

**THE BONDING OF PLASTICIZED POLYVINYL
CHLORIDE SHEET TO METAL**

by

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I. INTRODUCTION

In recent years, rapid advances have been made in bonding plastic sheeting to metal by means of synthetic adhesive. The product, a pre-finished material, has the strength of the base metal and exterior finish of the plastic. It can be deep-drawn, crimped, punched or stamped as ordinary metal, and because of the properties of the plastic, it offers a number of advantages over paint, varnish and lacquer coatings. Plastic-metal laminates have found applications in radio and television cabinets, furniture, chemical containers and automobiles. In fact, this is a new branch of the surface coating industry, and is now of growing importance.

Although the practice of bonding plastic to metal has advanced to a considerable extent, little is known about the basic principles. In practice, many questions may arise as to optimum working conditions, the nature of the bond, effect of deformation of the product, and the factors affecting the bond strength. A knowledge of the factors involved would be helpful in the effort to improve bonding practice. Since polyvinyl chloride has exceptional resistance to corrosive chemicals, moisture and abrasion, and has high dielectric strength and other desirable

electrical characteristics, it is an excellent coating material. But the very properties, chemical inertness, toughness and insolubility, which make polyvinyl chloride so valuable, make it difficult to bond to metal.

It was the purpose of this investigation to study the factors affecting the bond strength between polyvinyl chloride sheeting and metal.

II. LITERATURE REVIEW

Bonding⁽¹³⁾ is the process of joining surfaces by adhesive action, and the bodies held together are called adherends. Owing to the dissimilarity between the metallic bond and other types of valence, it is not surprising that the bonding of plastic to metal is a comparatively recent development. A plastic sheet can be bonded to a metal through an intermediate layer of adhesive which holds them together by surface attachment. It is said⁽⁸⁾ that by selecting a suitable adhesive and proper working condition, good adhesion can be obtained.

Nature of the Bond

According to Epstein⁽⁶⁾ the strength of the bond is dependent upon a combination of two forces. These are cohesion and adhesion. In the case of plastic to metal bonding, cohesion is the force within the adhesive itself holding the molecules together. It is usually a strong covalent bond. The second force, adhesion, is the molecular attraction which is exerted across the surfaces of contact between adhesive and metal and adhesive and plastic. There are two types of adhesion, mechanical and specific. Mechanical adhesion is the phenomenon which occurs when tendrils

of adhesive enter into the pores of the base material. Specific adhesion, on the other hand, occurs between smooth, dense surfaces, and depends upon the chemical affinity of the adhesive and the adherend for one another. Since metals are relatively non-porous, specific adhesion is of greater importance than mechanical anchoring. In dealing with the nature of the bond between adhesive and metal, Epstein postulated that there were unbalanced forces on the metallic surfaces. These forces had an affinity toward molecular groups of certain adhesives, especially those which were highly polar.

Schmidt and Marlies⁽¹⁸⁾ said the metal adhesion depended upon the formation of a two-dimensional metal compound at the interface. For high strength bonds this compound would be of primary valence character. Bonds of lower strength could be those of secondary valence. They pointed out in the case of polyvinyl acetate, "if this adhesive is applied between metal adherends and the joint is effected via a cold pressing technique, a joint strength of the order of 200 pounds per square inch results. Probably the adhesion at the metal-adhesive interface depends upon the orientation of the carbonyl group toward the metal, a secondary effect. By contrast, if hot pressing is employed, a joint strength of 2,000 pounds per square inch is

attained. Perhaps acetic acid released during heating forms a two-dimensional metal acetate, which is linked by hydrogen bonding to the acetate radicals in the adhesive."

Factors Affecting Bond Strength

The ultimate goal in the bonding of plastic sheet to metal is to obtain a permanent bond. In order to attain this end, several factors must be taken into consideration.

Choice of Adhesive. An adhesive ⁽¹⁵⁾ should be selected in accordance with the chemical nature and thermal expansion coefficient of the adherend surface. High specific adhesion of the set adhesive to the adherend is essential.

Thickness of Adhesive. The adhesive layer ⁽⁶⁾ should be kept as thin as possible but sufficiently thick to eliminate possibility of a starved joint. Generally the thickness is 0.002-0.006 inch.

Surface Conditions. It is essential ⁽⁵⁾ that the surface of the metal be free of foreign matter. Chemical cleaning and degreasing with suitable solvents are generally desirable. Solvent wash, hot alkaline cleaning and hot acid pickling, each offers its own advantage, the choice

being contingent upon the ultimate physical properties required. Sanding of the surface with a fine grade of sand paper or light sandblasting is often employed to roughen the bonding surface while simultaneously cleaning it. The effect of a roughened surface is to increase the ratio of the actual to the apparent bonding area and also permit a greater degree of mechanical adhesion.

Crewdson⁽²⁾ used different acids to etch low carbon steel for bonding to plasticized polyvinyl chloride sheeting. It was observed that the metal untreated with acid had the smallest surface roughness, yet the greatest peel strength. He explained this fact from the standpoint of salt formed. When the metal was treated with acid, a metallic salt was formed on the surface. If this salt was not completely removed after etching, some of the adhesive would adhere to the salt instead of base metal, and thus would result in a weaker bond.

Fisher and Bobalek⁽⁷⁾ pointed out that the typical faults of surface condition for bonding are:

1. Air pockets,
2. Poor coverage at sharp projections,
3. Non-metallic inclusions that ruin adhesion,
4. A dirt or grease spot causing a pulling away of the adhesive to form a pore or "fisheye".

Any of these faults on the metal surface would cause considerable decrease in bond strength.

In addition to these faults, air normally contains variable amounts of moisture, organic matter, carbon dioxide and sulfurous gases which would cause tarnishing, rusting, oxidation, hydrolysis, "filming", "aging" and "grease pickup" on the surface. It is advised ⁽¹⁴⁾ that surfaces be freshly prepared.

Methods of Bonding. There are four general methods of bonding plastic sheet to metal. These are:

Solution Adhesives. A bond ⁽²¹⁾ can be formed by the use of a resin compound dissolved in a solvent. In use, the solution forms a bond with each of the surfaces being joined, and the success of this method depends upon the holding power of the solution itself. These adhesives set very slowly, and have little strength immediately after application. In order to obtain a faster set, a short forced dry at elevated temperature is satisfactory. Various methods can be used in plastic-metal bonding. One of these ⁽¹⁹⁾ uses two layers of adhesives. The first layer, or primer coat, is applied to the metal adherend, followed by a layer of another adhesive. The primary coat is usually baked, thus giving excellent adhesion to the metal.

The following formulations for solution adhesive and adhesive primer, in weight per cent, were given by Bakelite Corporation (24).

Solution Adhesive

	XL-5174
Resin VYWW	10%
Methyl Ethyl Ketone	36%
Cyclohexanone	36%
Propylene Oxide	<u>18%</u>
	100%

Adhesive Primer

	XL-5252
Resin VMCH	28%
Flexol Plasticizer DOP	6%
Methyl Ethyl Ketone	20%
Methyl Isobutyl Ketone	20%
Propylene Oxide	1%
Toluene	<u>25%</u>
	100%

Pressure-Sensitive Adhesives. Pressure-sensitive adhesives (22), typified by rubber cements, are used in plastic-metal bonding because of their

high initial strength, and the wide range allowable in curing time. This type of adhesive depends upon the tackiness of a solvent-free film to adhere to the surfaces. Tackiness is usually obtained through the use of plasticizer or soft resins in the compound.

Heat-Melt Adhesives. Instead of using a solvent ^(4,17), an adhesive can be applied in liquid phase by heat-melt. It contains little or no solvent. This method has the advantages of quick-setting.

Reaction-Sensitive Adhesives. This type of adhesive ^(3,17) is compounded with catalyst prior to bonding. During curing, reaction takes place and a strong bond is obtained.

Conditions of Bonding. Pressure, temperature and time of curing have influence upon the strength of the bond. According to Schmidt and Marlies ⁽¹⁶⁾ the pressure used in bonding varies from 1 to 250 pounds per square inch or greater. The temperature varies from below 68° to 212° F. The time of curing may vary from a few minutes to a few days. The optimum conditions will depend upon the nature of the adherends and the adhesives.

Previous Work

Regarding the bonding of plasticized polyvinyl chloride sheet to metal, nothing has been found in the literature. However, some similar work has been done which may be used as reference.

Blake⁽¹⁾ suggested a method for bonding plasticized polyvinyl chloride paste to mild steel. He used two layers of vinyl copolymer adhesive, one of which was baked on metal, the other applied as solution. Upon the adhesive layer, a semi-rigid (lightly plasticized) polyvinyl chloride sheet was placed and then followed by the polyvinyl chloride paste. The polyvinyl chloride sheet acted as a barrier to prevent the migration of plasticizer from the paste to the adhesive. By jelling the paste in contact with the semi-rigid sheet a durable bond was said to be obtained.

Torii⁽²⁰⁾ used a copolymer of 1,3-butadiene and vinyl cyanide dissolved in benzene and acetone as adhesive. This adhesive was coated on a lead surface, the solvents were allowed to evaporate off, and the "vinyl chloride" was applied without heating or pressing.

Adhesives for Metals

In general, a successful adhesive⁽¹⁸⁾ contains valence bonds and composition similar to those of the adherend. Although this is not possible with the metal, one should expect adhesives for metals to contain groups that readily form metallic compounds. This is borne out in fact. The followings^(1,6,10,12,19,23,25) are the successful metal adhesives:

Acrylic resins

Butadiene-acrylonitrile rubber

Epoxy resins

Furan derivatives resins

Phenolic resins

Polyamides

Polyesters

Polyurethane resins

Polyvinyl acetal

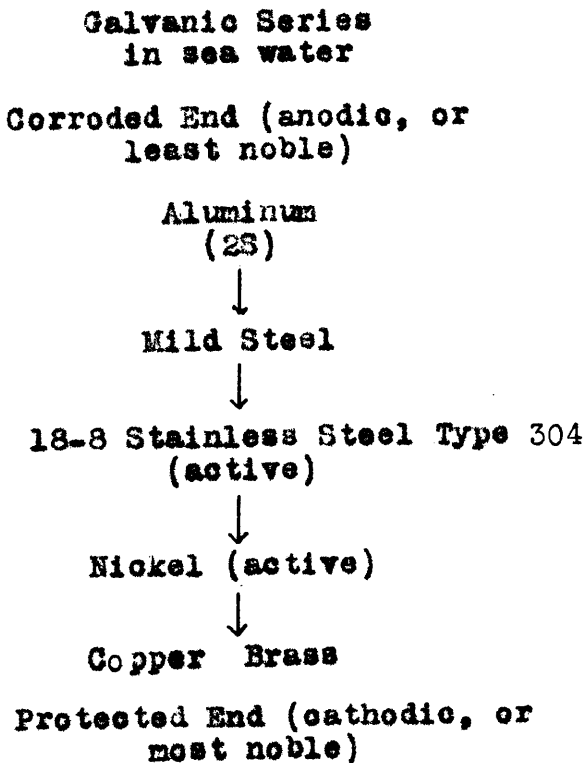
Polyvinyl acetate

Polyvinyl acetate-chloride copolymer

Silicones

Activity of Metals

Metals and alloys can be arranged in the order of their tendencies to form electrochemical or galvanic cells. In this arrangement, each metal or alloy is electropositive to any of those succeeding it, and electronegative to any of those preceding it. The farther apart the metals stand, the greater will be the galvanic tendency. Such an arrangement is termed galvanic series, it is useful to predict the probable direction of the galvanic effects and the relative chemical activity of the metals. The following galvanic series was given by Whitehead Metal Products Company (11).



The word "active" in the parenthesis means that the metal has a freshly prepared or active surface.

III. EXPERIMENTAL

This section is concerned with the purpose of the investigation, plan of experimentation, materials and apparatus used, method of procedure, and the data and results obtained in the investigation.

Purpose of Investigation

The purpose of this investigation was to study the factors affecting the bond strength between polyvinyl chloride sheeting and metal.

Plan of Experimentation

This investigation was carried out according to the following plan:

Literature Review. A survey of all available literature pertaining to the theories of adhesion, application of adhesives, methods of bonding, and metallurgy of bending and deep drawing was made to gain an insight into the work which had been done by previous investigators.

Metals used in this Investigation. The metals used in this investigation were aluminum, brass, copper, nickel, low carbon steel and stainless steel.

Adhesives used in this Investigation. Five different commercial adhesives were used in this investigation. These were:

Vinyl chloride-acetate resin, VYNW

Modified vinyl chloride-acetate resin, VMCH

Vinyl acetate resin, A-70

Vinyl alcohol-acetate resin, T-24-9

Vinyl alcohol-acetate resin, MA-28-18

Experimental Procedure. The specimens of metal sheet were cleaned to prepare active surfaces for bonding.

The adhesives were dissolved in suitable solvents and applied to the metals and plastic sheeting by brushing. Two layers were applied, one of which was applied to metal and dried in the atmosphere for 24 hours and the other one was applied to plastic and dried in the atmosphere until a greater part of solvent had been evaporated.

The plastic was then bonded to the metal with a hydraulic press. A pressure of 200 pounds per square inch was applied to the assembly to obtain a continuous glue line of uniform thickness. Heating was applied at the same

time to a temperature of 70°, 150°, 250° and 350° F. As soon as a pressure of 200 pounds per square inch was obtained the pressure was released.

The bonded products were tested for bond strength. A series of them were stretched to 10, 20 and 30 per cent elongation and the resulting products were tested for bond strength.

Materials

Acid. Hydrochloric, CP, reagent grade, lot No E-901013, assay 37-38 per cent, specific gravity 1.188-1.192. Obtained from General Chemical Division, Allied Chemical and Dye Corporation, New York, N. Y. Used to etch low carbon steel and stainless steel.

Acid. Hydrofluoric, reagent grade, lot No 71248, about 48 per cent. Obtained from J. T. Baker Chemical Company, Phillipsburg, N. J. Used to etch aluminum.

Acid. Nitric, CP, reagent grade, lot No 80726-E, assay 70 per cent, specific gravity 1.42. Obtained from Fisher Scientific Company, Silver Spring, Md. Used to etch aluminum.

Acid. Sulfuric, CP, reagent grade, lot No E-901009, assay 95.5-96.5 per cent, specific gravity 1.84.

Obtained from General Chemical Division, Allied Chemical and Dye Corporation, New York, N. Y. Used to etch brass, copper, nickel, low carbon steel and stainless steel.

Adhesive. Vinyl acetate resin, solution, grade A-70, 69-72 per cent by weight in acetone. Obtained from Bakelite Company, New York, N. Y. Used as adhesive for bonding plastic sheet to metal.

Adhesive. Vinyl alcohol-acetate resin, solution, grade MA-28-18, highest hydroxyl content, 28 per cent by weight in 82 per cent methyl acetate. Obtained from Bakelite Company, New York, N. Y. Used as adhesive for bonding plastic sheet to metal.

Adhesive. Vinyl alcohol-acetate resin, solution, grade T-24-9, lowest hydroxyl content, 24.5 per cent by weight in toluene. Obtained from Bakelite Company, New York, N. Y. Used as adhesive for bonding plastic sheet to metal.

Adhesive. Vinyl chloride-acetate resin, modified, powder, grade VMCH, tripolymer of vinyl chloride, vinyl acetate, and maleic acid in the ratio of 86:13:1. Obtained from Bakelite Company, New York, N. Y. Used as adhesive for bonding plastic sheet to metal.

Adhesive. Vinyl chloride-acetate resin, powder, grade VYNW. Obtained from Bakelite Company, New York, N. Y. Used as adhesive for bonding plastic sheet to metal.

Diethyl Phthalate. "Flexol Plasticizer DOP", chemical composition di-2-ethylhexyl phthalate, lot No TS20748. Obtained from Carbide and Carbon Chemicals Company, New York, N. Y. Used as plasticizer for modified vinyl chloride-acetate resin, VMCH.

Distilled Water. Obtained from Chemistry Department, Virginia Polytechnic Institute, Blacksburg, Va. Used to prepare etching solutions and to rinse metals after etching.

Metal Sheet. Aluminum, 2s-0 (soft), mill finish grade; B & S gauge No 20, thickness 0.032 inch, weight 0.452 pound per square foot. Obtained from Whitehead Metal Products Company, Inc., New York, N. Y. Used as metal to which plastic sheet was bonded.

Metal Sheet. Brass, soft, spinning and drawing grade; B & S gauge No 20, thickness 0.032 inch, weight 1.410 pounds per square foot. Obtained from Whitehead Metal Products Company, Inc., New York, N. Y. Used as metal to which plastic sheet was bonded.

Metal Sheet. Copper, cold rolled, annealed grade; B & S gauge No 20, thickness 0.032 inch, weight 1.50

pounds per square foot. Obtained from Whitehead Metal Products Company, Inc., New York, N. Y. Used as metal to which plastic sheet was bonded.

Metal Sheet. Nickel, cold rolled "A", annealed, plain finish grade; U. S. S. gauge No 22, thickness 0.031 inch, weight 1.44 pounds per square foot. Obtained from Whitehead Metal Products Company, Inc., New York, N. Y. Used as metal to which plastic sheet was bonded.

Metal Sheet. Steel, low carbon, cold rolled, carbon content 0.09-0.20 per cent; U. S. S. gauge No 20, thickness 0.037 inch. Obtained from O'Sullivan Rubber Corporation, Winchester, Va. Used as metal to which plastic sheet was bonded.

Metal Sheet. Steel, stainless, No 304, chromium 18 per cent, nickel 8 per cent; U. S. S. gauge No 22, thickness 0.031 inch, weight 1.313 pounds per square foot. Obtained from Whitehead Metal Products Company, Inc., New York, N. Y. Used as metal to which plastic sheet was bonded.

Plastic Sheet. Polyvinyl chloride, semi-rigid, embossed surface, about 22 per cent by weight tricresyl phosphate. Average thickness 0.0274 inch. Obtained from O'Sullivan Rubber Corporation, Winchester, Va. Used for lamination to metal sheet.

Plastic Sheet. Polyvinyl chloride, semi-rigid, smooth surface, about 22 per cent by weight tricresyl phosphate. Average thickness 0.036 inch. Obtained from O'Sullivan Rubber Corporation, Winchester, Va. Used for lamination to metal sheets.

Sodium Chloride. Granular, U. S. P. grade. Obtained from Merck and Co. Inc., Rahway, N. J. Used in acid solution to etch nickel.

Sodium Dichromate. Granular, U. S. P. grade. Obtained from Merck and Co. Inc., Rahway, N. J. Used in acid solution to etch brass and copper.

Sodium Hydroxide. Flake, 76 per cent, code No 2256. Obtained from General Chemical Division, Allied Chemical and Dye Corporation, New York, N. Y. Used in solution to etch aluminum.

Sodium Nitrate. Granular, CP, reagent grade, lot No 491892. Obtained from Fisher Scientific Company, Silver Spring, Md. Used in acid solution to etch nickel.

Solvent. Acetone, CP. Obtained from Commercial Solvents Corporation, Newark, N. J. Used as solvent for vinyl acetate resin.

Solvent. Cyclohexanone, reagent grade, lot No 34661. Obtained from Carbide and Carbon Chemicals Company,

New York, N. Y. Used as solvent for vinyl chloride-acetate resin.

Solvent. Methyl ethyl ketone, reagent grade, lot No 545265. Obtained from Carbide and Carbon Chemicals Company, New York, N. Y. Used as solvent for vinyl chloride-acetate resin.

Solvent. Methyl isobutyl ketone, reagent grade, lot No S22441. Obtained from Carbide and Carbon Chemicals Company, New York, N. Y. Used as solvent for vinyl chloride-acetate resin.

Solvent. Propylene oxide, reagent grade, 100 per cent, lot No S74551. Obtained from Carbide and Carbon Chemicals Company, New York, N. Y. Used as solvent for vinyl chloride-acetate resin.

Solvent. Toluene, reagent grade, lot No E153. Obtained from General Chemical Division, Allied Chemical and Dye Corporation, New York, N. Y. Used as solvent for vinyl chloride-acetate resin.

Trichloroethylene. Technical grade. Obtained from Fisher Scientific Company, Silver Spring, Md. Used as degreasing agent for metal.

Apparatus

Balance. Ohaus, triple beam, stainless steel, catalog No 2-034, capacity 610 grams, divisions 0.1 gram. Obtained from Fisher Scientific Company, Silver Spring, Md. Used to weigh reagents.

Beaker. Pyrex glass, capacity 600 ml. Obtained from Fisher Scientific Company, Silver Spring, Md. Used to hold distilled water, etching solutions and trichloroethylene.

Bond Strength Testing Machine. Scott, model XL, jaw speed 20 inches per minute, 75 pounds counterweight used. Manufactured by Scott Testers, Inc., Providence, R. I. Installed in laboratory of O'Sullivan Rubber Corporation, Winchester, Va. Used to measure the bond strength of the plastic-metal laminate.

Brush. "All Pure Bristle" No 1, small. Obtained from Blacksburg Hardware Company, Blacksburg, Va. Used to apply adhesive on metal and plastic sheet.

Graduated Cylinder. Capacity 100 ml, divisions 1 ml. Obtained from Fisher Scientific Company, Silver Spring, Md. Used to measure volumes of acids and solvents.

Graduated Cylinder. Capacity 500 ml, divisions 5 ml. Obtained from Fisher Scientific Company, Silver Spring, Md. Used to measure volumes of acids and solvents.

Graduated Cylinder. Capacity 1000 ml, divisions 10 ml. Obtained from Fisher Scientific Company, Silver Spring, Md. Used to measure volume of distilled water.

Hot Plate. "Autemp" heater, rating 450 watts, 115 volts, ac, with temperature regulator. Obtained from Fisher Scientific Company, Silver Spring, Md. Used to heat etching solutions.

Hydraulic Press. Carver, capacity 10 tons, serial No 16600-50, pressure range 0-20,000 psig, equipped with hot plates and thermostats, 110 volts, maximum temperature 400° F. Manufactured by Fred S. Carver Inc., Summit, N. J. Obtained from Fisher Scientific Company, Silver Spring, Md. Used to laminate plastic sheet to metal.

Micrometer. Machinist's micrometer, one-inch opening, 0.001 graduations. Obtained from Fisher Scientific Company, Silver Spring, Md. Used to measure thickness of adhesive.

Polishing Cloth. "Crystal Bay Crocus Cloth", composed mainly of iron oxide. Manufactured by Minnesota Mining and Manufacturing Company, St. Paul, Minn. Used to polish metal surface.

Pyrometer. Portable surface, type 4200, serial No 79292, temperature range 0-600⁰ F. Manufactured by Illinois Testing Laboratories, Inc., 420 N La Salle Street, Chicago, Ill. Used for measuring the temperature of the heating plates of the hydraulic press.

Sand Paper. "Wetordry Tri-M-Ite Paper", grit No 320 A, soft-back, composed mainly of silicon carbide. Manufactured by Minnesota Mining and Manufacturing Company, St. Paul, Minn. Used to polish metal surface.

Sand Paper. "Wetordry Tri-M-Ite Paper", grit No 400 A, soft-back, composed mainly of silicon carbide. Manufactured by Minnesota Mining and Manufacturing Company, St. Paul, Minn. Used to polish metal surface.

Shear. Serial No 436, mild steel capacity 18 gauge. Manufactured by Wysong and Miles Company, Greensboro, N. C. Installed in Metal Working Laboratory, Industrial Engineering Department, Virginia Polytechnic Institute, Blacksburg, Va. Used for cutting metal samples.

Tensile Strength Testing Machine. Tinius Olsen, capacity 12,000 psig, jaw speed 0.01-1.00 inch per minute, equipped with model 51 electronic recorder. Manufactured by Tinius Olsen Testing Machine Company, Willow Grove, Penna. Installed in Wood Research Laboratory, Virginia

Polytechnic Institute, Blacksburg, Va. Used to measure the shear strength and to stretch the laminate.

Thermometer. Range 0 to 300° F, divisions 2° F. Obtained from Fisher Scientific Company, Silver Spring, Md. Used to measure temperature of the etching solution.

Tongs. Crucible, stainless steel, improved form, catalog No 15-193. Obtained from Fisher Scientific Company, Silver Spring, Md. Used to handle the metal specimens used in this investigation.

Method of Procedure

The specimens of metal sheet were treated as follows. Three sets of specimens were prepared. The first set was degreased with solvent only, the second one was polished and degreased, and the third one was degreased and etched. Semi-rigid polyvinyl chloride sheet was bonded to the metal at a temperature of 70°, 150°, 250° and 350° F under a pressure of 200 pounds per square inch. A series of the bonded specimens was stretched in the Tinius Olsen tensile testing machine until the elongation was 10, 20 and 30 per cent. Specimens were then tested for bond strength by laboratory personnel of the O'Sullivan Rubber Corporation at their laboratory. A series of steel-steel laminates bonded by different adhesives at 250° F under 200 pounds per square inch was also prepared. The specimens of this series were tested for shear strength. The plan of experiment was carried out according to the following method of procedure.

Treatment of Metals. The metal sheets were cut into a size of 2×4 inches with a shear and treated by one of the following three methods.

Solvent Degreasing. Trichloroethylene was used in this method. It was placed in a 600 ml beaker and held at a temperature of $70 \pm 2^{\circ}$ F. The metal specimens were dipped into the trichloroethylene and kept moving back and forth to maintain certain agitation. The treatment continued for 2 minutes. The specimens were removed and allowed to dry in the atmosphere.

Polishing and Solvent Degreasing. Since the metals used in this investigation had different surface finish, it was necessary to eliminate this factor in order to compare the chemical adhesion of different metals. The specimens were polished with sand paper and polishing cloth to approximately the same degree of smoothness. A sharp pencil point was placed vertically on the polished surface, the reflection was observed and compared. The polishing continued until all specimens gave the same reflection.

The polished metal sheets were rubbed with a clean, soft cloth to remove any grit and dirt. The specimens were then treated with trichloroethylene by the method described under solvent degreasing.

Solvent Degreasing and Etching. The metals were first treated with trichloroethylene and then etched by solutions.

The method of solvent degreasing previously described was used. The metals removed from trichloroethylene were dried in the atmosphere. After the solvent was completely evaporated, each kind of metal was treated with a special etching solution. These etching solutions were prepared according to the formulations recommended by Finishing Publication Inc. (9)

Etching of Aluminum. Aluminum was etched by alkaline and acid solutions successively. A three per cent sodium hydroxide solution was made by dissolving 40 grams of 76 per cent sodium hydroxide in 960 grams of distilled water. Five hundred milliliters of this solution was placed in a 600 ml beaker and heated to $190 \pm 2^{\circ}$ F.

The acid solution was prepared by diluting 10 grams of 48 per cent hydrofluoric acid and 10 grams of nitric acid with 980 grams of distilled water. Five hundred

milliliters of this solution was placed in a 600 ml beaker and maintained at $70 \pm 2^\circ$ F.

The specimen was first etched with alkaline and then with acid solution. The time of etching in alkaline solution was 30 seconds, in acid solution 2 minutes. While etching the specimen, it was kept moving back and forth in the beaker to maintain sufficient agitation.

Etching of Brass and Copper. The etching solution for brass and copper was prepared by dissolving 18 grams of sodium dichromate and 194 grams of sulfuric acid in 1798 grams of distilled water. Five hundred milliliters of this solution was placed in a 600 ml beaker and maintained at $70 \pm 2^\circ$ F.

The specimen was dipped in the solution and kept moving back and forth in the beaker to provide sufficient agitation. The time of treatment was 4 minutes for brass and 5 minutes for copper.

Etching of Nickel. The etching solution for nickel was prepared by dissolving 54 grams of sodium chloride, 54 grams of sodium nitrate

and 167 grams of sulfuric acid in 725 grams of distilled water. Five hundred milliliters of this solution was placed in a 600 ml beaker and heated to $185 \pm 2^{\circ}$ F.

The specimen was dipped in the solution and kept moving back and forth in the beaker to provide sufficient agitation. The time of treatment was 6 minutes.

Etching of Low Carbon Steel. The etching solution for low carbon steel was prepared by diluting 47 grams of sulfuric acid and 37 grams of hydrochloric acid with 916 grams of distilled water. Five hundred milliliters of this solution was placed in a 600 ml beaker and maintained at $70 \pm 2^{\circ}$ F.

The specimen was dipped in the solution and kept moving back and forth in the beaker to provide sufficient agitation. The time of treatment was 5 minutes.

Etching of Stainless Steel. The etching solution for stainless steel was prepared by diluting 157 grams of hydrochloric acid and 157 grams of sulfuric acid with 686

grams of distilled water. Five hundred milliliters of this solution was placed in a 600 ml beaker and maintained at $70 \pm 2^{\circ}$ F.

The specimen was dipped in the solution and kept moving back and forth in the beaker to provide sufficient agitation. The time of treatment was 5 minutes.

The metals removed from the etching solutions were flushed with running water for 10 seconds. The specimens were then rinsed successively with distilled water. Three 600 ml beakers were used. Each one contained 500 milliliters of distilled water. The time of rinsing in each beaker was 1 minute. Sufficient agitation was maintained by moving the specimen back and forth in the beaker. The metals were allowed to dry in the atmosphere after rinsing.

The complete process of solvent degreasing and etching is shown in Figure 1.

Preparation of Adhesive Solutions. Five different commercial adhesives were used in this investigation. They were dissolved in solvents to make solutions suitable for brushing.

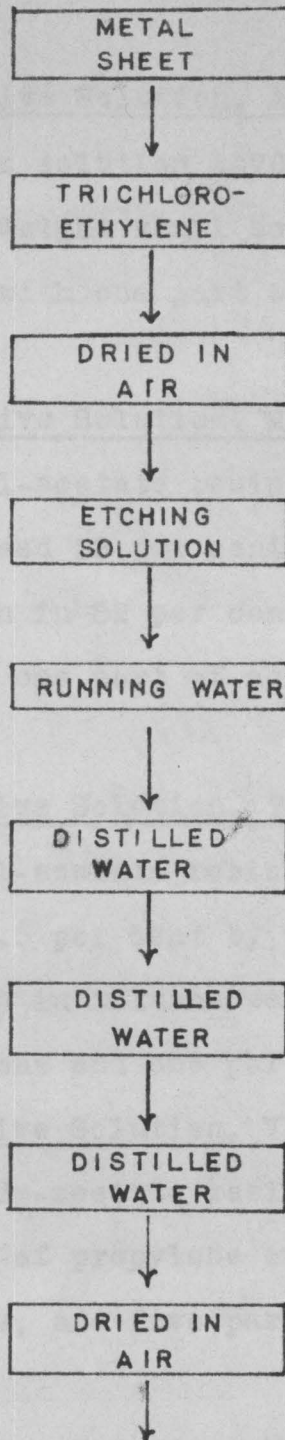


FIGURE 1. FLOW DIAGRAM FOR SOLVENT DEGREASING AND ETCHING

Adhesive Solution, A-70. One part of vinyl acetate resin solution A-70 which contained 69-72 per cent by weight vinyl acetate resin in acetone was diluted with one part of acetone and two parts of toluene.

Adhesive Solution, MA-28-18. One part of vinyl alcohol-acetate resin solution MA-28-18 which contained 28 per cent by weight vinyl alcohol-acetate resin in 82 per cent methyl acetate was diluted with one part of acetone and one part of toluene.

Adhesive Solution, T-24-9. One part of vinyl alcohol-acetate resin solution T-24-9 which contained 24.5 per cent by weight vinyl alcohol-acetate resin in toluene was diluted with one part of acetone and one part of toluene.

Adhesive Solution, VYNW. One part of vinyl chloride-acetate resin VYNW was dissolved in two parts of propylene oxide, five parts of cyclohexanone, and five parts of methyl ethyl ketone.

Adhesive Solution, VMCH. Ten parts of vinyl chloride-acetate resin VMCH was dissolved in eighty parts of mixed solvent which contained

1.5 per cent propylene oxide, 30 per cent methyl ethyl ketone, 30 per cent methyl isobutyl ketone, and 38.5 per cent toluene.

Adhesive Solution, VMCH, 5% DOP. Nine and one-half parts of vinyl chloride-acetate resin VMCH was dissolved in one-half part of dioctyl phthalate and 76 parts of mixed solvent which contained 1.5 per cent propylene oxide, 30 per cent methyl ethyl ketone, 30 per cent methyl isobutyl ketone, and 38.5 per cent toluene. This adhesive was called five per cent DOP VMCH in this investigation.

Adhesive Solution, VMCH, 10% DOP. Nine parts of vinyl chloride-acetate resin VMCH was dissolved in one part of dioctyl phthalate and 72 parts of mixed solvent which contained 1.5 per cent propylene oxide, 30 per cent methyl ethyl ketone, 30 per cent methyl isobutyl ketone, and 38.5 per cent toluene. This adhesive was called ten per cent DOP VMCH in this investigation.

Adhesive Solution, VMCH, 15% DOP. Eight and one-half parts of vinyl chloride-acetate resin VMCH was dissolved in one and one-half parts of

dioctyl phthalate and 68 parts of mixed solvent which contained 1.5 per cent propylene oxide, 30 per cent methyl ethyl ketone, 30 per cent methyl isobutyl ketone, and 38.5 per cent toluene. This adhesive was called fifteen per cent DOP VMCH in this investigation.

Adhesive Solution, VMCH, 17.65% DOP.

Fourteen parts of vinyl chloride-acetate resin VMCH was dissolved in 3 parts of dioctyl phthalate and 112 parts of mixed solvent which contained 1.5 per cent propylene oxide, 30 per cent methyl ethyl ketone, 30 per cent methyl isobutyl ketone, and 38.5 per cent toluene. The DOP content of this adhesive on a solvent-free basis was 17.65 per cent which was the same as in adhesive primer XL-5252 (24) recommended by Bakelite Corporation .

Adhesive Solution, VMCH, 20% DOP. Eight parts of vinyl chloride-acetate resin VMCH was dissolved in two parts of dioctyl phthalate and 64 parts of mixed solvent which contained 1.5 per cent propylene oxide, 30 per cent methyl ethyl ketone, 30 per cent methyl isobutyl ketone, and 38.5 per cent toluene. This adhesive was called twenty per cent DOP VMCH in this investigation.

Application of Adhesive. The adhesives were applied to both plastic sheet and metal sheet.

The plastic sheet was cut into a size of 2 X 4 inches. An adhesive solution VYNW was applied to the embossed sheet by brushing. Two sets of specimens were prepared, one of which was allowed to dry in the atmosphere for 10 minutes and one for 24 hours. The plastic with the adhesive coat dried 10 minutes was used to make laminate to be bonded at 70° and 150° F. That which dried for 24 hours was used for bonding at 250° and 350° F. The thickness of the adhesive coat after drying was about 0.001 inch in both cases.

The metal specimens treated by one of the three methods previously described were coated with adhesive by brushing. This coat of adhesive was allowed to dry in the atmosphere for 24 hours. The thickness of this coat after drying was measured with a micrometer. If it was less than 0.0015 inch, another coat was applied. This process was repeated until a thickness of 0.0015-0.0025 inch was obtained.

In all the above methods of applying the adhesive, only an area of 2 X 3 inches was covered so that when the specimens were laminated a part of the metal would not be bonded.

Bonding the Plastic to Metal. Five methods of bonding the plastic to metal were used in this investigation.

In the first method, the embossed plastic which had been coated with adhesive VYNW and dried in the atmosphere for 10 minutes was placed on top of the adhesive-coated metal. The assembly was placed between the two hot plates of the hydraulic press with the metal surface contacting the bottom hot plate. Both hot plates of the press were held at $70 \pm 2^{\circ}$ F. The temperature of the plates was measured with a surface pyrometer. The assembly was bonded under a pressure of 200 pounds per square inch. The time of loading from zero to 200 pounds was 10 ± 1 seconds. As soon as a pressure of 200 pounds per square inch was obtained, the pressure was released.

The second method was the same as the first one except that both hot plates were heated to $150 \pm 2^{\circ}$ F.

In the third method, the embossed plastic which had been coated with adhesive VYNW and dried in the atmosphere for 24 hours was placed on top of the adhesive-coated metal. The assembly was placed between the plates of the hydraulic press with the metal surface contacting the bottom hot plate. The bottom plate was heated to

250±2° F while the top plate was held at 70±2° F. The temperature of the plates was measured with a surface pyrometer. The assembly was bonded under a pressure of 200 pounds per square inch. The time of loading from zero to 200 pounds was 10±1 seconds. As soon as a pressure of 200 pounds per square inch was obtained, the pressure was released.

The fourth method was the same as the third one except that the bottom plate was heated to 350±5° F.

The fifth method used smooth plastic for lamination. The plastic was not coated with adhesive. It was placed on top of the adhesive-coated metal. The assembly was placed between the plates of the hydraulic press with the metal surface contacting the bottom hot plate. The bottom plate was heated to 350±5° F while the top plate was maintained at 70±2° F. The temperature of the plates was measured with a surface pyrometer. The assembly was bonded under a pressure of 200 pounds per square inch. The time of loading from zero to 200 pounds was 10±1 seconds. As soon as a pressure of 200 pounds per square inch was obtained, the pressure was released. These specimens were used for stretching tests.

Stretching the Laminates. The laminates bonded by the fifth method were subjected to stretching. Before the samples were stretched, they were cut into one-inch strips and conditioned in a room at 72° F and 50 per cent relative humidity for one hour. Two parallel lines two inches apart were marked crosswise on each sample. The samples were stretched in a Tinius Olsen tensile strength testing machine at a speed of 0.1 inch per minute to 10, 20 and 30 per cent elongation between the two marked lines.

Test for Bond Strength. The specimens were sent to O'Sullivan Rubber Corporation, Winchester, Va. for bond strength test. Prior to testing, the specimens were conditioned at 75° F and 50 per cent relative humidity for 30 minutes. The specimen was placed in a Scott testing machine and the plastic sheet peeled from the metal. The greatest force required to peel the plastic off the metal was recorded. The bond strength was reported in pounds per inch width of the sample. Figure 2 is the photograph of the Scott testing machine.

Bonding Metal to Metal. Low carbon steel was used in this experiment. The metal was cut into a size of 1 x 4 inches and treated by the degreasing and etching method. Different adhesives were used to bond the metal. In each case, the adhesive was applied on one end of the

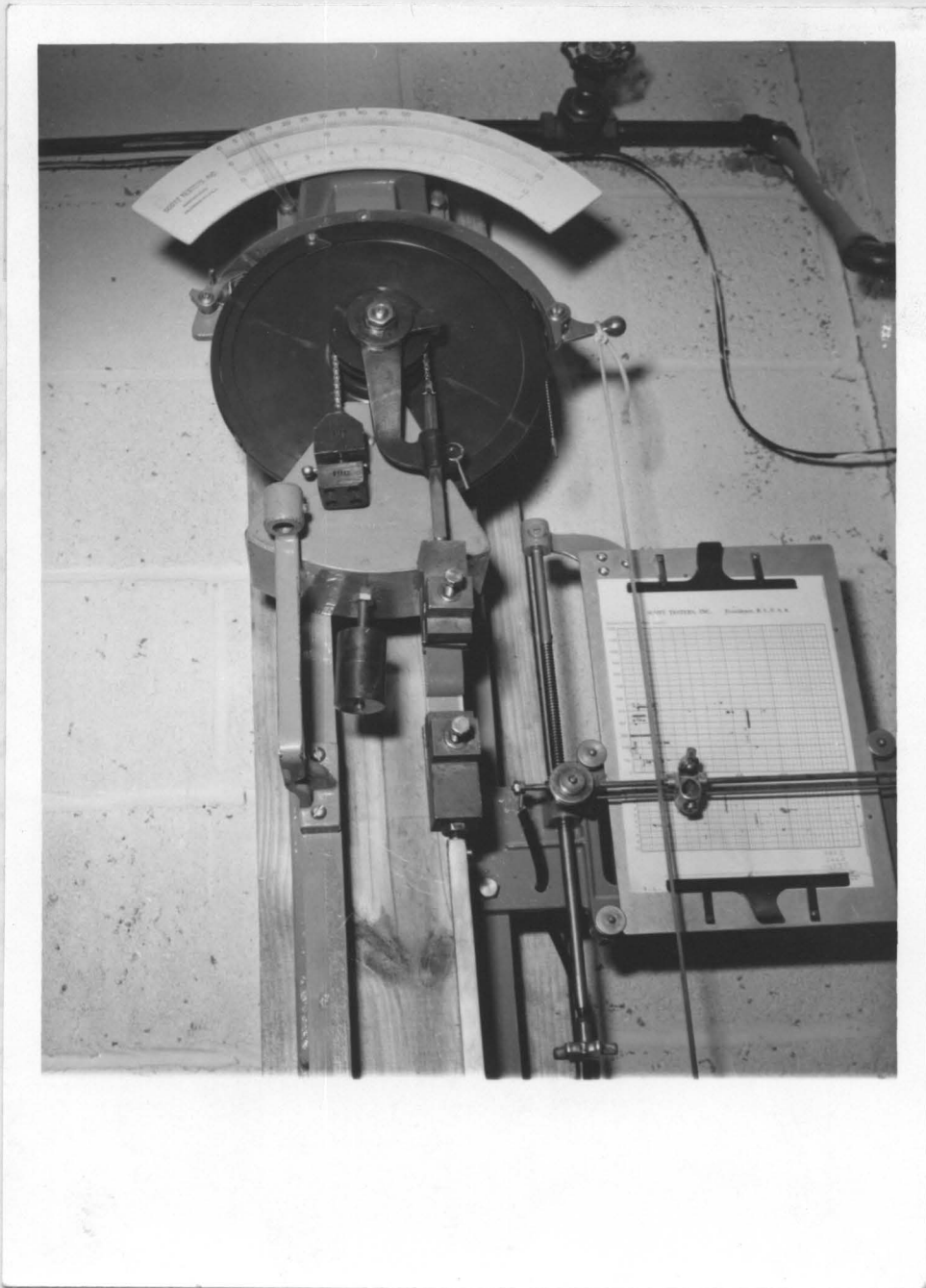


Figure 2. View of Scott Testing Machine

metal strip covering an area of 1X1 inch. Both adherends were coated. The adhesive coat was allowed to dry in the atmosphere for 24 hours. The adhesive-coated metal was placed on top of the other with the uncoated part in opposite direction. The assembly was placed between the two hot plates of the hydraulic press. Both hot plates were heated to $250 \pm 2^{\circ}$ F. The assembly was bonded under a pressure of 200 pounds per square inch for 2 minutes. These specimens were used for shear strength tests.

Test for Shear Strength. The metal-metal laminates bonded by different adhesives were tested for shear strength. Prior to testing, the specimens were conditioned in a room at 72° F and 50 per cent relative humidity for one hour. The specimen was placed in a Tinius Olsen tensile strength testing machine and pulled at a speed of 0.05 inch per minute. The shear strength was reported in pounds per square inch of bonding area.

Data and Results

The data and results obtained from this investigation are presented in the following section.

Bond Strength of Embossed Plastic-Metal Laminates Bonded by VYNW and Plasticized VMCH. The greatest force required to peel the plastic off the aluminum, brass, copper, nickel, low carbon steel, and stainless steel laminates bonded by adhesive VYNW and primer plasticized VMCH (17.65 per cent DOP) at various temperatures is given in Tables I, II, III, IV, V and VI, pages 46, 47, 48, 49, 50 and 51 respectively. Figures 3, 4, 5, 6, 7 and 8, pages 54, 55, 56, 57, 58 and 59 are the corresponding strength curves of these laminates. Figure 9, page 60, is a comparison of bond strengths of polished specimens of different metals. It was apparent from these results that the bond strength of metal laminates followed the order of these metals in the galvanic series.

Bonded Strength of Embossed Plastic-Metal Laminates Bonded by VYNW and A-70. Embossed plastic sheet was bonded to aluminum, brass, copper, nickel, low carbon steel and stainless steel by adhesive VYNW and primer A-70 at 70^o, 150^o, 250^o and 350^o F. The results of the pre-

liminary tests showed that the bond strength of these laminates was very weak. The bond of each of these laminates was easily broken by hand at the interface between adhesive VYNW and primer A-70. It was indicated that primer A-70 did not adhere to adhesive VYNW.

Bond Strength of Embossed Plastic-Metal Laminates

Bonded by VYNW and MA-28-18. Embossed plastic sheet was bonded to aluminum, brass, copper, nickel, low carbon steel and stainless steel by adhesive VYNW and primer MA-28-18 at various temperatures. The results of the preliminary tests showed that the bond strength of these laminates was very weak. The bond of each of these laminates was easily broken by hand at the interface between adhesive VYNW and primer MA-28-18. It was indicated that primer MA-28-18 did not adhere to adhesive VYNW.

Bond Strength of Embossed Plastic-Metal Laminates

Bonded by A-70. Embossed plastic sheet was bonded to aluminum, brass, copper, nickel, low carbon steel and stainless steel by adhesive A-70 at various temperatures. The results of the preliminary tests showed that the bond strength of these laminates was very weak. The bond of each of these laminates was easily broken by hand at the interface between plastic and adhesive A-70. It was

indicated that adhesive A-70 did not adhere to plasticized polyvinyl chloride sheet.

Bond Strength of Embossed Plastic-Metal Laminates

Bonded by MA-28-18. Embossed plastic sheet was bonded to aluminum, brass, copper, nickel, low carbon steel, and stainless steel by adhesive MA-28-18 at various temperatures. The results of the preliminary tests showed that the bond strength of these laminates was very weak. The bond of each of these laminates was easily broken by hand at the interface between plastic and adhesive MA-28-18. It was indicated that adhesive MA-28-18 did not adhere to plasticized polyvinyl chloride sheet.

Effect of Amount of Plasticizer in Adhesive. The

bond strength of smooth plastic-low carbon steel laminates bonded by adhesive VMCH with various amounts of dioctyl phthalate plasticizer at 350° F is given in Table VII, page 52, and Figure 10, page 61. The laminates were stretched to various degree of elongation to study the bond strength when the products were deformed by bending and deep drawing. It was indicated that the rate of decrease in strength with stretching was inversely proportional to the amount of plasticizer in the adhesive.

Shear Strength of Steel-Steel Laminates Bonded by Different Types of Adhesives. The shear strength of steel-steel laminates bonded by different types of adhesives at 250° F is given in Table VIII, page 53. It was apparent that the adhesion of adhesive to metal was proportional to the polarity of the adhesive.

TABLE I

Bond Strength of Embossed Plastic-Aluminum Laminates
Bonded by VYNW and Plasticized VMCH^a

Bonding Temperature	Bond Strength	
	Etched	Polished
°F	lb/in	lb/in
70	23 P/A	16 P/L
150	24 P/A	17½ P/L
250	24 T/A	14½ P/L
350	27 T/L	14 P/L

Type of failure: P - bond failure, plastic peeled from metal at least one quarter inch.
 T - combination bond and plastic failure, plastic torn in an irregular pattern.
 A - failure in adhesive layer.
 M - adhesive left on metal.
 L - adhesive left on plastic.

a - Plasticizer, dioctyl phthalate, 17.65 per cent.

TABLE II

Bond Strength of Embossed Plastic-Brass Laminates
Bonded by VYHW and Plasticized VMCH^a

Bonding Temperature	Bond Strength	
	Etched	Polished
^o F	lb/in	lb/in
70	4 P/L	1 P/L
150	3 P/L	2 P/L
250	8 P/L	1 P/L
350	8 P/L	6 P/L

Type of failure: P - bond failure, plastic peeled from metal at least one quarter inch.
T - combination bond and plastic failure, plastic torn in an irregular pattern.
A - failure in adhesive layer.
M - adhesive left on metal.
L - adhesive left on plastic.

a - Plasticizer, dioctyl phthalate, 17.65 per cent.

TABLE III

Bond Strength of Embossed Plastic-Copper Laminates
^a
Bonded by VYNE and Plasticized VMCH

Bonding Temperature	Bond Strength	
	Etched	Polished
°F	lb/in	lb/in
70	4 P/L	3 P/L
150	6 P/L	6 P/L
250	8 P/L	7 P/L
350	14 P/L	12 P/L

Type of failure: P - bond failure, plastic peeled from metal at least one quarter inch.
T - combination bond and plastic failure, plastic torn in an irregular pattern.
A - failure in adhesive layer.
M - adhesive left on metal.
L - adhesive left on plastic.

a - Plasticizer, dioctyl phthalate, 17.65 per cent.

TABLE IV

Bond Strength of Embossed Plastic-Nickel Laminates
Bonded by VYNU and Plasticized VVCH^a

Bonding Temperature	Bond Strength	
	Etched	Polished
°F	lb/in	lb/in
70	19½P/L	5 P/A
150	---	11 P/ML
250	14 P/L	9 P/L
350	17½P/L	9 P/L

Type of failure: P - bond failure, plastic peeled from metal at least one quarter inch.
T - combination bond and plastic failure, plastic torn in an irregular pattern.
A - failure in adhesive layer.
M - adhesive left on metal.
L - adhesive left on plastic.

a - Plasticizer, dioctyl phthalate, 17.65 per cent.

TABLE V

Bond Strength of Embossed Plastic-Low Carbon Steel
Laminates Bonded by VYNN and Plasticized VMCH^a

Bonding Temperature	Bond Strength		
	Etched	Polished	Degreased
°F	lb/in	lb/in	lb/in
70	15 P/ML	20 P/A	11½ P/L
150	14 P/L	22 T/ML	11 P/L
250	15 P/L	18½ P/L	10 P/L
350	13 P/L	18 P/L	9 P/L

Type of failure: P - bond failure, plastic peeled from metal at least one quarter inch.
 T - combination bond and plastic failure, plastic torn in an irregular pattern.
 A - failure in adhesive layer.
 M - adhesive left on metal.
 L - adhesive left on plastic.

a - Plasticizer, dioctyl phthalate, 17.65 per cent.

TABLE VI

Bond Strength of Embossed Plastic-Stainless Steel
Laminates Bonded by VYNH and Plasticized VMCH^a

Bonding Temperature	Bond Strength	
	Etched	Polished
^o F	lb/in	lb/in
70	20 P/A	16 P/A
150	18 P/A	14½ P/L
250	18 P/L	14½ P/L
350	23 P/L	10 P/L

Type of failure: P - bond failure, plastic peeled from metal at least one quarter inch.
T - combination bond and plastic failure, plastic torn in an irregular pattern.
A - failure in adhesive layer.
M - adhesive left on metal.
L - adhesive left on plastic.

^a - Plasticizer, dioctyl phthalate, 17.65 per cent.

TABLE VII

Effect of Amount of Plasticizer in Adhesive on Bond Strength of Smooth Plastic-Low Carbon Steel Laminates in Stretching

Plasticizer ^a	Bond Strength			
	Per Cent Elongation			
	0	10	20	30
%	lb/in	lb/in	lb/in	lb/in
0	18 P/M	10 P/L	7 P/L	4 P/L
5	25 P/M	24 T/A	16 T/A	8 P/L
10	16 P/ML	14 P/A	10 P/M	6 P/L
15	12½ P/ML	11 P/L	7 P/ML	6 P/L
20	7 P/M	6 P/L	7 P/L	5 P/L

Type of failure: P - bond failure, plastic peeled from metal at least one quarter inch.
 T - combination bond and plastic failure, plastic torn in an irregular pattern.
 A - failure in adhesive layer.
 M - adhesive left on metal.
 L - adhesive left on plastic.

a - Plasticizer, DOP; adhesive VMOH.

TABLE VIII

Shear Strength of Adhesives in Steel-Steel Laminates

Adhesives	Shear Strength		
	No 1	No 2	Average
	lb/in ²	lb/in ²	lb/in ²
Vinyl chloride-acetate resin, VYNW	502 P	448 M	475
Vinyl acetate resin, A-70	702 P	696 P	699
Vinyl alcohol-acetate resin, T-24-9	802 N	886 L	844
Vinyl alcohol-acetate resin, MA-28-18	900 L	907 L	903.5
Modified vinyl chloride- acetate resin, VMCH	974 L	1000 L	987

Type of failure: L - failure in adhesive layer.
M - failure at adhesive-metal interface.
N - a large part of failure in adhesive layer, small part at interface.
P - a large part of failure at interface, small part in adhesive layer.

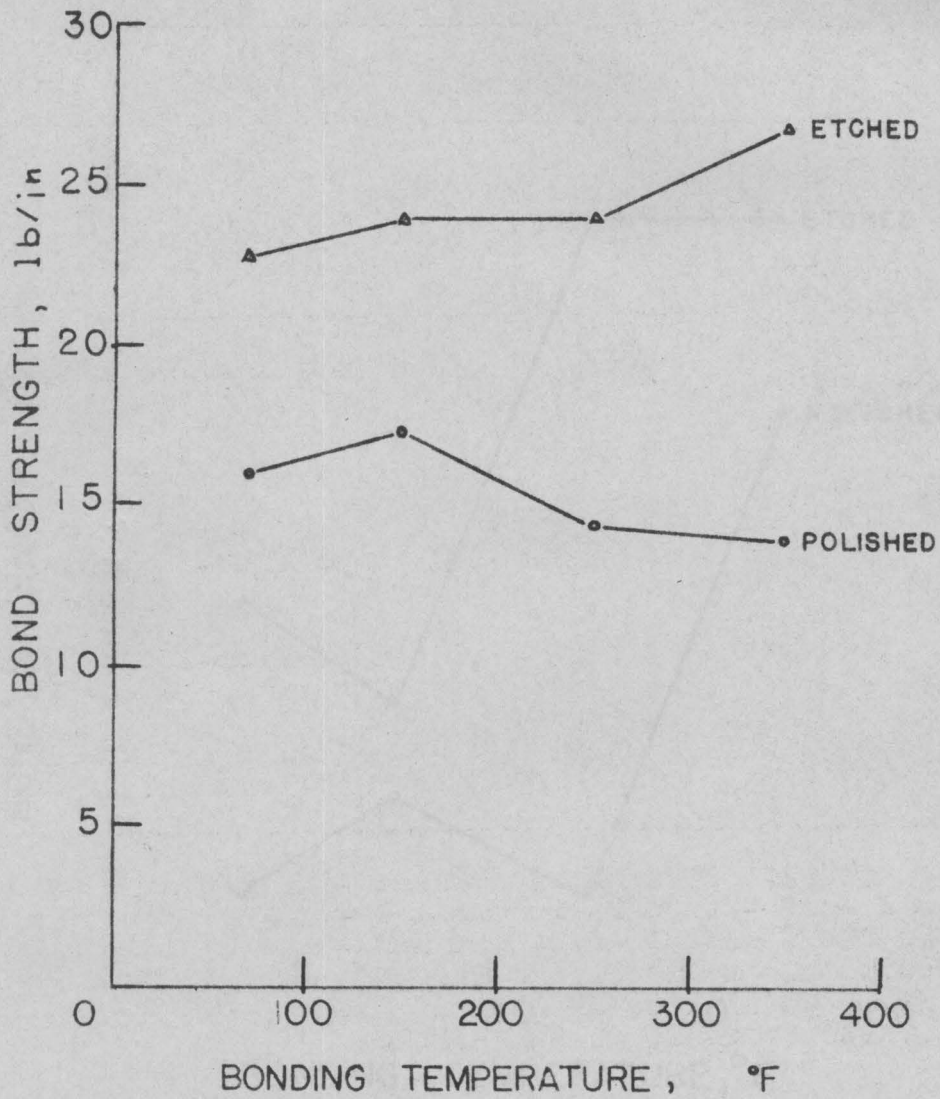


FIGURE 3. BOND STRENGTH OF PLASTIC-ALUMINUM LAMINATES BONDED BY VYNW AND PLASTICIZED VMCH

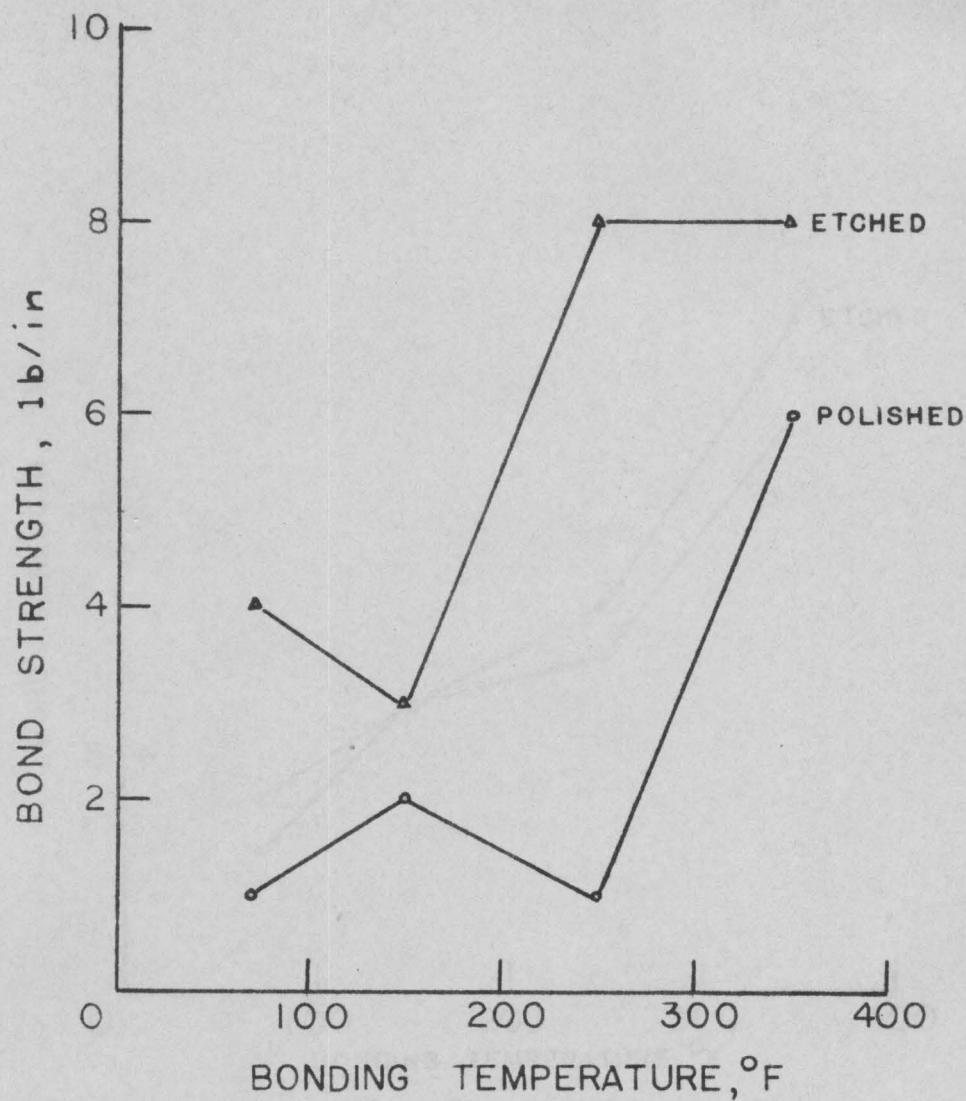


FIGURE 4. BOND STRENGTH OF PLASTIC-BRASS LAMINATES BONDED BY VYNW AND PLASTICIZED VMCH

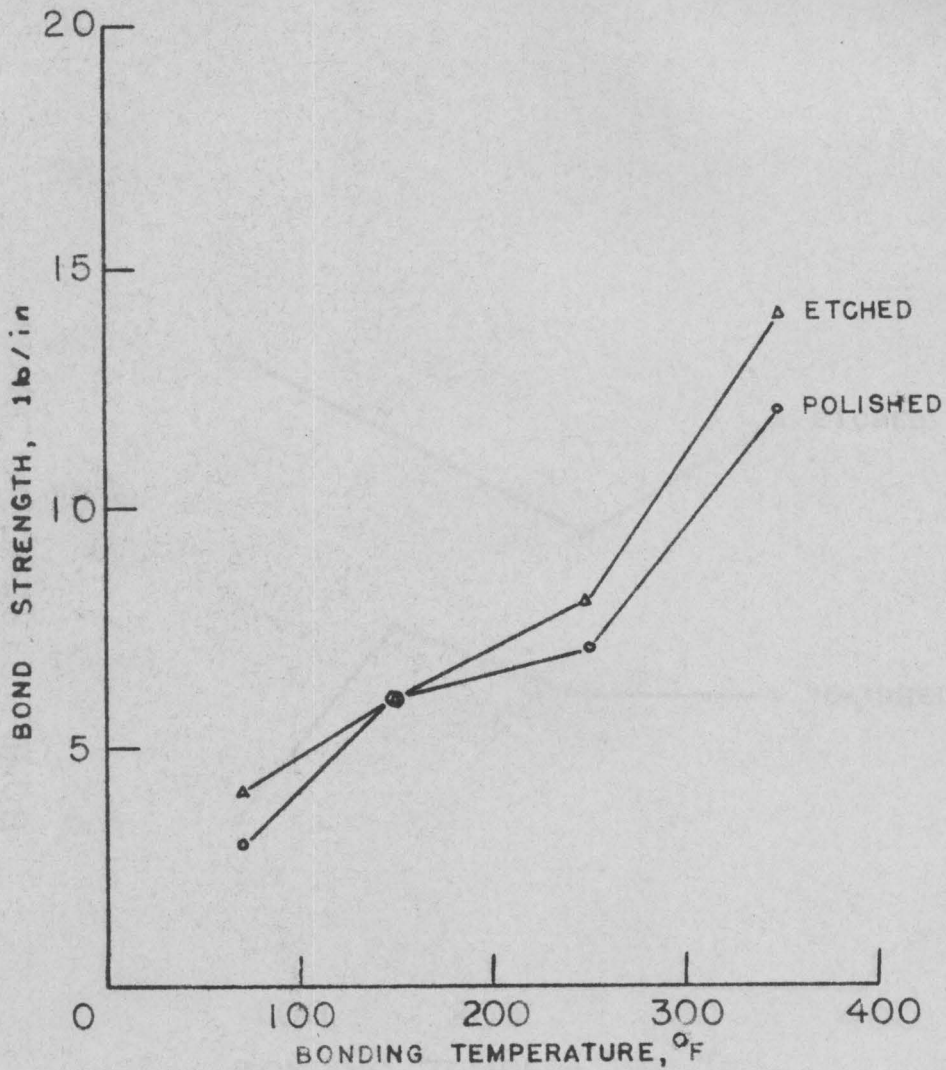


FIGURE 5. BOND STRENGTH OF PLASTIC-COPPER LAMINATES BONDED BY VYNW AND PLASTICIZED VMCH

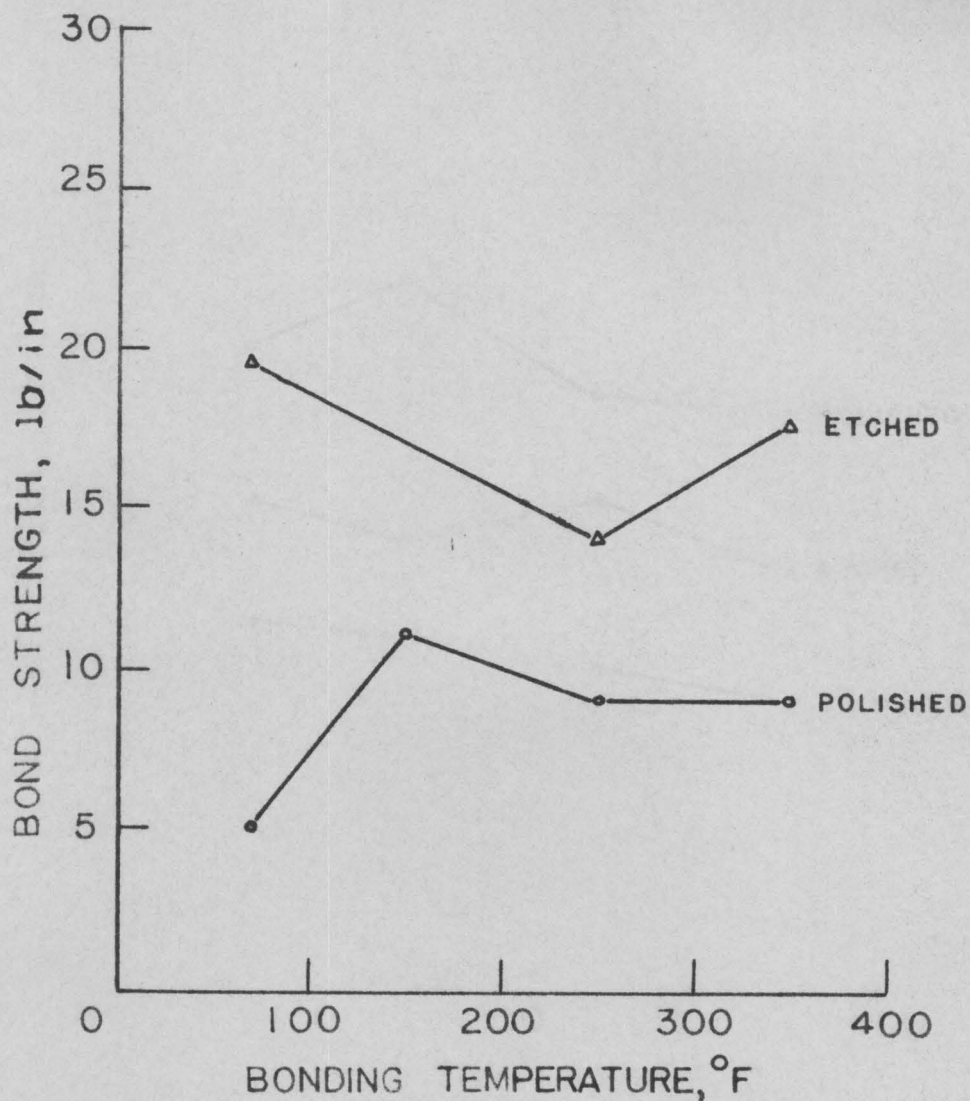


FIGURE 6. BOND STRENGTH OF PLASTIC-NICKEL LAMINATES BONDED BY VYNW AND PLASTICIZED VMCH

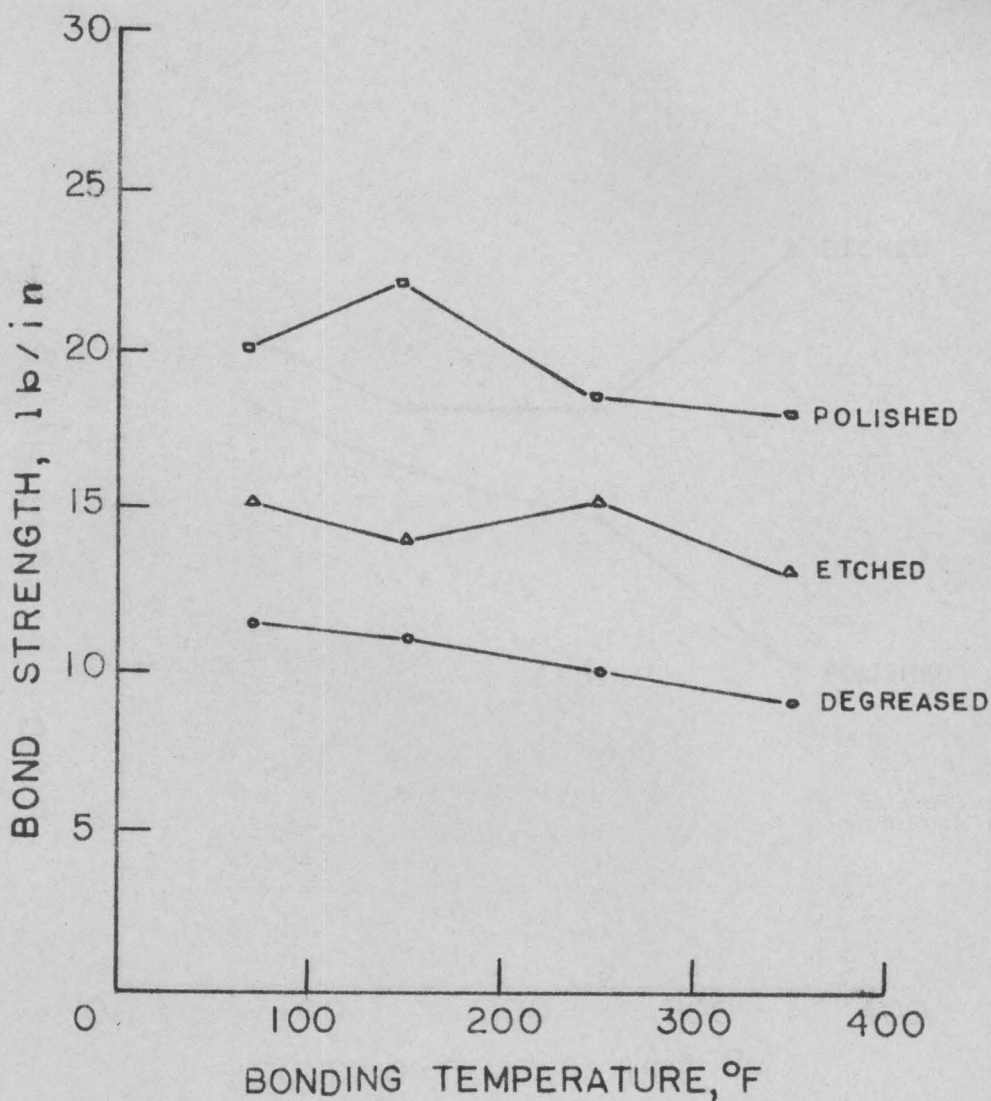


FIGURE 7. BOND STRENGTH OF PLASTIC-LOW CARBON STEEL LAMINATES BONDED BY VYNW AND PLASTICIZED VMCH

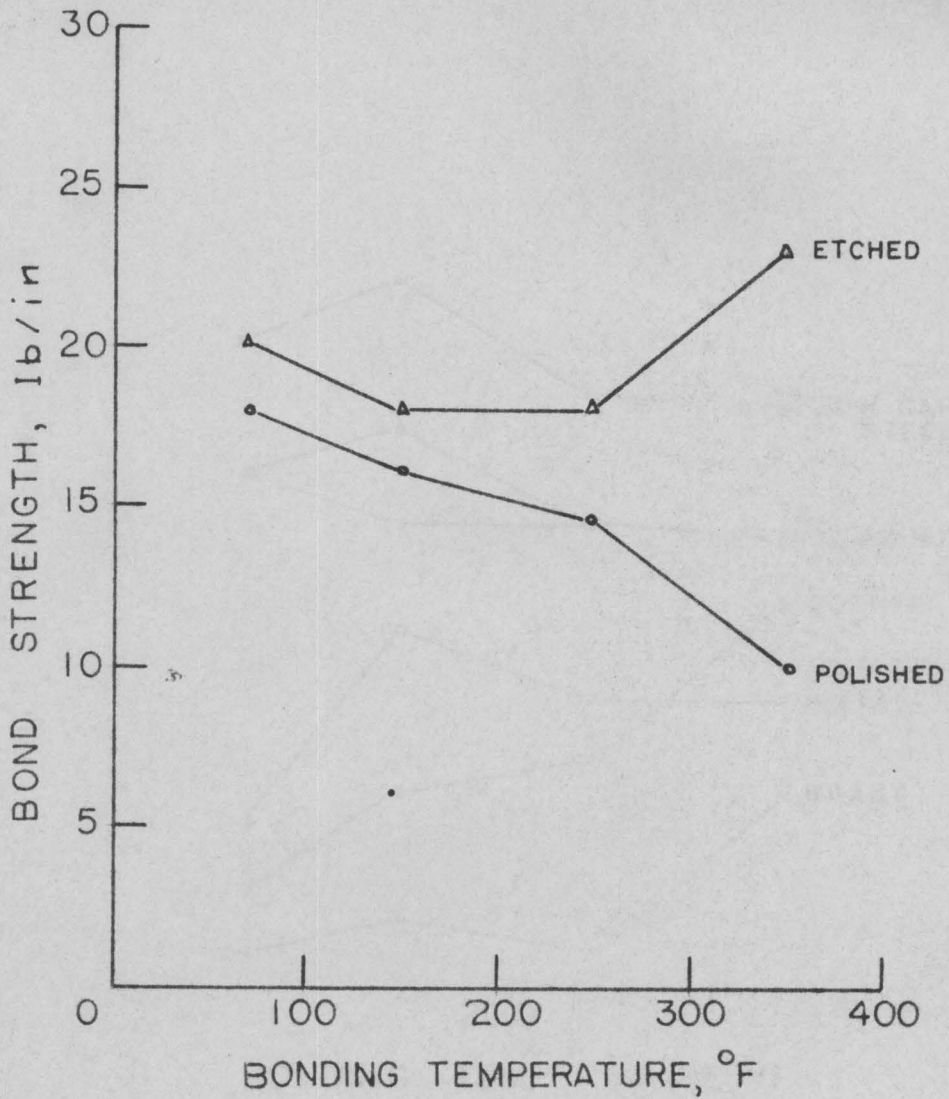


FIGURE 8. BOND STRENGTH OF PLASTIC-STAINLESS STEEL LAMINATES BONDED BY VYNW AND PLASTICIZED VMCH

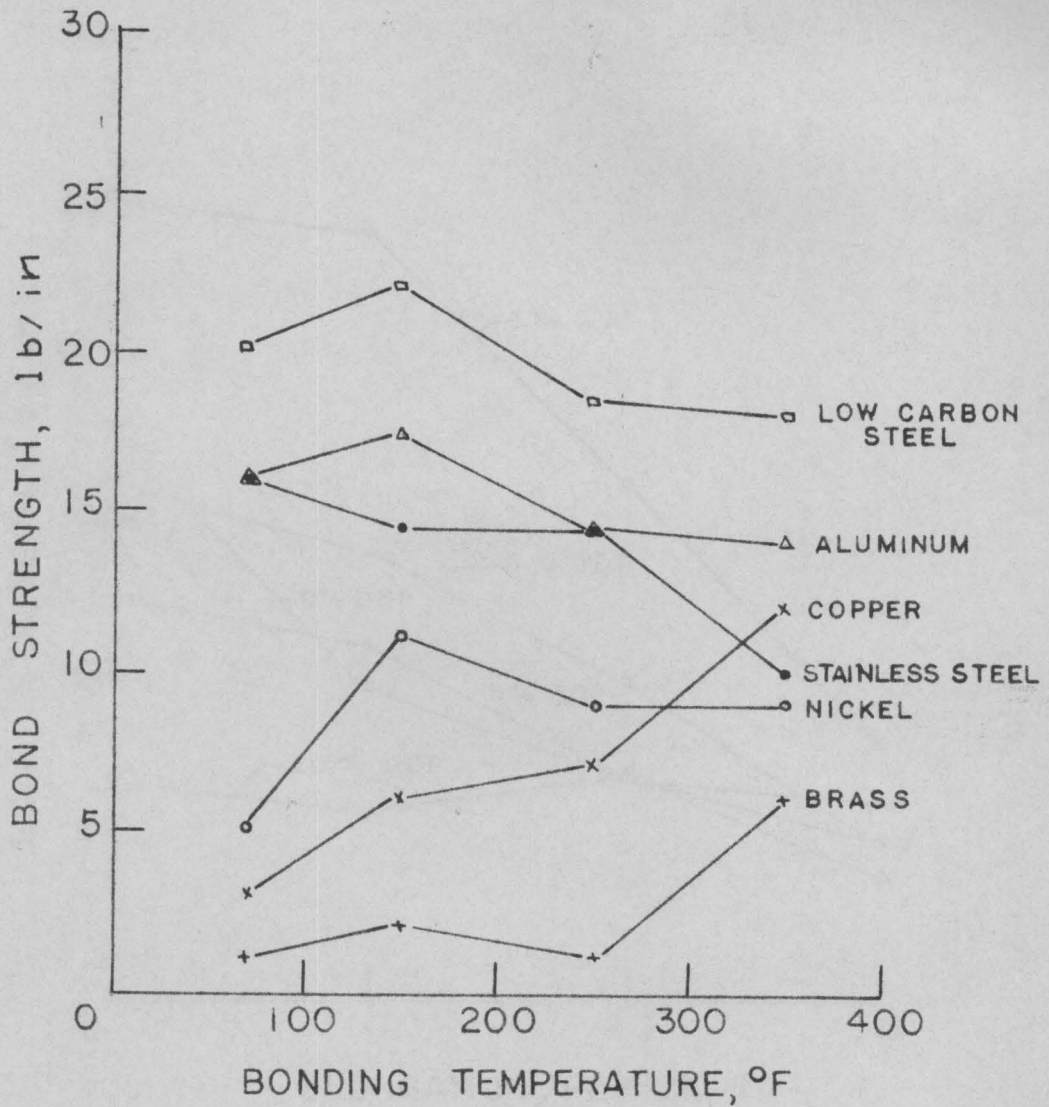


FIGURE 9. COMPARISON OF BOND STRENGTH OF DIFFERENT METAL LAMINATES BONDED BY VYNW AND PLASTICIZED VMCH

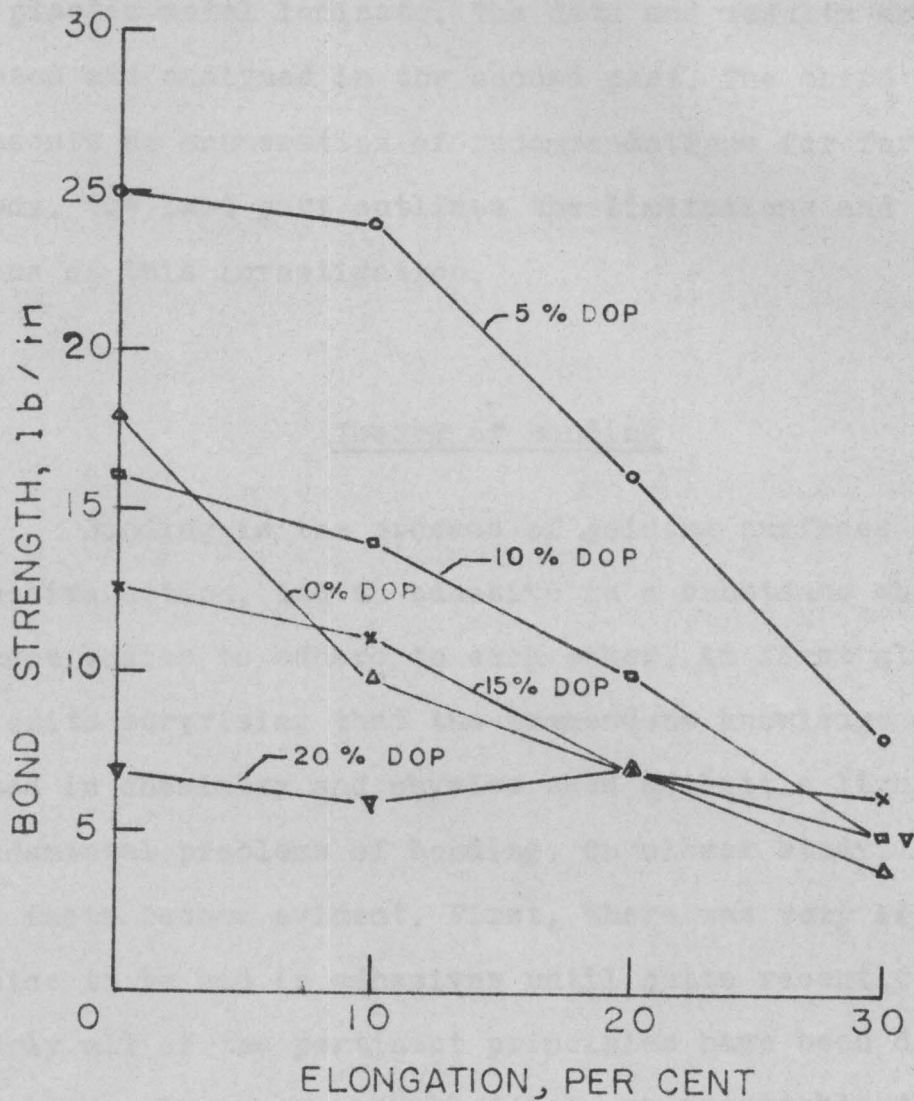


FIGURE 10. EFFECT OF AMOUNT OF DOP PLAS-
TICIZER IN ADHESIVE ON BOND STRENGTH
OF PLASTIC-STEEL LAMINATES IN
STRETCHING

IV. DISCUSSION

This section contains four parts. The first part attempts to interpret a general theory of bonding applied to plastic-metal laminate. The data and results are discussed and analyzed in the second part. The third part presents an enumeration of recommendations for further study. The last part outlines the limitations and restrictions of this investigation.

Theory of Bonding

Bonding is the process of joining surfaces by adhesive action, and an adhesive is a substance which causes bodies to adhere to each other. At first glance it is quite surprising that the tremendous knowledge accumulated in chemistry and physics shed so little light on the fundamental problems of bonding. On closer study, however, two facts become evident. First, there was very little choice to be had in adhesives until quite recently. Second, nearly all of the pertinent principles have been disclosed and have only to be assembled into an acceptable theory of bonding.

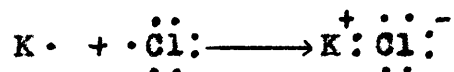
The strength of an adhesive joint is governed by two factors. These are adhesion at the interfaces between adhesive and adherends and cohesion within the adhesive layer itself. When a bonded assembly is subjected to stress, it may fail (1) at either of the two adhesive-adherend interfaces against adhesive forces, or (2) within either adherend or the adhesive layer against cohesive forces. There are thus essentially five possible regions of failure. Actual rupture will occur at the weakest point of the assembly.

In dealing with the nature of the adhesive bond, it is convenient to recognize two general types of bonding forces which hold atoms and molecules together in all matter. These are primary valence forces and secondary valence forces.

Primary Valence Forces. These forces hold one atom to another within a molecule and are also termed intramolecular forces. They have been found to be of different kinds.

The Ionic Bond. According to the octet theory, the most stable configuration of the outermost electrons of an element is a shell of eight. Atoms of many elements attain an outer group of eight electrons by exchanging electrons with other atoms, thus forming

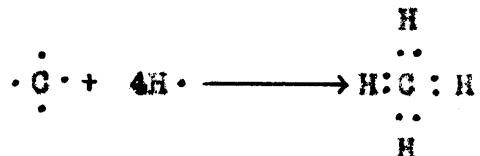
ionic bonds. An ionic bond is a chemical bond between atoms or groups, in which one or more electrons are given up by the electropositive element and transferred to the electronegative one, the resultant ionic structures are held together by the mutual attraction of opposite electric charges. The formation of potassium chloride, for example, is due to the transfer of an outermost electron of potassium to chlorine. This transfer results in the formation of charged ions which are attracted to one another to form potassium chloride.



The ionic bond has also been called the polar bond.

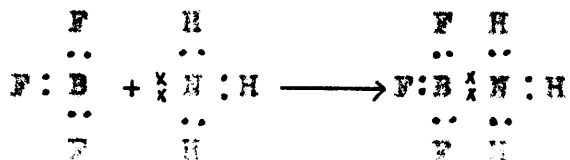
The Covalent Bond. Many chemical compounds, particularly the organic, are nonionic in character. The atoms in these compounds attain stable shells of eight, not by losing or gaining electrons, but by sharing electrons with an adjacent atom. A covalent bond is a chemical bond between atoms or groups in which, one or more pairs of electrons shared, the atoms or groups are held together by the stable configuration resulting from the sharing. In this type of linkage, each atom or group contributes an electron to the shared pair. In methane,

four bonds are formed between one carbon and four hydrogen atoms. The covalent bond is sometimes



called the nonpolar bond.

The Coordinate Bond. In this type of linkage, the force which holds atoms together is also due to the sharing of a pair of electrons. But in this case, both the electrons of the shared pair are supplied by the one atom. This atom in effect loses one electron, one-half of the shared pair, to form a covalent bond. The donor of electrons therefore becomes positive and the acceptor negative. This type of chemical bond has a tendency to ionize so that it is sometimes called a semipolar bond.



An example of a coordinate bond is given by the formation of ammoniate of boron trifluoride. In the above notation, the electrons of particular interest are shown x.

The Metallic Bond. The metals owe their unique properties to a type of bond that was not recognized until rather recently. In a metal, there is such a general dearth of valence electrons that the concept of the formation of stable electronic configurations by electronic transfer or electron pair sharing is unfruitful. In order to explain the characteristic properties of metals, it is now recognized that the metallic bond is a very special case of a covalent bond.

When two metallic atoms approach one another, their electrons form a valence bond. Owing to the small number of valence electrons available, it is far short of a complete shell. Accordingly, on the approach of another metallic atom, this atom joins the group. This effect continues as more and more atoms enter the aggregate, forming a crystal rather than a definite molecule. The metallic bond is essentially covalent in nature but spread throughout the entire group of atoms, holding them all together. It differs from the ordinary covalent bond in (1) that there is no saturation of valence, so that any number of atoms can be held together, (2) that the bond is not directional. The lack of direction in

the bonding forces enables the individual atoms in the lattice to move past one another without suffering any essential change in environment. This results in the characteristic properties of ductility, malleability, and cold working. Another conspicuous property of metal is electrical conductivity. This is also a consequence of small number of valence electrons relatively distant from the balance of the atom. The sharing of the valence electrons with many neighboring atoms loosens the electron and permits free motion. The structure is one of electropositive ions immersed in a cloud of mobile electrons.

Secondary Valence Forces. The forces which hold one molecule to another are known as secondary valence forces, sometimes called intermolecular forces. They are due to residual fields left about the molecule as a whole after two or more atoms have combined through primary bonds to form the molecule itself. These forces account for (1) melting and boiling points, (2) solubility and miscibility, and (3) mechanical strength. The secondary forces are weak in comparison with the primary valence forces, but when molecules grow very large, these forces assume spectacular importance.

They are generally divided into three classes. These are dispersion forces, induction forces and electrostatic forces.

The dispersion forces are by far the most common and the most important of the three classes. They are the non-polar forces which act between all atoms. This type of force contributes to the temporary relative displacement of the nuclei and electrons during the vibration of the several parts of the molecule with respect to one another. This displacement results in temporary dipoles that make intermolecular attractive forces possible.

The second class of secondary valence forces is numerically the least important. If an electric system ordinarily devoid of dipole moment is placed in an electric field which causes a relative displacement of the positive and negative charges, a moment is induced. For many molecules it has been found that an electric field causes slight displacement of the electrons and nuclei, thus leading to such an induced moment. Since the permanent dipole of one molecule can induce a moment in a second molecule, this effect is particularly important in the attractions occurring between polar and nonpolar compounds.

The third class of secondary valence forces is due to the electrostatic attraction between the charged spots

on the molecules. A molecule or part of a molecule may possess charges at its surface traceable to the electron transfer that has taken place. In the case of a covalent bond, no electron transfer occurs, consequently this bond is inherently of low polarity. But molecules held together by covalent bonds may nevertheless be polar due to the location of the positively charged nuclei. The polar hydrogen atom in alcohols, acids and amines is just a positive spot like any other, but it is a very active one by reason of the small size of that atom. The energy of the hydrogen bridge or hydrogen bond is considered to be mainly of an electrostatic nature.

Surface Properties. The surface of solids differs appreciably from their interior in regard to energy relationships. A molecule in the interior of a solid is surrounded on all sides by neighbors and intermolecular attraction is the same in all directions. In the surface, however, the attractive force is unbalanced, there are very few molecules in the gas phase. Therefore, there is a residual field of attraction in the surface, and that is a state of unbalance or unsaturation. In order to extend the area of the surface, work must be done to bring molecules from the interior into the surface against inwardly directed attractive forces. It follows that the surface molecules have

greater energy than the interior molecules, and this additional energy content is termed the surface energy. It is this energy that causes adhesion to unlike molecules.

Since a surface is characterized by unbalanced valence forces, it is certain that adsorption or reaction soon occurs at almost any freshly exposed surface. The intensity of this force depends on the specific nature of the material and the physical condition of the surface. The greater the activity which a surface possesses the greater is the force of attraction with other material. In order to improve the adhesive force or bond strength of a joint, it is generally necessary to have the surface well cleaned before the adhesive is applied.

Nature of the Plastic-Metal Bond. The bonding of plastic to metal involves, like other types of bonding, two requirements for the formation of an adhesive joint. These are cohesion in the adhesive layer and adhesion at the adhesive-plastic and adhesive-metal interfaces. Although this is a comparatively recent development, the nature of this type of bonding can be satisfactorily interpreted in terms of the previous discussion.

Cohesion in the Adhesive. The nature of the cohesive forces in the adhesive layer depends on the type of adhesive used. In the case of thermoplastic or long chain compounds such as vinyl acetate resin, the cohesive forces between chains are secondary valence in character. The magnitude of these forces depends on the nature of the resin, molecular weight, polarity, symmetry and orientation. In some cases, thermoplastic resins such as polyvinyl alcohol-acetate may acquire some degree of cross-linking when they are heated at high temperature. When this is so, the cohesive forces within the adhesive layer are a combination of primary and secondary valence forces. It is effective in raising the softening point and increasing the mechanical strength.

If the adhesive used is a thermosetting resin such as phenol-formaldehyde, it is set by polymerization and a three-dimensional structure results. The cohesive forces are thus strong covalent bonds and the gross physical properties are those of great strength and stability.

Adhesion between Adhesive and Plastic. As previously pointed out, the surface of a solid differs from its interior in regard to energy relationship. It possesses a higher energy content than the interior and this energy may cause adhesion or reaction to foreign molecules. However, the activity of a surface depends on the nature of the material and the physical condition of the surface itself, the cleaner the surface, the greater the activity. An ordinary surface is likely to become contaminated by impurities from outside. The activity of the surface is therefore greatly reduced.

In the bonding of plastic to metal, it is usual to bond the plastic as such without any previous treatment to prepare a clean surface. Due to the contamination by impurities which an ordinary surface has, the surface energy of the plastic is rather low. It is certain that this energy is not a major factor to cause adhesion.

In plastic-metal bonding, the adhesive is usually applied to the plastic in the form of a solution. In this case, the solvent in the adhesive solution solvates or even dissolves the plastic

surface. This enables the adhesive molecules to penetrate into the plastic and mix with it like a true solution. After the solvent evaporates off, the adhesive is held to the plastic by secondary valence forces.

If the adhesive is first coated on metal and the plastic is bonded to it by a heat-melt method, the melted adhesive still can penetrate into the plastic and form a strong bond of the secondary valence character.

The ability of an adhesive to penetrate into the plastic depends on the interfacial tension between these two materials. If the interfacial tension is high, the adhesive molecules can not penetrate into the plastic and these two materials are incompatible. The fact that vinyl acetate resin and vinyl alcohol-acetate resin do not adhere to plasticized polyvinyl chloride is simply due to the high interfacial tension between the adhesive and the plastic. The failure of penetration results in very weak adhesive forces.

Adhesion between Adhesive and Metal. It has been stated that the structure of a metal is one of

electropositive ions immersed in a cloud of mobile electrons. These positively charged metallic ions have a tendency to attract electronegative atoms or groups. When a polar compound comes in contact with a metal, an electrostatic field is built up and the compound orientates itself with the polar group or electronegative spots toward the metal. These two substances are then held together by electrostatic forces, a secondary-valence effect. When the molecules of the compound are very large, these forces become extensive and a good bond is obtained.

Because the magnitude of an electrostatic force is proportional to the electropotentials of the charged spots, it follows that the bond strength is dependent upon the electropotential of the metal and the polarity and molecular weight of the adhesive. As a matter of fact, nearly all the successful metal adhesives contain one or more polar groups and this is a strong evidence that the adhesion between adhesive and metal is through the electrostatic forces.

In some cases, adhesives may contain one or more reactive groups. These groups react with metal in the process of bonding to form metallic compounds.

Modified vinyl chloride-acetate resin VMCH, for example, is a tripolymer of vinyl chloride, vinyl acetate and maleic acid. By including as low as one per cent of the acid in the polymerization a marked increase in adhesion as compared to vinyl chloride-acetate resin VYNW is obtained. In this case, some degree of a primary valence bond of the coordinate type is formed. The adhesive forces as a whole are primary and secondary valence in character.

Discussion of Results

This section contains two parts. The first part is a discussion of the factors affecting the bond strength between polyvinyl chloride sheet and metal. This includes surface conditions, bonding temperature, nature of metal, nature of adhesive and the amount of plasticizer in adhesive. The second part discusses the requirements for a good adhesive.

Surface Conditions. Three methods were used in this investigation to prepare a metal surface for bonding. These were: (1) degreasing, (2) polishing and degreasing, and (3) degreasing and etching. From Table V, page 50, and Figure 7, page 58, it can be seen that the low carbon

steel laminates in which the metal was polished and degreased had the highest strengths. Those in which the metal was degreased and etched had the medium strengths, and the laminates in which the metal was treated by degreasing alone had the lowest values. In regard to the results obtained by the degreasing method, it is to be expected that the lowest strength will be obtained because some of the impurities such as metallic oxide and dirt can not be removed by this method and the surface activity is therefore reduced.

Table I, page 46, and Figure 3, page 54, present the bond strengths of aluminum laminates. It can be seen that the etched samples had higher strengths than the polished ones. The same results were obtained from brass, copper, nickel and stainless steel laminates. They all showed etched samples had stronger bonds than polished samples.

It can be seen from these data that the degreasing and etching method gave better results for all kinds of metal laminates except low carbon steel. This is because (1) the metal treated by etching has a cleaner surface for bonding and a higher surface activity, (2) the etching of a metal provides some degree of roughness, and this will increase the actual bonding area and cause mechanical anchoring. The fact that polishing is better than etching

for low carbon steel can be explained from the standpoint of salt contamination. Since the low carbon steel was etched by a mixture of hydrochloric and sulfuric acids, iron chloride and iron sulfate were formed on the surface of the metal. Iron chloride which is soluble in water could be removed by subsequent washing. Iron sulfate, on the other hand, is only slightly soluble in water, subsequent washing could only remove a part of it. When the plastic was bonded to the etched low carbon steel, the adhesive was adhering mainly to the iron sulfate instead of the base metal. Thus, the bond strength would probably be weaker than that obtained with polished metal even though it had greater roughness.

Crowdson⁽²⁾ obtained a similar result in the preparation of the surface of low carbon steel for bonding to plasticized polyvinyl chloride sheet. He used different acids to etch the metal and found that the metal untreated with acid gave the greatest bond strength for the laminate.

From the results obtained by different methods, it can be concluded that the bond strength is related to the cleanliness and roughness of the metal surface. The cleaner and rougher the surface, the stronger is the bond.

Bonding Temperatures. A study of the effect of bonding temperature on bond strength was made by changing the bonding temperature while other variables were held constant. The laminates were bonded at 70°, 150°, 250° and 350° F, and the results were studied. From Tables I, IV, V and VI, pages 46, 49, 50 and 51 respectively, it can be seen that the bond strengths of aluminum, nickel, low carbon steel and stainless steel laminates did not change definitely with the bonding temperature. But Tables II and III, pages 47 and 48, show that the bond strengths of brass and copper laminates seemed to increase with the increase of bonding temperature. The effect of bonding temperature on strength varies with different metals. Some change their bond strengths with bonding temperature, but others do not change at all. The variation in strength of aluminum, nickel, low carbon steel and stainless steel laminates bonded at different temperatures may be regarded as experimental error rather than the effect of bonding temperature. The reason for this interpretation is that they do not change definitely with bonding temperature.

The fact that the effect of temperature on strength varies with different metals can be explained from the standpoint of electrostatic forces. When an adhesive comes

in contact with a metal, an electrostatic field is built up. The electrostatic forces of this field cause the polar groups to orientate. An increase in temperature will increase the fluidity of the adhesive and this might help the adhesive molecules to get better orientation, and a stronger bond results. On the other hand, if the electropotential of the metal is high, the force produced might be strong enough to cause complete orientation of the polar groups. An increase in temperature will have no effect as far as orientation of the adhesive is concerned. In this case, the bond strength will not change with the bonding temperature. Because brass and copper are of low order in the galvanic series, their electropotentials are rather low. An increase in temperature helps to produce a stronger bond.

As previously pointed out, the magnitude of the electrostatic force is proportional to the electropotential of the metal and the polarity of the adhesive, so the type or nature of the adhesive also has influence on this factor. Moreover, the temperature range chosen for study must be specified. Some laminates might have a steady bond strength in one temperature range but a variable bond strength in another range. It may be concluded that the effect of bonding

temperature on strength depends on the nature of metal, type of adhesive, and the temperature range chosen for study.

Nature of Metals. It has been emphasized under the section of theory of bonding that the adhesion between adhesive and metal is due to the electrostatic force. Because the magnitude of an electrostatic force is proportional to the potential difference of the charged spots, it follows that the bond strength is dependent upon the electropotential of the metal and the polarity and molecular weight of the adhesive.

In accordance with this postulation, it is expected that the bond strengths of different metal laminates follow the order of these metals in the galvanic series. The galvanic series is an arrangement of metals or alloys which indicates their tendencies to form electrochemical cells. In other words, they are arranged in the order of decreasing electropotentials, so that one can predict the probable direction of the galvanic effects. According to Whitehead Metal

(11)
Products Company, the metals used in this investigation have the following order in the galvanic series.

<u>Galvanic Series</u>	<u>Order of Bond Strengths of Laminates</u>
Aluminum	Low Carbon Steel
Low Carbon Steel	Aluminum
Stainless Steel (active)	Stainless Steel
Nickel (active)	Nickel
Copper	Copper
Brass	Brass

The average bond strengths of polished specimens of low carbon steel, aluminum, stainless steel, nickel, copper and brass were 19.6, 15.5, 14.2, 8.5, 5.5, and 2.5 pounds per inch respectively. Also from Figure 9, page 60, it can be seen that the bond strengths of these laminates had the order shown above. Since the metals of these laminates had been polished to the same degree of smoothness before the adhesive was applied, the surface factor was therefore eliminated and the bond strengths could be compared. It has been found that the bond strengths follow the order of the metals in the galvanic series except aluminum. The reason that aluminum laminate fell in the second place instead of the first might be due to the formation of an

oxide film. Aluminum is an active metal, it reacts readily with oxygen leading to the formation of aluminum oxide. When a freshly prepared surface is exposed to the atmosphere, it is quickly oxidized and a film of aluminum oxide is formed on the surface. This film acts as a coating on the metal to prevent further oxidation, and at the same time it reduces the surface activity of aluminum. When the plastic was bonded to the metal, the adhesive was actually in contact with the oxide film instead of the base metal. Thus, the bond strength was considerably reduced.

Types of Adhesives. In accordance with the postulation given above, it is to be expected that the polarity of the adhesive is a factor on bond strength. Several adhesives of different polarities were used in this investigation. All of them were vinyl resins. According to Schmidt and Harlies⁽¹⁷⁾, chlorine bearing vinyl resins may be classified as low polarity, vinyl esters as medium polarity, and polyvinyl alcohol as

high polarity. So the adhesives used in this investigation can be arranged in the order of increasing polarity as:

Vinyl Chloride-Acetate Resin, VYNW

Modified Vinyl Chloride-Acetate Resin, VMCH

Vinyl Acetate Resin, A-70

Vinyl Alcohol-Acetate Resin, T-24-9

Vinyl Alcohol-Acetate Resin, MA-28-18

The difference between VYNW and VMCH is that VYNW is just a copolymer of vinyl chloride and vinyl acetate, but VMCH is a tripolymer of vinyl chloride, vinyl acetate and maleic acid. The amount of maleic acid in this adhesive is one per cent. The resin T-24-9 differs from MA-28-18 in that T-24-9 contains a lower hydroxyl content than MA-28-18.

Tests revealed that A-70 and MA-28-18 did not adhere to VYNW which was coated on plastic. The bonds were easily broken by hand at the interfaces of A-70 and VYNW and of MA-28-18 and VYNW. Similar results were obtained with A-70 and MA-28-18, which did not adhere to the plastic. The bonds were easily broken by hand at the interface between plastic and adhesive. This can be explained by the relative cohesive force and adhesive force. If the cohesive force of one material is much higher than its adhesive force to another

material, these two materials will have high interfacial tension and they are incompatible. A very weak adhesion is obtained.

In order to study the effect of polarity on bond strength, a series of metal-metal laminates were made by bonding with adhesives of different polarities. As previously postulated, the adhesion between adhesive and metal is due to the electrostatic force acting between them. The magnitude of this force is determined by the polarity of the adhesive and the electropotential of the metal. It is expected that high polarity adhesive will give a strong bond.

Table VIII, page 53, presents the bond strengths of different adhesives. It was found that the bond strength increased with the increase in polarity of the adhesive. The only exception in this case was VMCH which gave the highest strength in this series. The following is the

comparison of polarity to bond strength obtained in low carbon steel to low carbon steel bonding.

<u>Polarity of Adhesive</u>		<u>Order of Bond Strength</u>
VYNW	Low	VYNW
VMCH	↓	A-70
A-70		T-24-9
T-24-9		MA-28-18
MA-28-18	High	VMCH

The bond of VYNW failed at the interface between VYNW and low carbon steel. This indicated that the cohesion of the adhesive was stronger than the adhesion between adhesive and metal. In the case of A-70, the bond failed at the interface and in the adhesive layer, but a greater part of the failure was at the interface. This indicated that an increase in adhesion was obtained, but the cohesion was still stronger than the adhesion. Laminates bonded by T-24-9 also failed at the interface and in the adhesive layer, but most of the failure was in the adhesive. Resins MA-28-18 and VMCH had good adhesion to the metal, the bonds were broken in the adhesive layers with the adhesives

left on both of the adherends. In this case, the adhesion was much stronger than the cohesion, and the cohesive force within the adhesive layer determined the strength of the bond. Since the cohesive force of an adhesive is determined by the intermolecular attraction, so any factor which may affect the intermolecular attraction will come into effect in case the adhesion of adhesive to metal is very strong.

The marked increase in bond strength given by VICH was undoubtedly due to the radical of maleic acid. It was supposed to react with the metal to form a metallic compound, and some degree of primary valence bond of the coordinate type was formed. This resulted in the strong adhesion to the metal.

Amount of Plasticizer in Adhesive. Table VII, page 52, and Figure 10, page 61, show the effect of plasticizer DOP on bond strengths when the laminates were subjected to stretching. It can be seen from the figure that the laminates bonded by adhesives containing different amounts of plasticizer showed different behavior to stretching. Laminates bonded by adhesive of zero per cent DOP showed a marked decrease in strength when they were stretched, and the curve had the steepest slope at this point. This effect diminished as the amount of plasticizer in adhesive

increased. Laminates bonded by adhesive of 20 per cent DOP content decreased in strength very slightly when they were subjected to stretching. The slope of the curve was nearly zero.

The addition of plasticizer increases the distance between molecules, so the intermolecular attraction force is reduced. This results in a decrease of bond strength but an increase in flexibility and extensibility. Whenever the laminates are used in a bending or deep drawing process, it is recommended that a certain amount of plasticizer should be added to the adhesive.

Requirements for a Good Adhesive. Adhesives used for different purposes vary in many ways. In a number of uses the most important consideration in choosing an adhesive are the cost and strength; in other uses, it may be the working properties and durability.

Certain working properties are desirable in a large number of different applications. For example, the adhesive should be capable of being stored, without deterioration, for a period of time. It should be easy to prepare for application and should have a sufficiently long working life without foaming or changing viscosity.

As far as strength is concerned, an adhesive should contain active groups that will combine with or strongly

adhere to the adherend surfaces. This results in forming primary valence bonds or secondary valence bonds depending on whether reaction takes place. In the case of plastic-metal bonding, the adhesive should have a composition similar to that of the plastic and at the same time possess active groups to enable it to adhere to the metal. The reason for choosing an adhesive of similar composition lies in the fact that the adhesive and plastic will have a low interfacial tension and this enables the adhesive to penetrate into the plastic and hold to it by secondary valence forces. The function of the active groups is evident. For reasons previously indicated, they serve to adhere to the metal and thus complete the joint. However, active groups in the adhesive must be kept within limits. This is because they usually increase cohesion within the ultimate adhesive layer, reducing its plasticity and increasing its brittleness. If carried too far, this may result in a weak joint.

In dealing with durability, an ideal adhesive should be resistant to weather condition, water, solvent and chemicals. The bond strength should not change markedly over a long period of time. Unfortunately, an adhesive containing active groups which make it valuable for adhesion is

easily attacked by water and chemicals. Polyvinyl alcohol-acetate, for example, is excellent in metal-metal bonding. But the hydroxyl groups in this adhesive make it easily attacked by humidity, thus weakening the bond.

Although there are a few versatile adhesives that are generally applicable to most of the bonding processes, an adhesive having outstanding properties for all types of surfaces and bonding problems is impossible. The choice of adhesive for a given purpose depends on the relative merits of these factors. As in all other fields, research on adhesives is toward improved properties in particular directions so that many modern adhesives are synthesized for specific applications.

Recommendations

The following recommendations are offered as suggestions for further study in the field of bonding plastic sheeting to metal.

1. The method used in this investigation to measure the bond strength is essentially a peeling test. It measures the greatest force required to peel the plastic off the metal. This force is actually the strength of a particular line in the joint, it can not represent the strength of the whole bonding area. The value of this test is doubtful for two reasons.

Firstly, actual rupture of an adhesive joint will occur at the weakest point of the assembly, but this method reports the strongest point of the joint. It is possible that two samples may have the same strong points but different weak points. In this case, the sample which has the weaker point of these two will rupture first when the laminates are in actual service.

Secondly, the stresses to which a plastic sheet-metal laminate may be subjected in actual service are shear, compression, impact and flexural forces rather than peeling. So, it is doubtful that peeling test can represent the actual bond strength.

For reasons indicated above, it is recommended that other methods should be used in order to evaluate the actual bond strength. Because of the thickness, compression, impact and flexural properties of a laminate can not be tested by ordinary methods. In view of this, it is suggested that a small ball mill be used for this purpose. This ball mill should have every specification fixed, including shape, inside diameter, number and weight of balls, and speed of revolution. The specimen of specified size will be put into the ball mill and the balls allowed to strike on it during revolution. The stress which the specimen would suffer should be a combination of shear, compression, impact and flexural forces. The time required for the plastic to part from the metal will be counted, and this will serve as a standard to measure the bond strength. In case the plastic wears out before it is separated, bigger balls should be used. This is just a proposed method for testing the bond strength. It surely needs criticism.

2. It is recommended that the effect of adhesive thickness on bond strength be studied between the range of 0.001-0.006 inch.

3. From the results obtained, it can be seen that the bond strengths of different metal laminates followed the order of these metals in the galvanic series except aluminum. The strength reduction of aluminum laminate might be due to the formation of an oxide film. It is suggested that aluminum be treated under an atmosphere of inert gas before the adhesive is applied. In this case, the aluminum should not be oxidized, and the reason of strength reduction can be determined.

4. It is suggested that low carbon steel be treated with acids which may form soluble salts. This would serve to determine if the strength reduction of the etched low carbon steel laminate is due to the metallic salts left on the metal.

5. It is recommended that other types of plastic, such as polyethylene and cellulose, be bonded to metal, so that the postulations given in the discussion can be checked.

Limitations

This investigation was conducted under the following limitations.

1. The metals used in this investigation were limited to aluminum, brass, copper, nickel, low carbon steel, and stainless steel.

2. The adhesives were limited to vinyl chloride-acetate resin, VYNW, modified vinyl chloride-acetate resin, VMCH, vinyl acetate resin, A-70, vinyl alcohol-acetate resin, T-24-9, vinyl alcohol-acetate resin, MA-28-18. Since only the first two adhesives adhered to plasticized poly-vinyl chloride, they were used to bond the plastic to metal for bond strength test.

3. The methods used to treat the metal were limited to solvent degreasing, polishing and degreasing, and degreasing and etching.

4. The plastic was bonded to metal at 70^o, 150^o, 250^o and 350^o F under a pressure of 200 pounds per square inch. The time of loading from zero to 200 pounds was 10±1 seconds. As soon as the desired pressure was obtained, the pressure was released. The actual temperature of the metal was not measured, the temperature of the hot plates was taken as the bonding temperature.

5. To obtain the effect of amount of plasticizer on bond strength, the amount of dioctyl phthalate in modified vinyl chloride-acetate resin, VMCH, was varied from 0, 5, 10, 15, to 20 per cent. This was used in bonding plastic sheet to low carbon steel, the laminates of which were stretched to 10, 20, and 30 per cent elongation.

6. The greatest force required to peel the plastic off the metal was regarded as bond strength. Prior to testing, the specimens were conditioned at 75° F and 50 per cent relative humidity for 30 minutes.

7. The steel-steel laminates were bonded at 250° F under a pressure of 200 pounds per square inch for 2 minutes. The shear strength of these specimens was tested. Before testing, they were conditioned at 72° F and 50 per cent relative humidity for one hour.

V. CONCLUSIONS

The investigation of the effect of surface conditions, of bonding temperatures, of nature of metals, of types of adhesives, and of amount of plasticizer in the adhesive upon the bond strength between plasticized polyvinyl chloride sheet and metal led to the following conclusions.

1. The highest bond strengths, in pounds per inch, of the etched specimens were: aluminum, 27; stainless steel, 23; nickel, 19.5; low carbon steel, 15; copper, 14; and brass, 8.

2. The highest bond strengths, in pounds per inch, of the polished specimens were: low carbon steel, 22; aluminum, 17.5; stainless steel, 16; copper, 12; nickel, 11; and brass, 6.

3. The highest bond strength of degreased low carbon steel laminates was 11.5 pounds per inch.

4. The bonding temperature had no definite influence on the bond strengths of aluminum, nickel, low carbon steel and stainless steel laminates.

5. The bond strengths of copper laminates increased with bonding temperature from 4 pounds at 70° F to 14

pounds at 350° F for the etched specimens, and from 3 pounds to 12 pounds for the polished specimens.

6. The bond strengths of brass laminates increased with bonding temperature from 4 pounds at 70° F to 8 pounds at 350° F for the etched specimens, and from 1 pound to 6 pounds for the polished specimens.

7. The average bond strengths, in pounds per inch, of the polished specimens were: low carbon steel, 19.6; aluminum, 15.5; stainless steel, 14.2; nickel, 8.5; copper, 5.5; brass, 2.5. The strengths of these metal laminates followed the order of these metals in galvanic series except aluminum.

8. The adhesion between adhesive and metal varied with the polarity of the adhesive. Vinyl chloride-acetate resin, VVW, which had low polarity gave a shear strength of 475 pounds per square inch for steel-steel laminate bonded at 250° F, whereas vinyl alcohol-acetate resin, MA-28-18, which had high polarity gave a shear strength of 903.5 pounds per square inch.

9. The bond strength was also affected by the amount of plasticizer in the adhesive. With modified vinyl chloride-acetate resin, VMCH, as adhesive, the best result was obtained with five per cent dioctyl phthalate.

10. The bond strength after stretching decreased with increase of plasticizer in the adhesive.

11. Vinyl acetate resin, A-70, and vinyl alcohol-acetate resin, MA-28-18, did not adhere to plasticized polyvinyl chloride sheet.

VI. SUMMARY

The bonding of plastic sheeting to metal is a recent development of the surface coating industry. The product, a pre-finished material, has the strength of the base metal and exterior finish of the plastic. It is used in making radio and television cabinets, furniture, and chemical containers.

In order to improve the bonding practice, it is necessary for the engineer to be well informed of the fundamental principles involved. A knowledge of the theory of bonding, the nature of the bond, and the factors affecting the bond strength would be helpful in attaining this goal.

The purpose of this investigation was to study the factors affecting the bond strength between polyvinyl chloride sheeting and metal.

The metals used in this investigation were aluminum, brass, copper, nickel, low carbon steel, and stainless steel. The plastic sheeting was bonded to the metal by means of synthetic adhesive. Five different commercial adhesives were used. These were vinyl chloride-acetate resin, VYNW, modified vinyl chloride-acetate resin, VMCH, vinyl acetate

resin, A-70, vinyl alcohol-acetate resin, T-24-9, and vinyl alcohol-acetate resin, MA-28-18.

The specimens of metal were treated as follows. Three sets of specimens were prepared. The first set was degreased with solvent only, the second one was polished and degreased, and the third one was degreased and etched. Semi-rigid polyvinyl chloride sheeting was bonded to the metal at a temperature of 70°, 150°, 250°, and 350° F under a pressure of 200 pounds per square inch. A series of the bonded specimens was stretched to 10, 20, and 30 per cent elongation. A series of steel-steel laminates was prepared. They were bonded by different adhesives at 250° F under 200 pounds per square inch. The specimens of this series were tested for shear strength.

The effect of surface conditions, of bonding temperatures, of nature of metals, of types of adhesives, and of amount of plasticizer in the adhesive upon the bond strength between polyvinyl chloride sheet and metal was determined and the following conclusions were reached:

1. The bond strength was affected by surface conditions. Etched specimens had higher bond strength than polished ones except low carbon steel.

2. The bonding temperature had no definite influence on the bond strengths of aluminum, nickel, low carbon steel, and stainless steel laminates.

3. The bond strengths of brass and copper laminates increased with the bonding temperature.

4. The average bond strengths of polished specimens of low carbon steel, aluminum, stainless steel, nickel, copper, and brass were 19.6, 15.5, 14.2, 8.5, 5.5, and 2.5 pounds per inch respectively. The strengths of these metal laminates followed the order of these metals in the galvanic series except aluminum.

5. The adhesion between adhesive and metal varied directly with the polarity of the adhesive.

6. The bond strength was also affected by the amount of plasticizer in the adhesive. When the laminates were subjected to stretching, the rate of decrease in strength was inversely proportional to the amount of plasticizer in the adhesive.

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