The following heat cycle was used:

64 hours at 41-50 deg. C.
23 hours at 65 deg. C.
24 hours at 85 deg. C.

At the end of 64 hours, the parchment paper was split and the top glass plate was removed.

The sheet was clear and smooth with very few flaws. The thickness was 0.20 ± 0.01 inch over the whole sheet. Since the spacers were 0.25 inch thick, the shrinkage was 20% of the original thickness. There was practically no shrinkage in length or width.

The interlayer was cut to 37" x 42.75" and covered on both sides with Mylar film.

B. Cell 154

Another casting was produced using the same technique as before, except that Plexiglas spacers were used instead of steel, and rouge used instead of lampblack as a polishing agent. Also, after splitting the parchment paper, the glass plates were left in place during the entire cure cycle.

The sheet had more flaws than 151. There were many pinholes and protrusions and the sheet was quite hazy in some spots. The pinholes may have been caused by dust. Future work emphasized the elimination of dust from the casting solution and the cell, and greater attention to cleanliness in cell handling.

The sheet was cut to 37" x 42.5" and measured 0.195 ± 0.012 inch in thickness.

C. Cells 230, 231, 232, 233 and 234

The presence of microscopic dust particles in the air and on the surface of cleaned glass had contributed to the difficulty of casting an optically perfect sheet. It was felt that each particle served as a locus of either increased or decreased polymerization activity and resulted in a final sheet with so-called "pimple" distortion. In an attempt to obviate this difficulty, arrangements were made with a manufacturer of methyl polymethacrylate to make cells and cast five large sheets in a dust free plant.

On the basis of experimental cell work it was decided to produce three of these cells of syrup and two from monomer. The syrup used contained no photo-catalyst since experimental cells established that this procedure produced cells with the least amount of color. The initial cure temperature, 47° C., was the average of that used for cells 151 and 154.

The syrup cells were saturated with air before charging. Monomer cells 233 and 234 were degassed after charging since under the expected dust and seed free conditions it was felt that polymerization might not otherwise occur in 72 hours at 47° C. Inside cell size was 40" x 40" since this was the largest their oven could accommodate. All weighed, pre-cooled raw materials were supplied in separate sealed containers (new 5-gallon steel cans for the ethyl acrylate monomer and syrup) containing the correct amount of each material to be mixed for each cell. They were also supplied with the 0.364 inch O.D., 0.320 inch I.D. polyethylene gasket as well as the filtered 10% Aerosol OT mixture in distilled water. Cells after filling were inclined for several hours at room temperature.

Cells were placed in the oven at 47° C. in order with cell 230 being at the bottom. When the cells were examined 18 hours later it was noted that monomer cells 233 and 234 had polymerized to a white bubbled mass in the center of each cell. The edges and cells 230, 231 and 232 had not polymerized.

Contrails

Forty-two hours from the time the cells were placed in the oven, the cells were examined again. All cells still appeared liquid at the edges but cell 231 had a 6" x 9" section in the middle that contained bubbles and was white. All cells had "sink" marks ("lakes") in them over the entire polymerized surface.

At the end of 72 hours the cells had decreased in thickness about 0.030" from the original 0.250", and all had polymerized to the edges. Clamp pressure could not be increased at this time without danger of breaking the glass. Final cure was as shown in Table III. No difference is known that would explain why the center of cell 231 prematurely polymerized while adjacent cells 230 and 232 did not. In all probability, the 5" spacing of cells, although twice as great as used in methyl polymethacrylate production, did not permit as good heat transfer as the forced air circulation oven at Monomer-Polymer.

Cells were also polymerized in a more nearly horizontal position which may have prevented the bubbles formed from leaving the mix.

"Pimples" or "fisheyes" were, if anything, more prevalent than in cell 154. Whether this was caused by differences in heat history of the dried vinyl polysiloxane, dirt, or other factors has not been established.

III. SUMMARY AND CONCLUSION

Milling and calendering experiments were unsuccessful in producing the desired interlayer of a copolymer of 99 parts of ethyl acrylate and 1% vinyl polysiloxane because the "nerve" of the ethylpolyacrylate prevented the formation of a homogeneous sheet.

A method of thermal polymerization was developed that appears better for quantity production of large interlayers than the photo-polymerization method. The method consists in polymerizing an air-saturated mixture of 0.01% benzoyl peroxide, 1% vinyl polysiloxane, and 98.99% ethyl acrylate (as monomer or pre-polymerized syrup), at 40-47° C. for 72 hours, 57° C. for 8 hours, and 80° C. for 8 hours. Careful attention to the following details is essential to achieve the best optical qualities:

1. Absence of dust.
2. Absence of water or dirt in monomer.
3. Preparation of dry vinyl polysiloxane at low temperatures.
4. Purity and age of distilled monomer.
5. Purity of catalyst.
6. Collapsible gasket construction of inert material.
7. Good heat transfer.
8. Uniformity of pressure on cell during polymerization without adversely affecting uniform heat transfer.
9. Uniform presence of dry air in monomer.
10. Absence of stray irradiation.
11. Method of applying parting agent to glass.

Seven large sheets were prepared by this method. Optical imperfections of these sheets can be explained by deviations from the desired technique.

WADC tests indicate that two other submitted samples of thermally polymerized interlayers may not be as good as WADC 54-57 material. Comparison tests at Monomer-Polymer indicated that the materials were equal in measured properties.

Experiments at Libbey-Owens-Ford Glass Company showed that good lamination could not be achieved without the use of an adhesive. It is believed that residual traces of the required parting agent is preventing good lamination.

Conclusions

Four unsuccessful laminations with adhesives were made at Pittsburgh Plate Glass. Since the adhesives themselves are not normally tested at 400° F. the reported failures are not too significant. It is recommended that the three adhesives suggested in the body of this report be tested.

On the basis of the experience gained on this program, it is considered advisable that future work give due consideration to the following points in the effort to prepare consistently successful interlayer materials:

1. Substitution of Linde resin Y-1004 for solvent-free Linde resin X-31 as a source of vinyl polysiloxane.
2. Construction of cell gaskets from a flexible material such as thin-wall polyethylene tubing (1/32" wall thickness or less).
3. Application of uniform pressure to the cell face immediately after gelling under conditions that permit uniformly good heat transfer (such as circulating oil bath at 40° C. in a pressurized autoclave, or a series of turn-buckle type rod jacks).
4. Use of freshly distilled, air saturated, moisture- and iron-free monomer or syrup, with 0.01% of benzoyl peroxide and 1% of Linde resin Y-1004, for cell charges.
5. Investigation of siloxane and other suggested types of adhesives for lamination of the interlayer between glass.

IV. BIBLIOGRAPHY

1. Cox, R. P., Yaeger, L. L., Buetow, R. W., Roth, R. J., WADC 54-57, "Development of Heat Resistant Interlayer Materials for Laminated Plastic and Laminated Glass", July 1954.
2. Polmanteer, K. E., Cretzmeyer, J. W., Erwin, J. W., WADC 54-207 Part I, "Development of Silicone or Fluorocarbon Interlayer Materials for Laminated Glass and Laminated Plastic", October 1954.
3. Riddle, E. H., "Monomeric Acrylic Esters", Reinhold Publishing Company 1954.
4. Blout, E. R., Mark, H., "Monomers", Interscience Publishing Co., 1951.
5. Mandel, R., "Evaluation of Transparent Ethyl-Acrylate-Vinyl Polysiloxane Interlayer Developed by Bjorksten Research Laboratories under Contract AF 33(600)-22723", Technical Note WCRT 54-250, December 1954.
6. Schildknecht, "Vinyl and Related Polymers", John Wiley and Sons, 1952.
7. "Alkenyl Siloxanes", 19 February 1951, Linde Air Products Company

Contrails

V. APPENDIX

WADC TR 55-296

-16-

<u>Trade Name</u>	<u>Chemical Type</u>	<u>Source</u>
Solricin 535	Soap	Baker Castor Oil Company
Bomb-lube	Silicone spray	Price-Driscoll Corp.
D-C Silicone	Silicone greases	Dow Corning Co.
Tween 61	Mono stearate	Atlas Corp.
Glycerine	Alcohol	Shell Chemical Co.
Slipicone	Silicone	Dow Corning Co.
Aerosol OT	Sodium dioctyl sulfosuccinate	American Cyanamid Co.
Aerosol AY	Sodium diamyl sulfosuccinate	American Cyanamid Co.
Stearic Acid	Fatty acid	Emery Industries, Inc.
Cellophane	Cellulose film	E. I. du Pont de Nemours Company, Inc.
Tincture of Green Soap	Soap	Drug store grade
Oleic Acid	Fatty acid	Emery Industries, Inc.

WCRTR-4

9 March 1955

Monomer-Polymer, Inc.
Attn: Dr. B. D. Halpern
Leominster, Massachusetts

Gentlemen:

Reference is made to your letter dated 18 February 1955.

Preliminary test data have been acquired on the two 15"x12"x0.175" ethyl acrylate-polyvinyl siloxane interlayer sheets which were furnished gratis to this Center. The results are as follows:

1. Haze = 5.2%. Elimination of foreign matter and waviness will significantly reduce the haze. Foreign matter adheres to sticky interlayer surface.

2. Tensile strengths and percent elongation at elevated temperatures. An Instron with an inclosed temperature jacket was used. The rate of pull was five inches per minute with an initial two inches between the jaws. Specimens were loaded with 0.4-1.0 pounds at the beginning of the soaking period because the interlayer specimens relax upon heating.

<u>Specimen Number</u>	<u>Temp.</u>	<u>Soaking Time at Temp.</u>	<u>Tensile Strength</u>	<u>Percent Elongation</u>
1	75° F	15 Min.	150 psi	285
2	"	"	160 psi	312
3	150° F	19 "	51 psi	144
4	"	15 "	86 psi	162
5	200° F	"	74 psi	140
6	"	"	61 psi	109
7	250° F	"	55 psi	140
8	"	"	56 psi	97

3. Thermal stability tests. 1.5"x3"x0.175" specimens were conditioned for 3 days at 75° F and 25% relative humidity. Heating was conducted in a preheated forced air oven.

- a. 250° F for 17 hours. Very slight yellow tint; sticky; retained shape.
- b. 300° F for 5 hours. Slight yellow tint.
- c. 350° F for 3 hours. Yellowed. Color would be objectionable in a laminate.
- d. 400° F for 30 minutes. Yellowed; retained shape.
- e. 450° F for 30 minutes. Deep yellow; retained shape.
- f. 500° F for 30 minutes. Charred, dark brown, retained shape.

4. Low temperature flexibility. Material was brittle around 0° C and lower.

WADC TR 55-296

Contracts

Hq. WADC (WCRTR-4)

9 March 1955

Monomer-Polymer, Inc.

Any comments or suggestions on the above test results will be appreciated.

Future correspondence in this matter should be referred to the attention symbol, WCRTR-4.

Sincerely,

s/s Floyd J. Wilcox
Floyd J. Wilcox
Capt., USAF
Project Officer, Plastic Products Section
Organic Materials Branch
Materials Laboratory
Directorate of Research

October 13, 1954

Monomer-Polymer, Inc.
Leominster, Massachusetts

When you phoned us yesterday we informed you that we were shipping to your attention via express two laminates made with the interlayer material shipped us by you and described in your letter to us of September 16, 1954.

Following your recommendations as outlined in your letter of September 25th we laminated the interlayer material without the use of an adhesive. The technique employed for laminating, while employing heat and pressure, was one developed for handling soft interlayer materials such as the samples presented. As a matter of fact, we tried a number of techniques before finalizing the one employed in making the samples sent you.

It was our observation that the laminates, on standing around after the lamination process was complete, underwent separation in numerous areas, indicating lack of adhesion between the glass and interlayer surfaces.

These observations were confirmed by conducting what we call a "hammer crush test" at both normal and 0° temperature.

As of this date we have only conducted a few tests on similar laminates but we do plan to carry out others, the results of which will be reported to you at a later date.

We heated one 6" x 6" sample at 350° for one hour. The glass outer faces under this heat treatment separated from the interlayer over a substantial area. It was difficult because of the initial condition of the laminate to state whether or not bubbling of the interlayer occurred as a result of this heat treatment. Examination of the sample after removal from the oven did show localized discoloration of the laminate to a pale amber color.

As indicated to you over the telephone, we shall be happy to further cooperate with you in your interlayer development work and as outlined above shall report on any further observations we make when other tests have been completed.

Very truly yours,

LIBBEY-OWENS-FORD GLASS COMPANY

s/s J. D. Ryan
Associate Director of Research

JDRyan/v

October 26, 1954

Monomer-Polymer, Inc.
Leominster, Massachusetts

Since our last letter to you, we have now tested two 12" x 12" laminates prepared with the acrylic interlayer sheeting you furnished us recently.

One 12" x 12" laminate was impacted at 0°F employing a 7-3/4 lb. steel ball falling freely from a distance of 18 feet (this size ball and the distance employed were chosen on the basis of the performance of a comparable thickness of polyvinyl butyral plastic interlayer). On impact, large pieces of glass were thrown for distances up to 10 feet, indicating little or no adhesion between the interlayer and the glass. The interlayer material was punched out of the laminate free of glass over a 6" diameter. This indicates to us embrittlement of the interlayer at temperatures as low as 0°F.

A second 12" x 12" laminate was impacted under the same conditions at 120°F. Large fragments of glass were freed from the interlayer on impact, being thrown distances of 6 feet. While the interlayer did not puncture under these conditions, the laminate folded up allowing the ball to pass through the break-testing wooden supporting frame.

As reported earlier, we feel that the laminates produced have little, if any, adhesion. As indicated to you over the phone, we shall be glad, however, to laminate additional material when you make it available employing adhesives.

Yours very truly,

LIBBEY-OWENS-FORD GLASS COMPANY

s/s J. D. Ryan

Associate Director of Research

JDRyan/v

Contrails

COPY

P I T T S B U R G H

PLATE GLASS COMPANY

L. A. Keim, Technical Representative
PRODUCT DEVELOPMENT DEPARTMENT
Glass Division

GENERAL OFFICES: GATEWAY CENTER
420 Fort Duquesne Boulevard
Pittsburgh 22, Pennsylvania

April 18, 1955

Monomer-Polymer
Chemical Division
The Borden Company
Leominster, Massachusetts

Gentlemen:

Our test results on the three 12" x 12" experimental sheets of ethyl acrylate-vinyl polysiloxane interlayer material you submitted may be summarized as follows:

1. The sheeting was tested only in glass laminations.
2. With no adhesive on the glass, considerable delamination resulted in samples broken at 0° F.
3. Adhesion at 0° F was not improved using any of our "usual" laminating adhesives.
4. Any lamination that was made failed completely in a 400° F oven test.

Very truly yours,

L. A. Keim

LAK:cte

cc: Commander
Wright Air Development Center
Wright-Patterson Air Force Base, Ohio
Attn: WCRTR-4

WADC TR 55-296

-22-

TABLE I
Bulk Polymerizations of Ethyl Acrylate to Syrup

Run No.	EA # Monomer	Sil-oxane	Catalyst	Induction Period Min.	Reaction Period Temp. °C.	Notes
EA-PVS-1	300 ml.	0	0.20 g.** Diacetyl - UV irradi. 15 watt 6" away	130	35 52	Rapid reaction caused fluid to gel rapidly. Yellow, rubbery solid produced. No PVS added.
EA-PVS-2	338	3 g.	0.25 g. Diacetyl - UV irradi.	135	12 41.5	Rapid reaction-syrup viscosity = 14,000 cps @ 35°C. % Solids = 4.2. Syrup was yellow.
EA-PVS-3	290	3	0.20 g. Diacetyl - UV irradi.	80	3 41.5	Rapid reaction-yellow syrup was produced with viscosity of 1750 cps @ 24°C.
EA-PVS-4	300	3	None - Thermal	60	10 85	No reaction at 41 and 50°C. Slow reaction at 60°C. Fast reaction at 85°C. Clear syrup with viscosity of 2450 cps @ 65°C.
EA-PVS-5	310	3	None - Thermal	65	10 74	Fast reaction - clear syrup with viscosity of 5000 cps @ 42°C.

*Ethyl Acrylate Rohm & Haas redistilled
**Diacetyl -Lucidol Division-Agene-Novedel Corp.

TABLE II
Experimental Cast Interlayers

Cell No.	Cell Construction	Parting Agent	Interlayer Material	Irradiation 15 Watt UV Lamp, 6" away	Cure	Results
C-1	6" x 6" glass plates-vellum gaskets and "C" clamps	Solricin 535	EA-PVS-3	2 hrs.	16 hrs. @ 80°C. 1 hr. @ 105°C.	Material uncured, did not sheet out.
C-2	"	"	"	25 hrs.	None, room temp. for 4 days	35 mil clear sheet, soap spots on layer, good parting.
C-3	"	"	"	20 hrs.	"	1/8 in. thick clear sheet, soap spots on 1 layer.
C-4	"	"	"	24 hrs.	16 hrs. @ 67°C. 24 hrs. @ 72°C.	Partially cured interlayer, only portion re-covered as solid material, remainder evaporated.
C-5	Wire rod spacers gelatin seal 6"x6" glass plates	"	"	52 hrs.	24 hrs. @ 70°C.	Clear, unbroken sheet 3/4 mils thick
C-6	"	Bomb-Lube Price-Driscoll Corp.	EA-PVS-4	None	0.06% Benzoyl Peroxide Cat., 40 hrs. @ 60°C.	Clear material, 3/4 to 40 mils, cell leaked most of syrup. Good parting.

TABLE II (Cont'd.)

Experimental Cast Interlayers

Cell No.	Cell Construction	Parting Agent	Interlayer Material	Irradiation 15 Watt UV Lamp, 6" away	Cure	Results
C-7	Wire rod spacers gelatin seal 6"x6" glass plates	Dow-Corning Silicone grease	EA-PVS-4	None	0.06% Benzoyl Peroxide Cat. @ 30 hrs. @ 57°C.	Clear sheet-cloudy on one side parting not quite so easy.
C-8	"	Atlas Tween 61	"	None	"	Clear sheet-parting not quite so easy.
C-9	Rubber spacers gelatin seal	Glycerin	"	None	0.06% B.P. 24 hrs. @ 57°C.	Air bubbles and yellow at edges
C-10	"	Slipicone Dow-Corning.	"	None	"	Easy parting-sheet was cloudy.
C-19	6x6x1/8" Parchment	Aerosol OT	98.98% EA syrup 1% PVS 0.02% Bz ₂ O ₂	700 cps	None	Clear, some bubbles and smudges
C-20	6x6x1/8" Parchment	Aerosol OT	98.99% EA syrup 1% PVS 0.01% Bz ₂ O ₂	1500 cps	None	Clear, some bubbles and smudges

TABLE II (Cont'd.)

Experimental Cast Interlayers

Cell No.	Cell Construction	Parting Agent	Interlayer Material	Irradiation		Results
				15 Watt UV Lamp, 6" away	Cure	
131	6x6x1/8" parchment	Aerosol OT	98.99% EA Monomer 1% PVS 0.01% Bz ₂ O ₂	None	24 hrs. @ 40°C	Polymerized in 22 hrs.
				None	15 hrs. @ 50°C	Parted readily, clear
				None	24 hrs. @ 60°C	smooth sheet, no bubbles.
				None	4 hrs. @ 80°C	
136	15x15x1/8" parchment	Aerosol OT	"	None	20 hrs. @ 40°C	Monomer 2 days old, fully polymerized 44 hrs., opaque in spots, no fisheyes.
				None	24 hrs. @ 50°C	
				None	48 hrs. @ 55°C	
				None	24 hrs. @ 85°C	
144	15x15x1/4" parchment	Aerosol OT	"	None	69 hrs. @ 40°C	Monomer 13 days old, a clear sheet obtained, (0.165 ± .030") washing soapy water removed smudge marks. Laminated at Libbey-Owens-Ford.
				None	25 hrs. @ 45°C	
				None	48 hrs. @ 50°C	
				None	4 hrs. @ 65°C	
146	15x15x1/4" parchment	Aerosol OT	EA syrup only	None	72 hrs. @ 40-45°C	Filled within 2" of top.
				None	25 hrs. @ 50°C	35 hrs. polymerized at bottom but not top.
				None	48 hrs. @ 65°C	
				None	8 hrs. @ 80°C	Bubbly sheet obtained.

TABLE II (Cont'd.)

Experimental Cast Interlayers

Cell No.	Cell Construction	Parting Agent	Interlayer Material	Irradiation		Results
				Lamp, 6" away	15 Watt UV	
149	15x15x1/4" parchment	Aerosol OT	98.99% EA	None	135 hrs. @ 42-45°C	Monomer 5 days old, a clear sheet obtained (0.180 ± .005") not gelled after 39 hrs. Fisheyes and lakes present.
			1% PVS 0.01% Bz2O2		12 hrs. @ 65°C	
150	15x15x1/4" parchment	Aerosol OT	500 cps syrup, 1% PVS, No. Cat.	None	84 hrs. @ 45°C 12 hrs. @ 65°C	Clear, bubbles, distorted, weak spots (.210 ± .020"), Breaks

TABLE III
Large Interlayers, Preparation

Cell No.	Cell Type	Gas Content	PVS Content %	Benzoyl Peroxide %	Parting Agent	Syrup Viscosity Cps.	Heat Treatment
151	48x48x1/4" Parchment	Some	1	0.01	Aerosol OT	Monomer	64 hrs. 41-50°C. Average 48°C. 23 hrs. 65°C. 24 hrs. 85°C.
154	48x48x1/4"	Some	1	0.01	Aerosol OT	Monomer	4 days 41-50°C. Average 46°C. 8 hrs. 65°C. 16 hrs. 85°C.
230	40x40x1/4" Thin walled Polyethylene Gasket	Saturated, Air	1	0.01	Aerosol OT	1550	72 hrs. 47 ± 2°C. 8 hrs. 57 ± 2°C. 8 hrs. 67 ± 2°C. 8 hrs. 80 ± 2°C.
231 Rex No. 2	"	"	1	0.01	Aerosol OT	1180	"
232 Rex No. 3	"	"	1	0.01	Aerosol OT	Syrup similar to cells 230 and 231	"
233 Rex No. 4	"	Degassed	1	0.01	Aerosol OT	Monomer	"
234 Rex No. 5	"	Degassed	1	0.01	Aerosol OT	Monomer	"

TABLE IV
Large Interlayers, Characteristics

Cell No.	Approximate Length, Width and Weight	Thickness, in.	Remarks	Description of Interlayer
151	Cut to 37 x 42.5" 16# 7 oz.	0.200 ±.010	Gelled in 24 hrs. Top glass plate removed after 64 hours.	Clear, smooth, Lakes. Monomer 4 days old.
154	Cut to 37 x 42.5" 15# 3 oz.	0.195 ±0.012	One plate removed after 4 days but replaced for final cure.	Many pinholes, protrusions, lakes, some haze, more flaws than Cell 151.
230 Rex No. 1	38.5 x 38.5" 13# 3 oz.	0.190 ±.010	13# 7 oz. charge. Lakes present, liquid at edges, after 42 hrs.	Clear, lakes, fisheyes, few black specks, 7 bubbles, 1-1" cut in center.
231 Rex No. 2	38 x 38" 13# 8 oz.	0.198 ±.018	13# 8 oz. charge polymerized in center between 18 and 42 hr. Lakes present, fluid at edges at end of 42 hrs.	Clear, except for 11 x 9" white bled area in center. Lakes, fish-eyes, 6 pieces of dirt.
232 Rex No. 3	38.5 x 38.5" 13# 13 oz.	0.183 ±.015	13# 13 oz. charge. Lakes present, liquid at edges at end of 42 hrs.	Clear, lakes, fisheyes, 5 large pieces of foreign matter.
233 Rex No. 4	38.5 x 34" 12# 3 oz.	0.192 ±.024	13# 7 oz. charge. Cell leaked, polymerized in center within 18 hrs. Lakes within 42 hrs., fluid at edges.	Clear, except for 12 x 8" white bled area in center, fisheyes, lakes, 1 - 2-1/2" cut, many 1" cuts, few large bubbles.
234 Rex No. 5	38 x 34" 10# 10 oz.	0.158 ±.030	13# 7 oz. charge. Polymerized in center within 18 hrs. Lakes within 42 hrs., fluid at edges.	Clear, except for 11 x 5" white bled area in center, lakes, fisheyes, few 1" cuts, many bubbles near center

TABLE V
Further Cast Interlayers

Cell No.	Cell Type	Syrup Viscosity and Type	Gas Content	PVS Content %	Benzoyl Peroxide %	Parting Agent	Ultra-Violet and Heat Treatment	Final Condition of Cell	Remarks
210	15 x 15" Parchment 1/2 full	836 cps Benzoin	Degassed	1.0	0.01	None	7 days 34-45°C 1 day 45-84°C	Cloudy, Lakes, Fisheyes	Polymerized too fast first 15 hrs. Top 1" did not polymerize until 6th day.
211	15 x 15" Parchment Benzoin	700	Air Saturated	1.0	0.01	OT	6 days 34-45°C 1 day 45-84°C	Clear, Lakes, Fisheyes	Not polymerized completely during first 3 days, top position not polymerized in first 5 days.
212	15 x 15" Mylar Covered PVC 3/4 Full	700 Benzoin	Air Saturated Day Before Charging	1.0	0.01	OT	5 days 34-45°C 1 day 45-84°C	Mostly clear, Lakes, few small fisheyes	Cell cracked during cure. Polymerized between 2nd and 5th day.
213	6 x 6" Mylar Covered PVC Full	700 Benzoin Plus 1% Added H ₂ O	Degassed	1.0	0.01	OT	4 days 34-45°C 1 day 45-84°C	Opaque Discarded	Polymerized too fast in places, too slow in others. Worst of Cells 210, 211, 212, 213.

TABLE V (Cont'd.)

Further Cast Interlayers

Cell No.	Cell Type	Syrup Viscosity and Type	Gas Content	PVS Content %	Benzoyl Peroxide %	Parting Agent	Ultra-Violet and Heat Treatment	Final Condition of Cell	Remarks
214	6 x 6" Mylar Covered PVC Full	1350 Benzoin	Degassed	1.0	None	OT	1 1/2 hrs. under 300 Watt UV at 9" 4 days 80°C 1 day 105°C	Yellow at edges, very clear Few lakes, some shrinkage, monomer odor, sticky, no fisheyes	Cooled front of cell with cold water first 1 1/2 hrs. Syrup did not polymerize under portion shielded from U.V. by lettering of cell number. Interlayer stripped from glass before heat cure. Turned yellow during 105°C cure.
215	6 x 6" Mylar Covered PVC Full	1350 Benzoin	Degassed	1.0	None	OT	2 hours under 300 watt UV at 9 to 6" 4 days 80°C 1 day 105°C	Yellow at edges. No fisheyes, clear, few lakes, some shrinkage, mono-mer odor, sticky.	Cooled back of cell with cold water first 2 hrs. Interlayer stripped from glass before heat cure. Yellow tint appeared during 105°C cure.

TABLE V (Cont'd.)

Further Cast Interlayers

Cell No.	Cell Type	Syrup Viscosity and Type	Gas Content	PVS Content %	Benzoyl Peroxide %	Parting Agent	Ultra-Violet and Heat Treatment	Final Condition of Cell	Remarks
216	15 x 15" Parchment Benzoin 3/4 Full	1350	Degassed	1.0	None	None	3 1/2 hrs. under 300 watt UV at 36"	Clarity good. Lakes present on uncooled side.	Although cell was cooled by fan on one side, cell & surrounding air became heated above room temperature. Cell was discarded since glass could not be removed from interlayer.
217	15 x 15" Mylar Covered PVC 3/4 Full	4400 Benzoin	Degassed	1.0	None	OF	5 days 37°C 1 day 47°C 3 days 80°C 1 day 105°C	Cloudy, bubbles, lakes, few fisheyes, clear in places, many wave like optical imperfections.	Polymerization control very poor. Air plus 0.01% benzoyl peroxide syrup cells polymerize more smoothly. Much shrinkage from cell gasket occurred.

TABLE V (Cont'd.)

Further Cast Interlayers

Cell No.	Cell Type	Syrup Viscosity and Type	Gas Content	PVS Content %	Benzoyl Peroxide %	Parting Agent	Ultra-Violet and Heat Treatment	Final Condition of Cell	Remarks
218	15 x 15" Mylar Covered PVC 3/4 Full	4400 Benzoin	Degassed	1.0	0.01	OT	3 days room temperature 2 days 37°C 3 days 80°C 1 day 105°C	Lakes, shrinkage marks, some cloudiness, few fisheyes, yel. cast when observed from edges.	Cell cracked during assembly. Polymerized partially from sunlight. Became dirty during separation of broken glass from interlayer.
219	15 x 15" Mylar Covered PVC 3/4 Full	4200 Benzoin	Degassed	1.0	None	OT	4 days under 300 watt UV at 48" 3 days 80°C 1 day 105°C	Tacky, clear but lakes & waves present. Cell has a yellowish cast. No fisheyes but many bubbles.	Cell cooled front & back with fans. At the end of 8 hrs. all but the top 1/2" had polymerized. Clamps kept tight first four days. Clamps removed during heat cure. Edges uneven. Edges contained bubbles. Lakes appeared during heat cure.

TABLE V (Cont'd.)

Further Cast Interlayers

Cell No.	Cell Type	Syrup Viscosity and Type	Gas Content	PVS Content %	Benzoyl Peroxide %	Parting Agent	Ultra-Violet and Heat Treatment	Final Condition of Cell	Remarks
220	15 x 15" Mylar Covered PVC Full	4200 Benzoin	Air Saturated	1.0	None	OT	Overnight room temperature, 4 days under 300 watt UV at 48" 6 days 80°C 16 hrs. 105°C	0.146" thick, clear, lakes at edges, no yellow cast, no fish-eyes.	Cooled front & back until placed in oven. After U.V. treatment cell was lake free in the center & on U.V. exposed surface. After 6 hrs. at 80°C cell was cloudy but became clear during cure. Cell cracked during 80°C aging. (Clamps were tightened with cell hot, & cooling during inspection may have set up stresses.) Difficulty in removing cracked glass. This is the best cell of those listed above.

TABLE V (Cont'd.)

Further Cast Interlayers

Cell No.	Cell Type	Syrup Viscosity and Type	Gas Content	PVS Content %	Benzoyl Peroxide %	Parting Agent	Ultra-Violet and Heat Treatment	Final Condition of Cell	Remarks
221	15 x 15" Mylar covered rubber tubing full	400 Diacetyl	Air Saturated	1.0	None	OT	4 days under 300 watt UV at 9 ft. 6 days 80°C 1 day 105°C	Many lakes, extremely clear, some dust but no large fish-eyes, monomer odor, yellow color.	Lakes formed 1 1/2 hrs. after placed under U.V. probably due to sunlight although cell remained cool at all times.
222	15 x 15" Mylar covered PVC. 1 corner glass broken	630 No benzo- in or diacetyl	Air Saturated	1.0	0.01	OT	3 days 46°C 18 hrs. 55°C 18 hrs. 65°C 6 hrs. 72°C	Clear, many lakes, large fisheyes.	Aerosol spread with squeegee, & glass plates dried overnight at 105°C. before filling.
223	15 x 15" Mylar covered rubber tubing	0.03% Diacetyl Syrup	Air Saturated	1.0	None	OT from EA solution	5 days under 300 watt UV at 9 ft. 3 days 85°C 2 hrs. 105°C	Brown discoloration where Mylar seal leaked. Cell clear in center. (1 small lake) small fisheyes, shrinkage, waves, lakes & bubbles at edges.	Extra dry monomer used to prepare syrup. Cell polymerized between 16th and 22nd hour (when sunlight was in room but with no direct rays of sun on cell). Lakes not deep. Softer gas-ket appears to lessen lakes in this size cell.

TABLE V (Cont'd.)

Further Cast Interlayers

Cell No.	Cell Type	Syrup Viscosity and Type	Gas Content	PVS Content %	Benzoyl Peroxide %	Parting Agent	Ultra-Violet and Heat Treatment	Final Condition of Cell	Remarks
224	6 x 6" PVC	EA Monomer	Air Saturated	1.0	0.01	OT	7 days 46°C	No inter-layer	Gasket had softened, all monomer leaked out.
225	6 x 6" Polyethylene film covered PVC	EA Monomer	Air Saturated	1.0	0.01	OT	7 days 46°C	No inter-layer.	Gasket softened, all monomer leaked out.
226	6 x 6" Polyethylene film covered rubber.	630 No benzoin no diacetyl	Air Saturated	1.0	0.01	OT	5 days 46°C	No inter-layer	Syrup discolored and leaked out from a bulge in gasket wall.
227	6 x 6" Polyethylene	630 No benzoin or diacetyl	Air Saturated	1.0	0.01	OT	61 hrs. 47°C 39 hrs. 105°C	Slight yellow color, cell had shrunk from edges.	Polyethylene tubing does not inhibit polymerization.

TABLE V (Cont'd.)

Further Cast Interlayers

Cell No.	Cell Type	Syrup Viscosity and Type	Gas Content	PVS Content %	Benzoyl Peroxide %	Parting Agent	Ultra-Violet and Heat Treatment		Final Condition of Cell	Remarks
							Days	Temp		
228	15 x 15" Polyethylene tubing	630 No Benzoin or Diacetyl	Air Saturated	1.0	0.01	AY	5 days	46°C	Cell 0.177	Clamps kept tight at all times. Polymerized at 44 hrs. Lakes appeared at 48 hrs., but disappeared by end of cycle. Inside of gasket had ruptured. Aerosol AY as parting and washing agent. Glass removed readily when still hot.
							12 hrs.	55°C	± 0.007 ^m	
							8 1/2 hrs.	65°C	in thickness,	
229	15 x 15" Polyethylene tubing	Extra dry EA mono-mer plus 0.1% Ergy Stearic Acid.	Air Saturated	1.0	0.01	Stearic Acid Monomer	3 days	47°C	Shrinkage, fisheyes,	Cell was not polymerized after 3 days 47°C. Polymerized too fast at 55°C or in the first six hrs. at 65°C. Glass released readily hot.
							12 hrs.	55°C	many lakes, many stress marks, waves.	
							8 1/2 hrs.	65°C		
							6 1/2 hrs.	85°C		

TABLE VI
Comparison of Physical Properties of WADC 54-57 Interlayers
With Those Produced Under This Contract

Sample Description	C-3		C-5		C-19 0.02% Bz ₂ O ₂ , Syrup	C-20 0.01% Bz ₂ O ₂ , Syrup	C-131 0.01% Bz ₂ O ₂ , Monomer	#136 0.01% Bz ₂ O ₂ , Monomer	#146(c) EA Syrup Only, No PVS	#150 Syrup Catalyst
	Ultra- Violet Polymer- ization	Ultra- Violet Polymer- ization	Ultra- Violet Polymer- ization	Ultra- Violet Polymer- ization						
% Weight Loss after 4.2 hrs. @ 350°F.	2.0%	2.7(a)	2.6	2.6%	2.3 (a)	2.8 (a)	1.6	1.6	1.6	2.1
Shore A-2	27	27	30	25	25	23	23	23	23	24
Hardness	27	27	30	25	25	23	23	23	23	24
Shore A-2	21	23	25	18	20	20	20	20	23	20
Hardness After 4.2 hrs. @ 350°F.	23	25	25	18	20	20	20	20	21	20
Color After Heating 4.2 hrs. @ 350°F.	Yellow	Yellow	Yellow	Yellow	Amber spots on surface					
Swelling in Ethylene Dichloride	710	600	32	580	801	930	59	62.5	50	60
Weight %	600	600	32	520	535	920	59	62.5	50	60
Volume %	32	65.0	32	75	112	43	59	62.5	50	60
Tensile Strength	129(a)	171	129(a)	118(a)	118(a)	64(a)	250	183	183	245
After Heating 4 hrs @ 350°F.	171	171	171	118(a)	118(a)	64(a)	252	265	265	245

REMARKS:
a. 5 hrs. heating.
b. Sample 1/5 as thick as others.
c. Only cell shown without 1% PVS.