Iraqi Journal of Physics, 2011 *Vol. 9, No.15, PP. 55-62*

The Effect of metals as additives on adhesion properties of epoxy resin

Asmaa Shawky Harith I. Jafeer,and Ekram AL-Ajaj.

University of Baghdad /College of Science /Physics Department

Abstract

EP/ metal composites were prepared as adhesives between two steel rods. Epoxy resin (EP) was used as a matrix with metal as fillers (Al, Cu, Fe,).

The preparation method for tensile adhesion tests includes two steel rods with adhesive composites between the rods to measure adhesion strength S_{ad} and adhesion toughness G_{ad}.

Results of tensile adhesion tests show that EP/ metals composite have maximum strength S_{ad} for certain weight percentage of metals 2.95 and 9MPa at 10% for EP/Al and EP/Cu composite and 8.2MPa at 40% for EP/Fe composites.

Key words

Adhesion strength, particulate composite

Article info

Received: Dec. 2010
Accepted: Sep. 2011
Published: Oct. 2011

تأثير المعادن كمضافات على خصائص التلاصق لراتنج الايبوكسي

اسماء شوقي ، حارث ابراهيم جعفر ، اكرام عطا العجاج

جامعة بغداد- كلية العلوم- قسم الفيزياء

الخلاصة

تضمن البحث تحضير متراكبات الايبوكسي/معدن، التي أستخدمت كمادة لاصقة بين قطعتين معدنيتين، حيث تم أستخدام الايبوكسي كأساس للمتراكبات مع أستخدام المعادن (Al ,Cu ,Fe) كمادة مالئة.

اظهرت نتائج فحوصات الالتصاق للعينات المتضمنة المتراكبات ذات الحشوات المعدنية المستخدمة كلواصق ان قوة الالتصاق لهذه العينات تكون عند نسبة معينة للمضافات المعدنية. حيث اظهرت ان اعلى قيمة لمضافات الالمنيوم والنحاس عند النسبة الوزنية ١٠% و ٤٠% لمضافات النحاس.

Introduction

Epoxy resins are basically thermosetting resins, which can be reacted with curing agent to form a cross-linked polymeric structures .Their most outstanding property is their excellent adhesion to both metallic and non-metallic surface [1]. Cured epoxy resins are characterized by their good mechanical properties, thermal and electrical insulation properties. Many particle types of fillers are used to improve the other properties of matrix materials such as mechanical, thermal and electrical conductivity

An adhesive is a substance capable to join the surface of two materials (adherends) together to produce a joint with a high strength. Adhesive is one of the oldest join techniques, but adhesive technology progressed very slowly. In the twentieth century, reports showed that adhesives were used over three thousand years ago. Blitumen and Tar pits were used in early structures. They are known to have been used as mortar by the builders of the tower of Babel. The Egyptians used glue formulated from tree resin and eggs [1].

Polymer adhesives may be used to join a large variety of materials combination: metal-metal, metal-plastic, metal-ceramic, and so on.

Adhesion of resin compounds to materials is provided mainly by: Chemical or Adsorption Adhesion, Van der Waal's Forces, and Mechanical interlocking[2].

Metal to Metal Bonding

The selection of the correct method of application of an adhesion may be as important to the success of the bond as the choice of the adhesive itself. Each different type of adhesive may require its own special type of application [3].

The design of adhesive-bonded joints involves selecting the proper geometry, consideration of the adhesive and substrates to be employed ,the size and dimensions of the joint ,and the ease by which it can be tooled for, fabricated, and mass produced. The design engineer must consider the type of structure, service requirements, mechanical strength factors, service environment, and fabrication cast. These considerations are usually borne out by testing [5].

Mostovoy et al (1971), have determined the rate of stress corrosion cracking of epoxy-aluminum bond using double cantilever beam specimens in which they follow stress corrosion cracking as a function of applied stress. They present their data as crack rate vs. strain energy release rate .They found that y approached a limiting or threshold value; y of about 1.75 to 17.5 J/m^2 For bends of epoxy resin cured [6]. Baker (1983) studied the butt joints between stainless steel/epoxy/stainless steel. He studied the effect of ultra-clean adherend surface on the strength of epoxide-stainless steel but joints. He found that the stainless steel adherend surface which cleaned with ion-bombardment will not give joints as strong as those made from specimens abraded and cleaned in the normal way[7] . Moloney et al(1983), studied the effect of the surface roughness, the thickness of adhesive layers and the combined stresses on the adhesive strength are examined for the specimens of various metals bonded with epoxy resin.the results show that, tensile adhesive strength increases as the thickness of the adhesive layer decreases [8]. Duncan et al (2004), studies the mechanical properties of two kinds of epoxy adhesives, they study interfacial adhesion strength, and a number of alternative test methods for adhesion strength-pull-off, profiled butt joint, pull –out and 3 point bend as alternative methods for quantifying adhesion strength [9].

Aim of the work-

One of the main objectives of this study is to prepare and testing samples of particulate composite, which consists of epoxy resin as a matrix, with metal and metal oxide (Al, Cu, and Fe) as fillers with different weight percentage (10,20,30,40,and 50 %), To study the effect of filler weight percentage and it's type on the adhesion force properties of EP composites with metals (steel).

Experimental

<u>Materials Used</u>

The material used to prepare the test samples were epoxy resin (EP10 Conbextra) supplied by Fost Compeny with the hardener aliphatic amine (HY 956) and particle fillers (metal and metal oxide).Table (3-1) summarizes the materials and some of their properties.

Table (3-1) Some of properties of the used material

Material	Sample	Density (gm/cm^3)	Thermal conductivity (W/m.K)	Particle size (μm)
Epoxy	EP	1.2	.19	
Aluminum	Al	2.7	247	10.2
Iron	Fe	7.3	80	15.01
Copper	Cu	8.89	398	12.42

<u>Preparation of Adhesion Samples:</u>

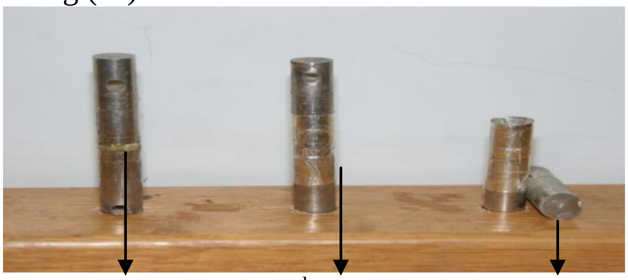
Adhesion test specimen consists of two steel rods with composite layer between their ends . A hand layup method is used to lubricate all the specimens in this study flowing these steps: (according to ASTM D-2094):

1- Two metal rods (adherend) machined from rod stock. Rough machining was done and finished with carbide. Tipped tools and grinding. The surface of the adherend metal (steel) was roughened with 220 grad.

2- Fix the lower adherend on a stage with a suitable balance and cover it with a partition around the steel rod to avoid the liquid mixed composite to flow as shown in Fig. (1-a), which is put on the roughened surface of the roughed surface of the lower adherend, then put the second adherend on the adhesion specimen (two pieces of adherend with adhesive layer between them) as shown in Fig. (1-b)

3- Leave the prepared specimen for 24 hours to cure at room temperature.

4- Clean around the specimen well to get the suitable specimens for tests as shown in Fig.(1c).



c b a

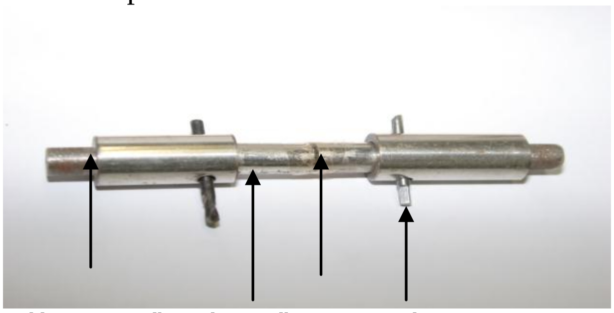
Fig. (1) Steps of preparation of adhesion sample

a- Instruments:

In order to get results for the required tests, the instruments must be prepared and calibrated by using standard samples to give more suitable results.

b- **Adhesion test**

The specimen of the tensile strength of adhesive consists of two steel cylinders bonded to each other at their ends which are roughened to subject the adhesive layer between the low ended, shown in Fig. (2). A according to the ASTM standard test method D2095 the rod type specimens was used in this method. The design of the specimens and procedures used in preparing them was in accordance with ASTM recommended practice. D2094 preparation of rod specimens for adhesion test. An Instron universal testing machine of (250 kN) full scale load capacity was used, the cross head speed was fixed in all tests to 0.1 mm/min at room temperature.



Holder Adherend Adhesive Steel Pin

Fig. (2) Adhesion test sample with holders

Results and discussion

Tensile Strength of Adhesion Force

The adhesion force between the two metal adherend depends on many parameters, such as surface roughness, kind of the filler, thickness of the adhesive, and the concentration of the filler [10]. Thus the surface roughness of the adherends is fixed, as shown in Fig. (3) as well as the adhesive thickness (1 mm) [11].

The tensile adhesion strength of the test specimens, which is calculated by dividing the breaking load by the area of the bonded surface, express this result in (MPa), and the toughness by the area under the curve show that the net epoxy specimen strength was about 1.9 MPa .

Fig. (3) The roughened surface of the adherend (10)

According to the obtained results from the tensile adhesion force, the strength v.s weight percentage for the adhesive consists of EP/Al composite as shown in Fig. (4). The results show that the maximum strength is got at (10%) Al filled EP composite, Table (2).

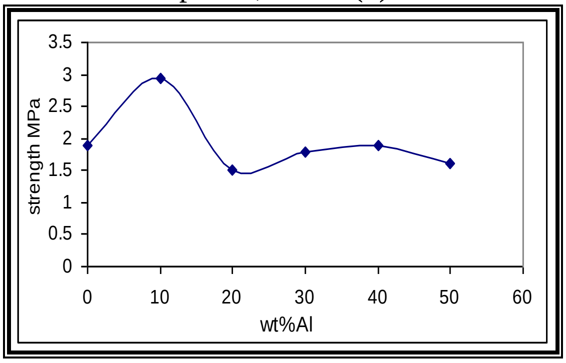


Fig. (4) The strength v.s wt% for EP /Al composites adhesive.

Table (2) Strength and toughness for tensile adhesion force (EP /Al Composites).

Materials	S_{ad} (MPa)	Toughness G_{ad} (J/m^3)
EP matrix	1.90	9.44
90% EP/Al 10%	2.95	14.45
80% EP/Al 20%	1.50	8.27
70% EP/Al 30%	1.80	5.87
60% EP/Al 40%	1.90	9.10
50% EP/Al 50%	1.60	5.88

Also, the result , for adhesive consisting of EP /Cu composites, show the increasing in strength at intended value at 10%, as shown in Fig. (5). The results show that the maximum strength value was (9.0MPa), Table (3).

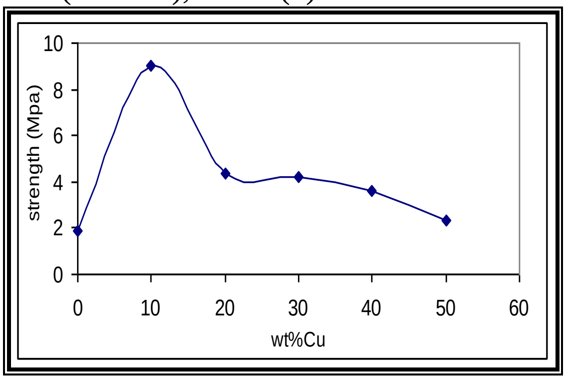


Fig. (5) The strength v.s wt% for the EP /Cu composite adhesive.

Table (3) Strength and toughness for tensile adhesion force (EP /Cu Composites).

Materials	S_{ad} (MPa)	Toughness G_{ad} (J/m^3)
EP matrix	1.9	9.44
90% EP/Cu !0%	9	186
80% EP/Cu 20%	4.35	68
70% EP/Cu 30%	4.2	28
60% EP/Cu 40%	3.6	26
50% EP/Cu/50%	2.35	19.7

EP /Fe composite adhesive gives the same mechanical behavior of EP/Cu, and EP/Al.The max value was at the (40%) weight percentage as shown in Fig. (4-26), Where the maximum value was (8.2 MPa) Table (4-11).

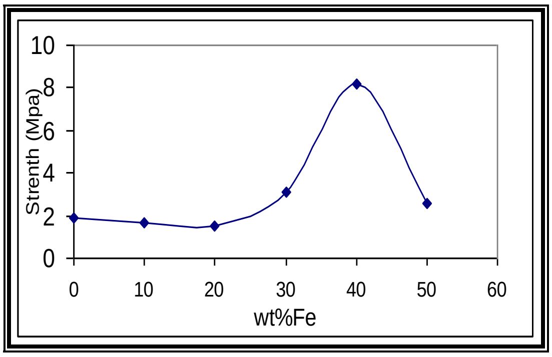


Fig. (6) The strength v.s wt% for the EP /Fe composite adhesive

Table (4) Strength and toughness for tensile adhesion force (EP /Fe Composites).

Materials	S_{ad} (MPa)	Toughness G_{ad} (J/m^3)
EP matrix	1.9	9.44
90% EP/Fe 10%	1.67	2.22
80% EP/Fe 20%	1.5	4.13
70% EP/Fe 30%	3.1	8.94
60% EP/Fe 40%	8.2	13.65
50% EP/Fe 50%	2.6	13.79

The addition of filler typically acts to decrease the flexural strength of a composite. This is normally attributed to poor adhesion at the filler /matrix interface, with the filler action as stress concentrators. From which crack s and ultimate failure is initiated [58]

For EP/Al composites adhesive specimens, the photographic pictures show that the crack propagation for the 10% weigt percentage starts from the interface pas through the adhesive as shown in Fig. (7).

That means; the adhesion between the metal and the composite adhesive is greater than the cohesive of the composite itself. These results with the results obtained by Bell [50].

To explain this mechanical behavior, the precipitate of Al particles between the grooves of the rough surface make good interlocking between the metal and the composite, at the same time the Al particles located inside the inter molecular spaces for molecular chains ,which leads to decrease the molecular chain's mobility during the test .so the existing of these particles will screen the bonds which contact between the main chain (branch bonds)so that the cross link density for the polymer will decreases with increasing weight percentage as shown in Fig. (8) . The higher Al particle weight percentage show that the decohesion happened at the interface of the adhesion surface as shown in Fig. (9), because of accumulate of particles, which prevent the resin to inter lock with the metal surface.

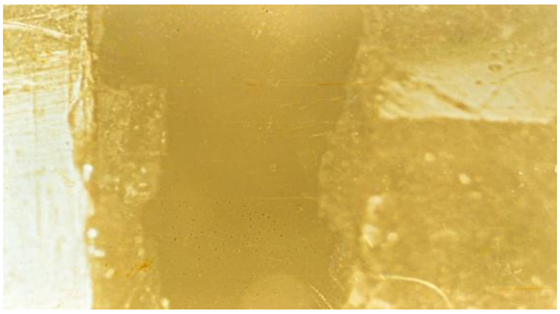


Fig. (7) 90% EP/ 10%Al composite adhesive (X 10)

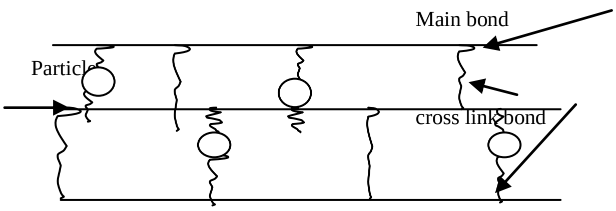


Fig. (8) The model for particles location inside intermolecular spaces.

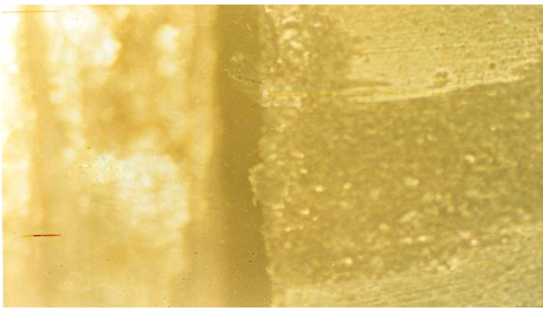


Fig. (9) 50% EP/50 Al composite adhesive X 10.

(EP/Cu) composite adhesive specimens at (10%) have maximum strength values;Fig. (10) show the surface fracture at the specimen.Figur(11) show the crack propagation from the metal/ adhesive interface through the adhesive and the adherend. The obtained results show that with increasing of the filler (wt %), the bonding between the adhesive and the adherend decrease, and the fracture happened at the interface (at high weight percentage) as shown in Fig. (12), leaving some of the adhesive material or the metal surface as shown in Fig. (13).That means that , the interlocking decreases between the adhesive and adhernd, with the increasing of additive weight percentage.

In this case, the shape and the size of the particles will be effective, because the accumulation of particles, which make a layer prevent the resin to make a good adhesion with the adherend surface.

Fig. (10) The composite adhesive 90%EP/10%Cu (X 10).

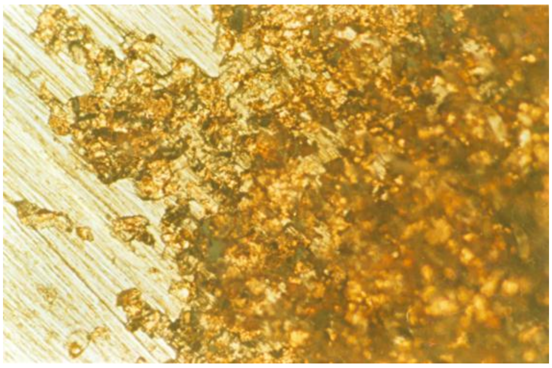


Fig. (11) The composite adhesive 90%EP/10%Cu (X 10).

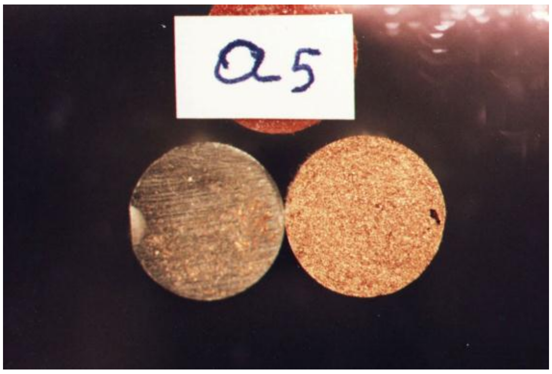


Fig. (12) The composite adhesive 90%EP /10%Cu (X 10).

Fig. (13) The composite adhesive 50%EP /50%Cu (X 10).

The same mechanical behavior appears with EP/Fe composite specimens, but the maximum value was at 40% weight percentage of the filler.

This results because of the precipitation of the (Fe) particle at other filler weight percentage which make two layers.The the first one of epoxy, the other of composite precipitation on the lower layers. This interne makes the decohesion appear at the epoxy layer, as shown in Fig. (14), but at (40% wt) which have more distribution of particles, it can be seen that the crack propagation through the adhesive layer as shown in Fig. (15).

These results agree with bending test, which show that at 60%EP /40%Fe, the material becomes more brittle, which lead to make the metal/composite adhesion greater than the cohesive.

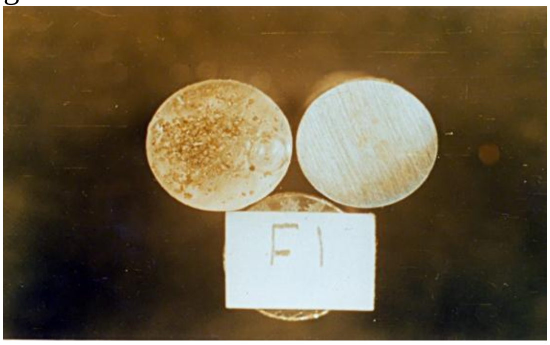


Fig (14) composite adhesive specimen for 90%EP /10%Fe (X10)

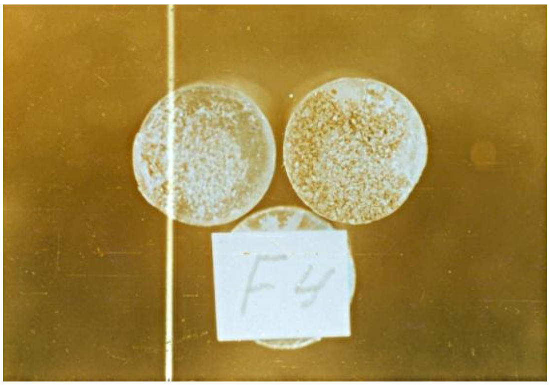


Fig. (15) The composite adhesive specimen for 60%EP/40%Fe the fracture growth from the interface through the adhesive (X 10).

From the obtained results, it can be said that the crack propagation flow two mechanisms, the first one was the crack growth at the metal/adhesive interface surface (upper and lower surface). The second mechanism was the crack initiation at the composite and propagate through it as shown in Fig.s (16), (17), and (18).

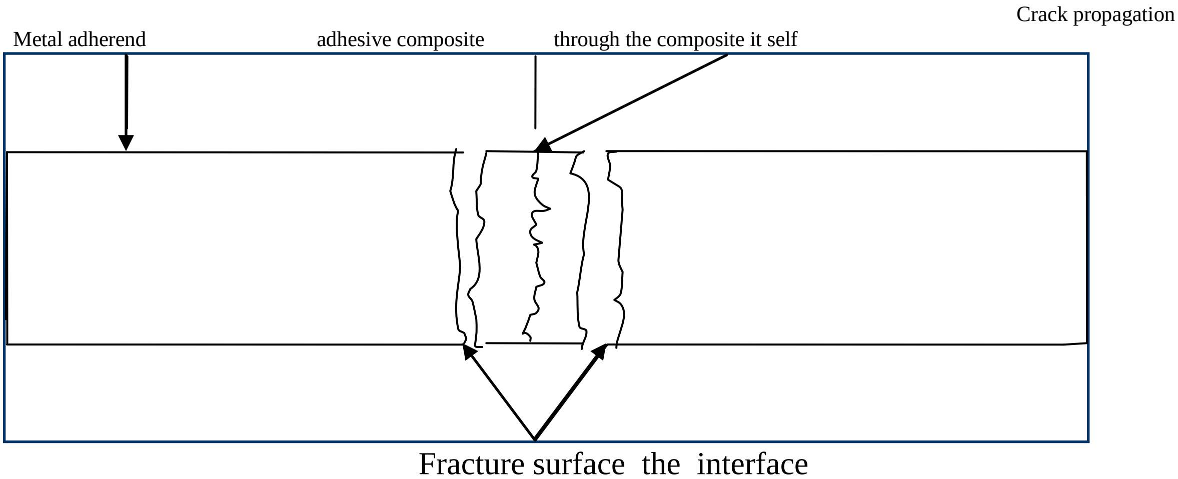


Fig. (16) The expected mechanism for the crack propagation in adhesive tensile specimen tests.

Fig. (17) The propagation of crack through the interface (X 10).

Fig .(18) The crack propagation through the composite itself (X 10).

For the specimens consisting of adhesive filled metal particles with EP composites ,the obtained results show that maximum value of strength was at intended weight percentage (i.e Al particles and Cu particles filled EP composite have maximum strength at 10% concentration ,where the Fe particle filled EP composite adhesive have the maximum strength at 40%.

The best obtained results were for metals particles filled EP composite was for Cu particle filler, which show good adhesion between the adhesive and the adherend at 10%.

General conclusions

EP/ metal composite show increase in adhesion force at certain filler weight percentage, where maximum stress was for EP/ Cu composite (9Mpa).

References

1. . P. Groover "Fundamentals of Modern Manufacturing" Prentice-Hall, Upper Saddle River, New Jersey 1996.
2. .V. Cagle "Adhesive Bonding" Mc Graw-Hill New York 1968.
3. M A. Carre and J. Schulitz "J. Adhesion" vol 15, (1983) pp151-163.
4. M. A.Meyers and K. Kumar "Mechanical Behavior of Material" prentice-Hall, Inc., 1999.
5. R. Klienholz and G. Molinier. "Vetrotex Fiberworld", No. 22, March 1986, pp: 11-20.
6. E. J. Ripling. S. Mostovoy and H. T. Corten" J. Adhesion" vol. 3. (1971), pp; 107.
7. F. S. Baker "J. Ahesion" Vol. 10, 1979. PP; 107-122.
8. A.C.Moloney, H.H. Kausch, and H.R.Stieger "J.Appl, Phys" Vol. 18, no.1, 1983 pp: 208-216.

9. Duncan, E. Arranz, and L. Crocker "Test for Strength of Adhesion", Crown Copy right UK 2004.
10. P.Bell and W.T.Mc Carvill "J. Appl. Poly. Sci."vol.18 (1974) pp; 2243-2247.
11. G. C. Papanicoloau and R. Mercogliano. "Plastics and Rubber Processing and Application" vol. 6 (1981), pp; 229-234.
12. Y. Agari, A. Ueda, and M. Tanaka "J. App. Poly. Sci." Vol. 40, 1990, PP; 929-941.
13. W. Wu and Z. Xu. "Acta polymerica Sinica". Vol. 1 (2001) pp; 99-104.
14. W. Gao, Q. Wang and S.Yan Yang "Acta polymerica Sinica." Vol. 1 2001 pp; 1-4.
15. R.H.Bott, L.T.Taylor,and T.C.Ward "J. of App. Poly. Sci." vol.36, issue 6 (2003) pp:- 1295-1304.
16. L.M. Grath, J.L.Lenhat, and R.Parnas "Composites"vol.1 November 2005.