

IMPROVE THE PERFORMANCE OF EPOXY RESIN AND POLY (VINYL BUTYRAL) AS AN ALUMINUM METAL ADHESION⁺

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Abstract:

The present work is mainly concerned with study the performance of epoxy resin which blended with poly (vinyl butyral) in order to be used as adhesive for aluminum metal. A different weight ratios of poly (vinyl butyral) was blended with epoxy resin (10-90 wt %) and these blends were cured at different conditions; time (10-60) minutes and temperature (100-200 C^o), different mechanical properties (shear stress, elongation and peeling stress) were carried out on the prepared samples.

Box – Wilson design method was adopted to find useful relationships between the three variables [(weight ratios and curing conditions (time & temperature)] with mechanical properties shear stress, elongation and peeling stress , it was found that the best adhesive properties of the prepared sample (i.e. best mechanical properties) obtained with the weight ratio of (40) wt% epoxy resin and (60) wt% poly (vinyl butyral) while the best curing condition obtained at a temperature of 160 C^o for (30) min.

المستخلص :

البحث الحالي يدرس تحسين أداء راتنج الايبوكسي بعد خلطه مع بولي فنيل بيوتيرال وذلك لاستعماله لاغراض لصق معدن الالمنيوم ، تم استخدام نسب وزنية مختلفة من راتنج الايبوكسي مع بولي فنيل بيوتيرال تتراوح بين (١٠-٩٠ %) كنسبة وزنية وهذه المخاليط تم انصاجها في ظروف مختلفة من درجات حرارة وزمن حيث تم استخدام درجات حرارة إنصاج تتراوح بين (١٠٠-٢٠٠) درجة سيليزية وزمن انصاج يتراوح بين (١٠-٦٠) دقيقة ، بعد ذلك تم قياس الخواص الميكانيكية (جهد القص ، الاستطالة وجهد التقشر) للنماذج المحضرة. تم استخدام تقنية Box – Wilson design من اجل ايجاد علاقات تربط المتغيرات الثلاثة (النسب الوزنية المختلفة ، درجة حرارة انصاج وزمن إنصاج) مع فحص الخواص الميكانيكية المختلفة وهي جهد القص ، الاستطالة وجهد التقشر ، حيث وجد أن احسن خواص لصق والتمثلة بافضل خواص ميكانيكية تم الحصول عليها عند النسبة الوزنية ٤٠% من راتنج الايبوكسي و٦٠% من بولي فنيل بيوتيرال بينما احسن درجة حرارة إنصاج كانت هي ١٦٠ درجة سيليزية وعند زمن إنصاج قدره ٣٠ دقيقة.

Introduction:

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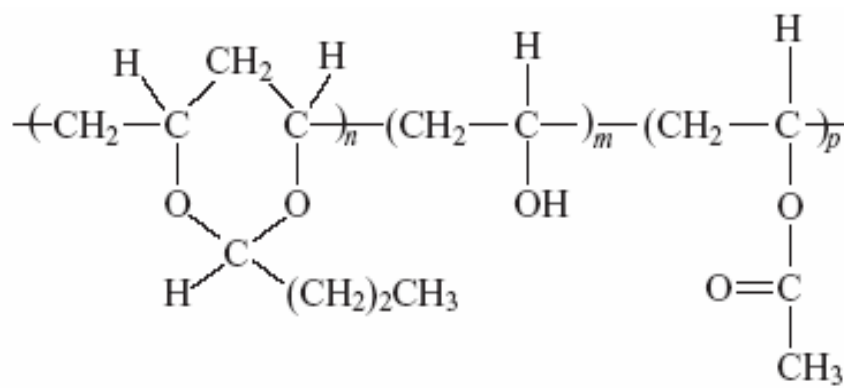
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Poly (vinyl butyral) classically belongs to a family of polymer termed as poly (vinyl acetal)s, poly (vinyl acetal)s were commercialized during 1930 and 1940 following development work at a number of firms including Dupont Shawiniquan Chemicals (now Monsanto) , Union carbide and other in the United States and Canada [1], only poly (vinyl acetal)s commercially available are poly (vinyl formal) and poly (vinyl butyral).

Poly (vinyl butyral) is mainly used in safety glass, adhesive and surface coating, the significant use is in lamination of safety glass (automotive windshields) ,other are structural adhesives , binders of rocket propellants , ceramics , in metallized brake linings and offset printing plates , in wash prime for protecting metal surface (e.g. naval vessels).[2].

Poly (vinyl butyral) is a member of the class of poly (vinyl acetal) resins, it is derived by condensing poly (vinyl alcohol) (PVA) with butyraldehyde in the presence of strong acid, poly (vinyl alcohol) reacts with the aldehyde to form six- membered rings primarily between adjacent , intramolecular hydroxyl group, leading to the structure shown below. [3].



Theory:

Poly (vinyl butyral) is used in combination with other thermoplastics and thermosetting resin to form tough strong structural adhesive , the most important adhesive for gluing of metals are bisphenol epoxy resins, phenolic resins modified by the addition of poly (vinyl acetal) [poly (vinyl butyral) and poly (vinyl formal)] and poly urethane adhesives [4].

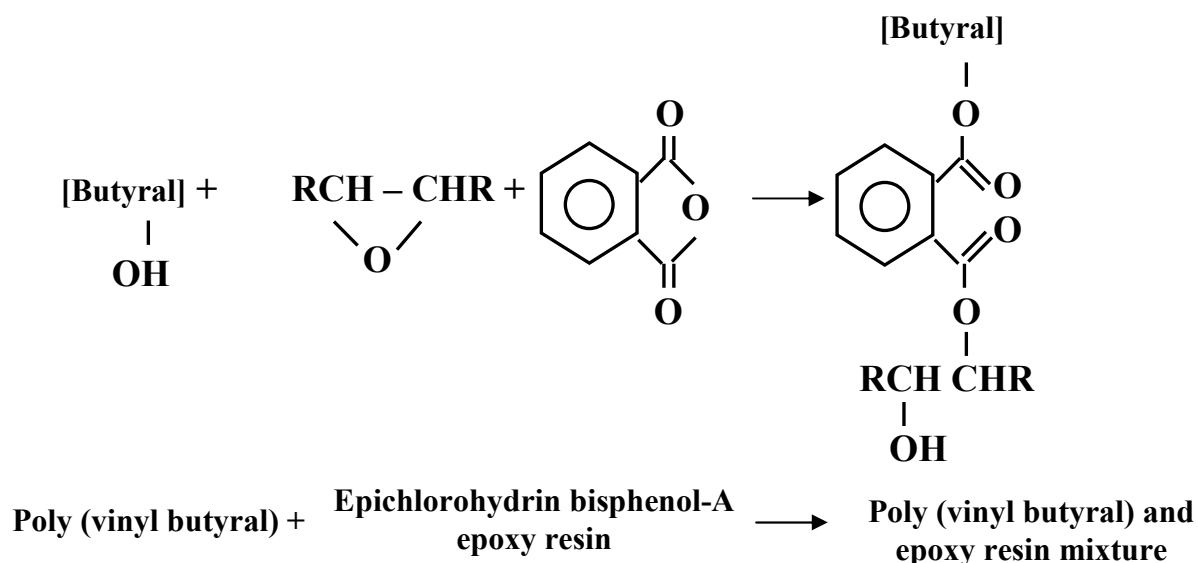
Epoxy or polyepoxide is a [thermosetting epoxide polymer](#) cured (polymerizes and crosslinks) when mixed with a [catalyzing](#) agent or "hardener". Most common epoxy [resins](#) are produced from a reaction between [epichlorohydrin](#) and [bisphenol-A](#). The first commercial attempts to prepare resins from epichlorohydrin occurred in [1927](#) in the [United States](#).

Epoxies are known for their excellent adhesion, chemical and heat resistance, good to excellent mechanical properties and very good [electrical insulating](#) properties, but almost any property can be modified, [5].

Many investigations studied the reaction between poly (vinyl butyral) and epoxy resin in order to prepare a suitable resin used for aluminum adhesive, Robert and Edward studied the mechanism of the reaction, they made many experiments depending on viscosity in order to know the reaction group in both resins, and they show that for epoxy resin and poly (vinyl butyral) the reactive group is hydroxyl group [6].

The chemical reaction of poly (vinyl butyral) is the characteristic reaction of the functional groups: the cyclic acetal, alcohol and acetate, the most reactive moiety is the hydroxyl group that undergoes reactions typical of hindered secondary alcohol, reaction via

the hydroxyl group with difunctional reactants provides a convenient method of cross-linking, and typical cross-linking reactions are shown below [1].



Reaction of poly (vinyl butyral) with epoxy groups

Jackson E.H. showed that in order to prepare a suitable adhesive for aluminum metal the mixture should consist of 80 gm of poly (vinyl butyral) and 30 gm of epoxy resin cured at temperature 180 C° all dissolved in 230 gm of ethanol solution[13].

Sumitomo Bakelite Co. showed that the adhesive for printed circuit board made from aluminum metal composed of the mixture of an epoxy resin, poly (vinyl butyral) and acrylonitrile butadiene copolymer [7], moreover “Matsuchita Electric Works” study the mixture of epoxy, phenolic and poly (vinyl butyral) composition used for the same application [8].

In this work poly (vinyl butyral) is blended with epoxy resin to prepare a suitable resin used for aluminum adhesive, for this purpose this work deals with finding the best condition for mixing poly (vinyl butyral) and epoxy resin to obtain good adhesive for aluminum metal.

Experimental Design:

In general, the purpose of an experimental design is to find useful relationships between controllable variable and observed response, in order to predict such relationships it can specify a combination of variable that will achieve some practical benefits, in the chemical industry, experimental design particularly applied to the study of process variable and how to affect the product, [9].

This analysis give a description for the system by correlation in order to predict the effect regarding the change of variables on the objective function, statistical software used to analysis of the mechanical properties testes that exhibits [shear stress, elongation and peeling stress) at different condition for [epoxy resin concentration wt%, curing time (t) and curing temperature (T)].

As mentioned this analysis will give a description of the system by a correlation relating all real variable to the objective function by a polynomial will the general form as shown in equation (1) [10].

$$Y=B_0+\sum B_iX_i+\sum B_{ii}X_i^2+\sum\sum B_{ij} X_iX_j+\dots\dots\dots i<j \dots\dots\dots (1)$$

Where:-

Y= objective function.

X_i, X_j = Controllable Variable

B_0, B_i & B_j = quadratic polynomial coefficients

K_i = number of variables in the quadratic polynomial model can be determined as follows.

$$\text{No. of terms} = [(K_i+1)(K_i+2)/2]\dots\dots\dots (2)$$

For three variables the quadratic polynomial can be represented as follows.

$$Y=B_0+ B_1X_1+ B_2X_2 +B_3 X_3+B_{11}X_1^2 + B_{22}X_2^2 + B_{33}X_3^2 + B_{12}X_1X_2+ B_{13}X_1X_3+ B_{23}X_2X_3\dots\dots\dots (3)$$

Experimental Work:

Complementary preparation of poly (vinyl butyral) were performed in the literature [11], in order to modify the adhesive properties of poly (vinyl butyral); epoxy resin was chosen as material to be mixed with poly (vinyl butyral), [2].

Raw Material:

Commercial poly (vinyl butyral) (B-73) consist of 80% butyral content , 17.5% hydroxyl content and 2.5% acetate content available from local substances was used during the experimental work.

Epoxy resin type (Epon 828) was used, this resin is an undiluted dysfunctional bisphenol-A /epichlorohydrin derived liquid epoxy resin manufactured by Shell Chemical Company. This resin has good mechanical, adhesive, dielectric and chemical resistance properties when cross-linked. In order to convert the resins into cross linked structures; which leads to the formation of a tightly bond three dimensional net-work of polymer chains, it is necessary to add curing agent (hardener).

The hardener used in present work for epoxy was tertiary amines which were commonly referred to as catalytic curing agent. Tri Ethylene Tetra Amine (TETA) is used as curing agent product of (Ciba Co.) trade name CY 956.

Epoxy resin was mix up with the appropriate combination ratios, component A (TETA) and component B (Epon 828) epoxy were mixed in the ratio of A: B = 1:5 by weight, using a mechanical stirrer to insure the complete mixing of the two components,[5].

Preparation of adhesion materials:

The present work was conducted in a batch process, variable percentage of epoxy resin (10-90) wt% in poly (vinyl butyral) – epoxy resin mixture were investigated, various reaction condition, time range (10-60) min and temperature range (100-200) C° were studied, two surface of aluminum metal in specific dimension depending on the desired tests, were adhesive to each other using the prepared blended resin.

Evaluation of prepared sample:

The following tests were used to evaluate the prepared sample of poly (vinyl butyral) –epoxy resin .The mechanical properties tests included [shear stress, elongation and peeling stress], shear stress & elongation tests were carried out according to the test specification of ASTM (D1002-72), [12], and moreover peeling stress test was carried out according to the test specification of ASTM (D903-49) [12].

Results and Discussion:

Mechanical properties of adhesives at optimum percentage of epoxy resin:

The results of mechanical properties tests which were carried out first on adhesion material are shown in tables (1,2 and 3) that exhibits (shear stress , elongation and peeling stress) under different conditions for each epoxy resin percentage (wt%) , curing time (min) and curing temperature (C°).

Table (1) shows the mechanical properties tests at different percentage of epoxy resin (10-90) wt% in poly (vinyl butyral) – epoxy mixture and at minimum value of curing condition [curing time (10 min) and curing temperature (100C°)], from this table it is clearly seen that the best percentage of epoxy resin in poly (vinyl butyral) – epoxy resin mixture is found to be (40) wt% this best concentration give the best mechanical properties ,thus this concentration chosen as the best in the following experiments.

Mechanical properties of adhesives at optimum curing time:

Table (2) shows the mechanical properties at different curing time rang (10-60) min and at best percentage of epoxy resin (40) wt% with minimum value of curing temperature (100) C°, from this table it is clear that the best curing time is (30) min give the best mechanical properties. Hence, best curing time (30) min was chosen for the followed experiments.

Mechanical properties of adhesives at optimum curing temperature:

Table (3) shows the mechanical properties at different curing temperature range (100-200) C° with optimum percentage of epoxy resin (40) wt% and optimum curing time (30) min ,from this table it is concluded the best value of curing temperature is (160) C° .

Finally from these tables (1, 2 and 3) we conclude that the best condition for epoxy resin used with poly (vinyl butyral) is (40) wt% epoxy resin and (160) C° curing temperature for (30) min give the best mechanical properties, the results give a good agreement with results obtained by Jackson [13].

Table (1):- Mechanical properties testes at different epoxy resin concentration (10-90) wt% cured at (100) C° temperature for (10) min .

Exp. No.	Epoxy Resin percentage . (wt)%	Mechanical Properties Testes		
		Shear stress (Kg /cm ²)	Elongation (%)	Peeling stress (Kg /cm)
1	10	17.6	77.6	1.9
2	20	17.9	78	1.93
3	30	18.2	78.6	1.97
4	40	18.5	79	2
5	50	16.2	71.5	1.65
6	60	13.7	66.2	1.23
7	70	11.5	54.3	0.95
8	80	8.4	42.6	0.54
9	90	6	39.8	0.25

Table (2):- Mechanical properties testes at different curing time (10-60) min and at optimum epoxy resin percentage (40) wt% and minimum curing temperature (100) C°.]

Exp. No.	Curing Time (min)	Mechanical Properties Testes		
		Shear stress (Kg /cm ²)	Elongation (%)	Peeling stress (Kg /cm)
1	10	18.5	79	2
2	20	18.8	80.2	2.2
3	30	19.3	81	2.3
4	40	19.2	80.9	2.3
5	50	19.3	81	2.3
6	60	19.3	81	2.3

Table (3):- Mechanical properties testes at different curing temperature (100-200) C° and at optimum epoxy resin percentage (40) wt% and optimum curing time (30) min.

Exp. No.	Curing Temperature (C°)	Mechanical Properties Testes		
		Shear stress (Kg /cm ²)	Elongation (%)	Peeling stress (Kg /cm)
1	100	19.3	81	2.3
2	120	19.7	81.8	2.45
3	140	20.4	82.6	2.5
4	160	21.6	83.4	2.6
5	180	19.4	81.1	2.33
6	200	19.9	81.7	2.4

The resulting mechanical properties (shear stress , elongation and peeling stress) are plotted versus epoxy resin percentage , curing time and curing temperature as shown in figures (1 ,2 and 3) .

Figure (1) shows the influence of epoxy resin weight ratios on the mechanical properties at minimum curing conditions [curing time (10) min and curing temperature (100) C°] , this figure show that increasing epoxy resin percentage more than (40) wt% causes a decrease in all mechanical properties , this may be attributed to the fact that poly (vinyl butyral) may act as plasticizing material while epoxy resin acts as a less plasticizing material and almost acts as a rigid material with no elasticity ,[14].

Figure (2) shows the effect of curing time on the mechanical properties at best percentage of epoxy resin (40) wt% and minimum curing temperature (100) C°, from this figure it is clear that increasing curing time leads to an increase in the cross linking between epoxy resin and poly (vinyl butyral) leading to an increase in the mechanical properties, after (30) min all the mechanical properties constant.

Figure (3) shows the influence of curing temperature on the mechanical properties at best percentage of epoxy resin (40) wt% and optimum curing time (30) min, from this figure

it is found that increasing curing temperature increase the mechanical properties which may be attributed to an increase in the cross linking at (160) C° after that the material starts lose its elasticity.

Finally from these figures (1, 2 and 3) one can conclude that poly (vinyl butyral) compatible with epoxy resin when (40) wt% epoxy resin is added to (60) wt% poly (vinyl butyral) cured at (160) C° for (30) min, the mixture was found to have a good capability to withstand stress and process high elasticity and adhesion properties which render the material suitable for being adhesive, our results agree well with results obtained by Gole and Jackson [5,13].

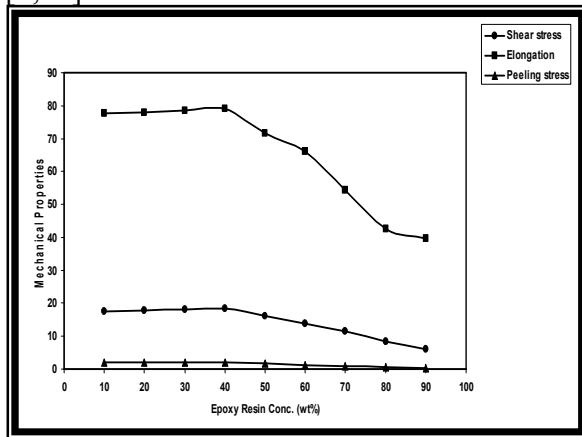


Fig. (1): Mechanical properties as a function of epoxy resin percentage (10-90) wt% and at minimum curing conditions [curing time (10) min & curing temperature (100) C°].

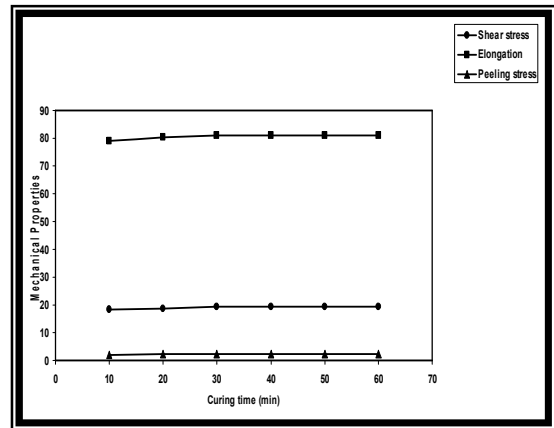


Fig. (2): Mechanical properties as a function of curing time (10-60) min and at optimum epoxy resin percentage (40) wt% and minimum curing temperature (100) C°.

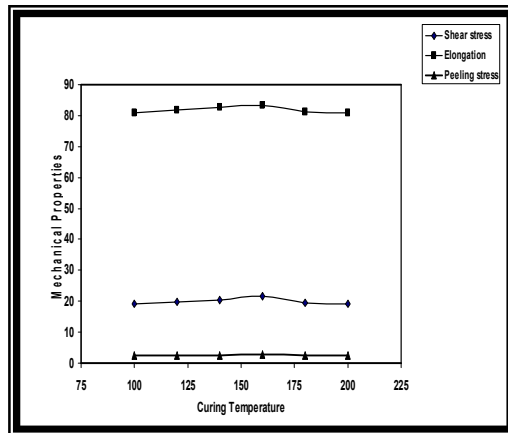


Fig. (3): Mechanical properties as a function of curing temperature (100-200) C° and at optimum epoxy resin percentage (40) wt% and optimum curing time (30) min.

5.4 Estimation of the Coefficients of the Second order Equation:

The coefficients of equation (3) for different mechanical properties testes can be determined respective by using statistical software.

For shear stress test equation (3) can be written as follows.

$$Y_1=19.34+1.189X_1-0.566X_2-1.58X_3-2.66X_1^2-0.4X_2^2-0.33X_3^3-0.077X_1X_2-0.051X_1X_3-0.052X_2X_3 \dots\dots\dots(4)$$

Correlation Coefficient =0.968

Average absolute error = 2.6%

For elongation test equation (3) can be written as follows.

$$Y_2=81.8+3.69X_1-2.67X_2-5.6X_3-8.45X_1^2-1.38X_2^2-1.26X_3^3-11.13X_1X_2-3.21X_1X_3-3.22X_2X_3 \dots\dots\dots(5)$$

Correlation Coefficient =0.984

Average absolute error = 2.12%

For peeling stress test equation (3) can be written as follows.

$$Y_3=2.41+0.16X_1-0.076X_2-0.24X_3-0.499X_1^2-0.12X_2^2-0.107X_3^3-0.003X_1X_2+0.0653X_1X_3-0.0651X_2X_3 \dots\dots\dots(6)$$

Correlation Coefficient =0.992

Average absolute error = 1.85%

These equations represent the best forms of the mathematical models that relate the mechanical properties tests with the three variables [epoxy resin concentration X_1 , curing time X_2 and curing temperature X_3].

From these equations (4, 5& 6) plots can be constructed for the relationships between experimental values and predicted values for mechanical properties testes, the plots are shows in figures (4, 5 & 6).

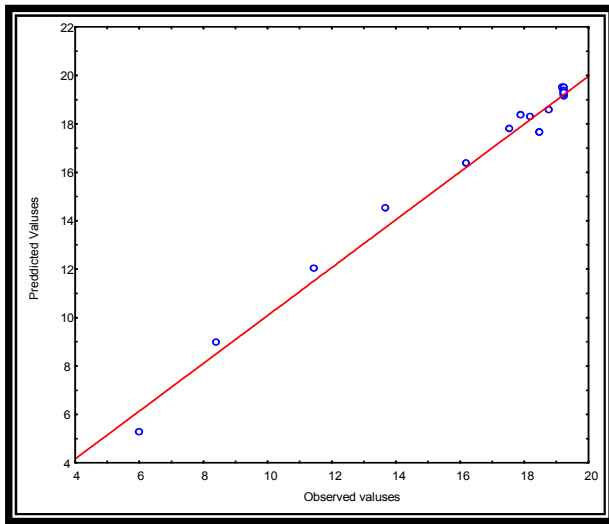


Fig. (4) Experimental versus predicted values for equation (4).

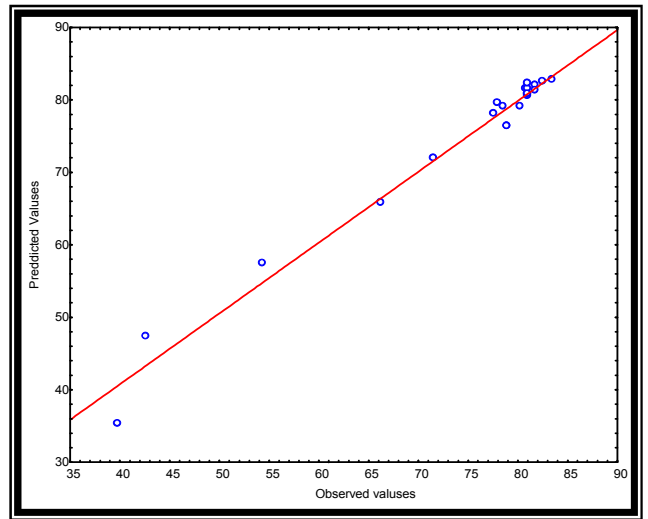


Fig. (5) Experimental versus predicted values for equation (5).

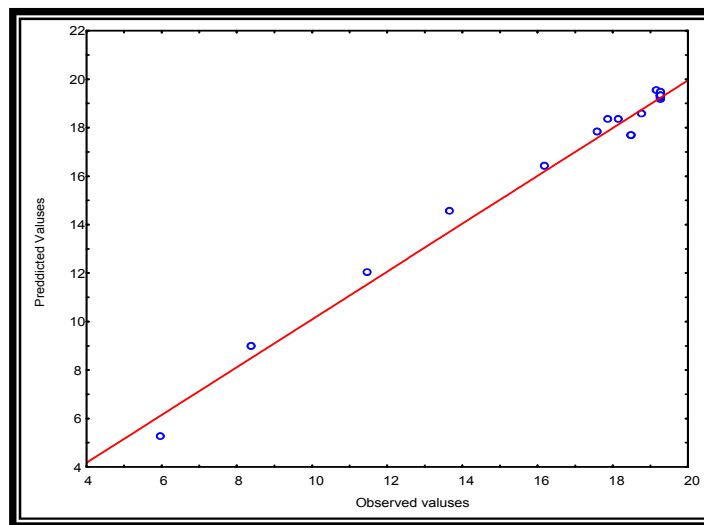


Fig. (6) Experimental versus predicted values for equation (6).

Three dimensional plots of mechanical properties (shear stress, elongation & peeling stress) versus (epoxy resin concentration wt%, curing time & curing temperature) using equations (4, 5& 6) respectively which are shows in figures (7, 8 & 9), these figures approximate the behavior of the system with reasonable accuracy for the values of (epoxy resin concentration wt%, curing time & curing temperature) in this studied range.

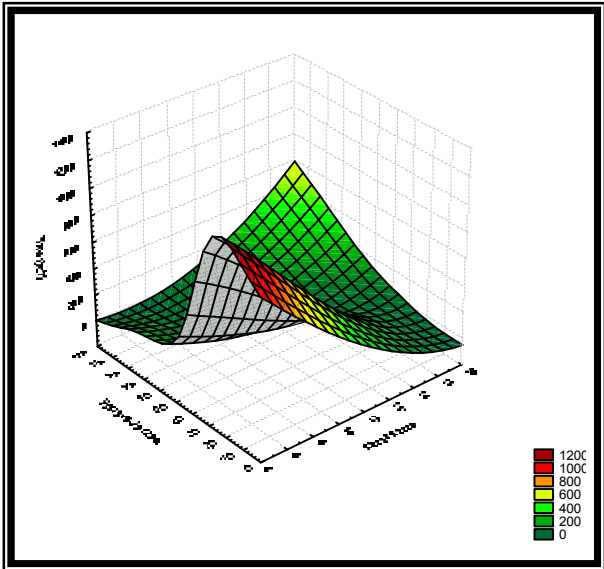


Fig. (7) Three dimensional representation of equation (4)

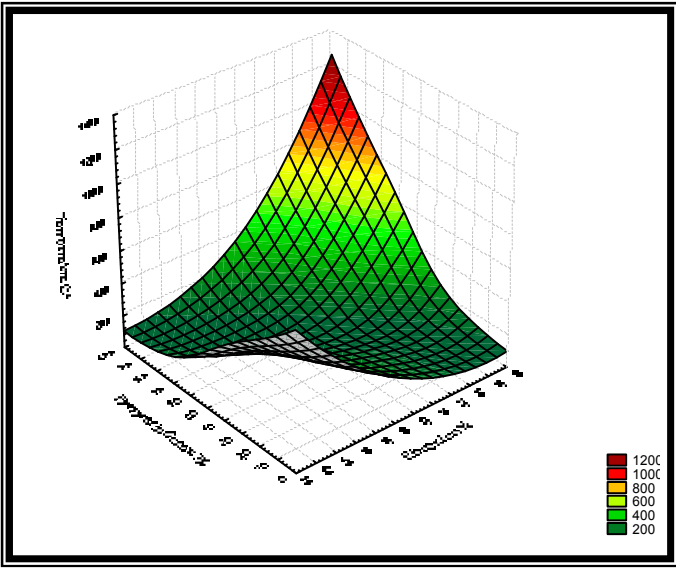


Fig. (8) Three dimensional representation of equation (5)

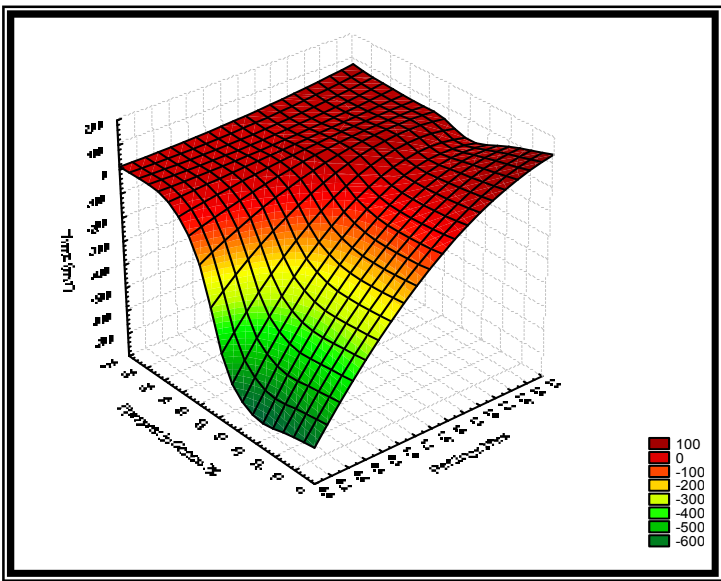


Fig. (9) Three dimensional representation of equation (6)

6. Conclusions:

- 1- Poly (vinyl butyral) is blended with epoxy resin in order to improve its mechanical properties.
- 2- Optimum condition in which poly (vinyl butyral) is compatible with epoxy resin by mixing (40) wt% of epoxy resin with (60) wt% poly (vinyl butyral) and cured at (160) C° for (30) min period which gives the best mechanical properties.
- 3- The prepared sample of poly (vinyl butyral) – epoxy resin blend is suitably to be used for aluminum metal adhesive according to the evaluation of most its mechanical properties which showed better results than those of the standard commercial grades of poly (vinyl butyral) alone.

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