The Effect of Particles as Additives on Water Absorption for Epoxy Resin

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ABSTRACT

Diffusivity behavior and diffusion coefficient was investigated in this paper for epoxy composite reinforced with metal(Al ,Cu , Fe) and metal oxide (Al2O3,Fe2O3,and CuO2 (with different weight percentage (5,10,15,20,and25%). The samples were immersed in water equal time for two months at constant temperature (room temperature 18C). The result show that the relative mass gain increased with increasing the immersion time till they tend saturation moisture mass (M∞) after that the relative mass gain decreased. Also the results show that the immersed samples in water filled with metals has maximum diffusion coefficient (4.73E-07cm2/sec) for EP/Al while the samples filled with metal oxides has maximum diffusion coefficient (3.41E-07cm2/sec) for EP/Fe2O.

Keywords: Particulate composite, water absorption

1.Intoduction:

EPOXY resin composites are widely used for the insulation of power transformers, switchgear, rotating machines, etc. The sensitivity of epoxy composites to humidity is a serious matter of concern because absorption of water may cause significant and possibly irreversible changes to the material [1]. Epoxy resins can absorb up to a few weight percent of water in a humid environment, leading to an overall degradation of the dielectric properties. Such a situation might be further exacerbated by inorganic fillers, which are often used to improve their mechanical and thermal properties and to reduce their cost[2].

Polymeric matrix composites differ from other materials in the sense that low- molecular weight substances such as watermay easily migrate even at room temperature generating variation of the material's structure, morphology, and composition [3]. In many cases, it could lead to an irreversible degradation of the material in the so- called humid aging that includes both chemical aging and physical aging. The epoxy matrix show moisture sensitivity due to Interactions between some polar groups of the macromolecule and the water molecules, which leads to a reduction of both glass transition (Tg) and mechanical properties. This sensitivity increases with the increasing degree of cross-linking and also with the polarity concentration of the molecular groups [4]. The physical phenomena that simultaneously occur are dissolution, diffusion, swelling, and relaxation, together with deformation and stress build up in the matrix

The relative mass gain can be obtained by:

M%= mass of wet sample – mass of dry sample x 100/Mass of dry sample   ….(1)

And the diffusion coefficient can be obtained by:

$$D = \pi \, (kb/4M\infty)^2 \ldots \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots. \ (2)$$

where:

D: diffusion coefficient (mm^2/day) , b: thickness of the sample(mm)., M∞: saturation moisture mass (gm).and K: the slop of the curve between mass gain and immersion time [5].

his diffusivity D, defined as the amount of liquid passing per second through a unit area under the influence of a unit gradient of concentration is still a function of the temperature T given by an Arrhenius expression:

$$D = Do \ exp \ (-H_D/RT) \qquad\qquad\qquad \ldots\ldots\ldots(3)$$

where: H_D: activation energy for diffusion. R: gas constant.
The diffusion time (t_D) defined as the duration of the transient and can be calculated as:[6]

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$t_D = \pi b^2/16\ D$ ……..(4)

Moisture diffusion in polymeric composites has shown to be governed by three different mechanisms [7, 8]. The first involves of diffusion of water molecules inside the micro gaps between polymer chains. The second involves capillary transport into the gaps and flaws at the interfaces between fiber and the matrix. This is a result of poor wetting and impregnation during the initial manufacturing stage. The third involves transport of micro cracks in the matrix arising from the swelling of fibers (particularly in the case of natural fiber composites). Generally, based on these mechanisms, diffusion behavior of polymeric composites can further be classified according to the relative mobility of the penetrate and of the polymer segments, which is related to either Fickian, non-Fickian or anomalous[9], A. Abd Alameer study electrical conductivity behavior of Cu-powder reinforced epoxy composite material in different solutions (distilled water, tap water &3.5%NaCl) with different a weight fraction [10], *W. S. Chow** study Water absorption of epoxy/glass fiber/organo-montmorillonite nanocomposites[1]

Although the diffusion coefficient is not a direct measure of mechanical deterioration, it is a relative measure of the rate at which the composite absorb water and the mechanical properties deteriorate.

2. Materials and testing procedure

The material used to prepare the test samples were epoxy resin (EPI0 Conbextra) with the hardener aliphatic amine (Hy 956) in ratio 3:1 for curing. Three type of metal (aluminum, iron, copper) and metal oxide (of purity 99.5%) used as fillers in this work. A weight amount of epoxy was mixed in (5%, 10%, 15%,20% and 25%) percentage. Table (1) summarizes the materials and some of their properties.

A hand lay up method was used to lubricate all the specimens in this work and many testes showed that the ratio of hardener to epoxy, which will be used in this study, is approximately [3:1], to prepare the composite sheets, which are cut later to get the samples(1*1)cm according to ASTM_D570 .The additives powder was added to the epoxy ,stirred for 5 minutes to be well mixed and for more diffusion the mixture was put in the oven at 50^0 C for 15 minute. The mixture was then left for 24 hours for curing.

All prepared sheets were checked by optical microscopic to see the good dispersion of the particles distribution.

The dray samples were weight and then immersed in water for (8 weeks), at the first week the samples weighted daily then every two weeks   with sensitive (starorius) by W-Germany, It was proven to be a sufficient time for reaching the steady state.

Table 1: some of properties of the used material

Material	Sample	Density (g/cm^3)	Particle size (µm)
Epoxy	EP	1.2	
Aluminum	Al	2.7	**10.2**
Iron	Fe	7.3	**15.01**
Copper	Cu	8.9	**12.42**
ALUMINUM OXIDE	**AL$_2$O$_3$**	**3.96**	**9.76**
CAPPER OXIDE	**CU$_2$O**	**6**	**15.2**

Water diffusion in epoxy materials was measured at room temperature (298K). Water sorption in the resins as a function of the square root of processing time (hours) was monitored by this gravimetric method. The specimens were periodically removed from the water, wiped down, and quickly weighed on the microbalance until the percent weight did not change.

3. Results and discussion

To determine the diffusion coefficient, the specimens were soaked in water at room temperature and the weight changes were monitored as a function of time. The percentage weight change M was calculated by equation (1).

The plotted curves of figs . (1) , (2) ,and (3) show the variation of water gain M against the square root of the immersion time $\sqrt{t}$ for neat epoxy resin and its different metal, and metal oxide composites. It is seen that the water uptake increased linearly with increasing of the square root of immersion time, then gradually slow until an equilibrium plateau is reached. This is similar to the prediction of Fickian behavior. The plateau is defined as the apparent maximum water content M_∞[1].

Water diffusion of neat epoxy and its composites were calculated from the relationship between weight gain M and diffusivity during initial water uptake for Fickian diffusion which is given by equation (2) .

Diffusion coefficients values were evaluated and listed in Table (1) the results reveal that the diffusion coefficients of resin composites are less than that of net epoxy.

Table (2) also shows that the diffusion coefficient of specimens of epoxy / metal oxide composites are less than epoxy resin and epoxy / metal composites , because of reaction between epoxy and (H2O) through the interface making a separation between the epoxy and particles [11].

Water diffusion gives an indication of the affinity of the polymer to water and the degradation of the resin caused by water. The diffusion rate depends on the surface condition of the resin, i.e., airtight or ventilated, and the dispersion of micro voids in the resin. If the initial diffusion rate is quick, this suggests that the surface of the resin is porous, and the micro voids in the resin are connected [9].
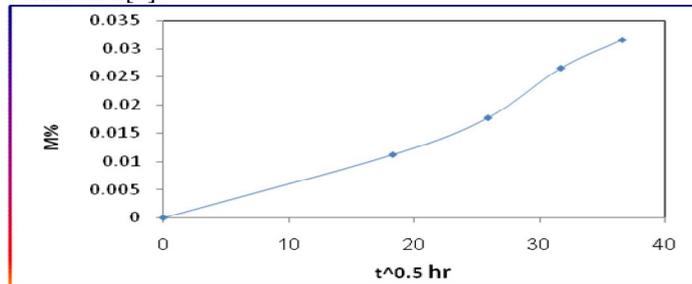

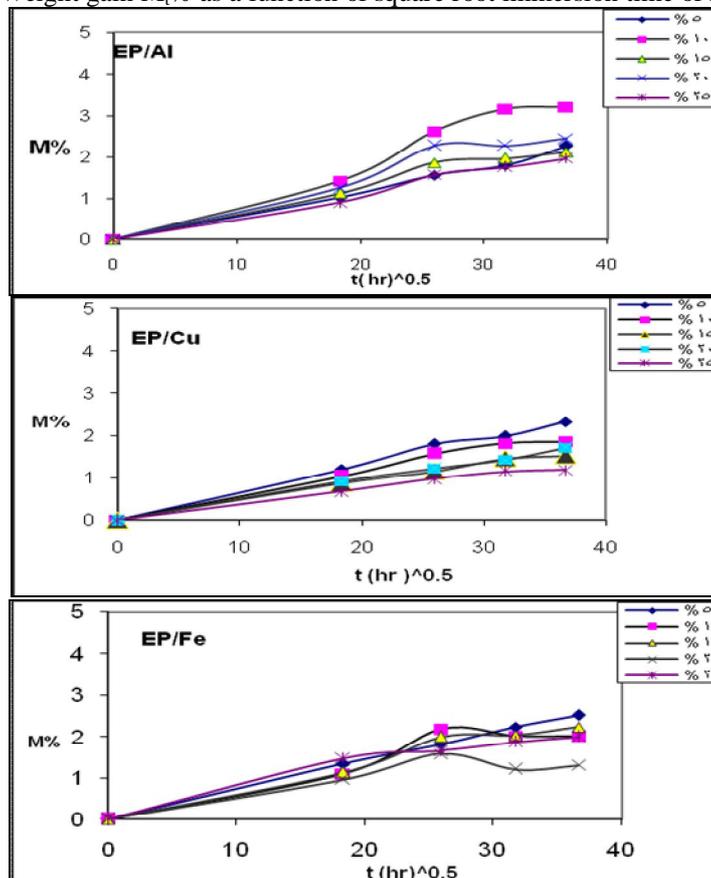**Figure 1:** Weight gain M_t% as a function of square root immersion time of Epoxy resin.

Figure 2: Weight gain M_t% as a function of square root immersion time
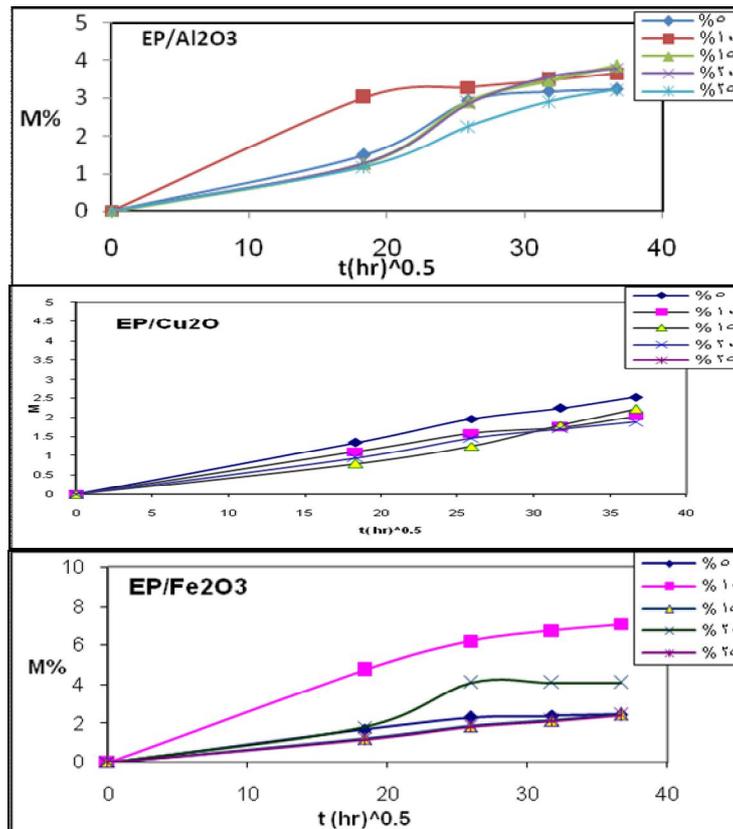
of Epoxy and EP/Metal composites

Figure 3: Weight gain $M_t\%$ as a function of square root v.s immersion time of EP/Metal Oxide composites

Table 1: Diffusion coefficient of neat epoxy resin and metal/epoxy composite

Composites	Diffusion coefficient (cm^2/sec)		
Wt%	Al	Cu	Fe
5%	4.73E-07	2.39E-07	4.73E-07
10%	1.78E-07	2.26E-07	1.78E-07
15%	2.19E-07	2.08E-07	2.19E-07
20%	2.22E-07	2.53E-07	2.22E-07
25%	2.62E-07	1.75E-07	2.62E-07
Epoxy resin	5.81E-07		

Table 2: Diffusion coefficient of neat epoxy resin and metal oxide/epoxy composite

Wt%	Diffusion coefficient (cm^2/sec)		
Composite	Al_2O_3	Fe_2O_3	Cu_2O
5%	3.08E-07	3.57E-07	1.90E-07
10%	2.73E-07	1.83E-07	1.85E-07
15%	1.81E-07	1.48E-07	2.13E-07
20%	1.38E-07	1.98E-07	1.19E-07
25%	2.19E-07	3.41E-07	1.57E-07
Epoxy resin	5.81E-07		

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The water absorption behavior of polymer-filled particles composites at a humid environmental condition is determined by many factors, such as processing techniques, polymer matrix and filler characteristics, composition of the composites, and so on [12].There are two situations for water sorption in composites filled with inorganic fillers:

• One is when there is a good interfacial adhesion established between the matrix and the particles. This may result in a higher density composite and less free volume around the particles. If there is a higher concentration of inorganic particles, the denser interface zones between particles and matrix means that less water can be contained in this region [12].

• Conversely, some bonds between epoxies and some filler (such as ceramic and aluminum) are thermodynamically unstable or not strong enough in the presence of water molecules. The thermodynamic work in the interfaces between fillers and matrix could become negative in the presence of water from the positive value when dried. For example, the thermodynamic work for the epoxy-ferric oxide interface changes from +291 to -255 mJ/m2 when there is an absorbed layer of water at the interface [12,13]. This change from a positive to negative work of adhesion provides a driving force for the displacement of adhesive on the interfaces between particles and epoxy matrix by water [2]. It is therefore to be expected that the interfaces between the inorganic fillers and matrix might be de bonded in the presence of water. At The higher concentration of inorganic particles, the more broken bonds, and therefore the water can be contained in this interfacial region. The generally accepted mechanism for solution sorption in polymers is an activated sorption-diffusion process.The molecules first dissolve into the polymer surface and then diffuse through the bulk of the polymer by a series of activated steps. The amount of absorbed water depends on the temperature, the structure and the morphology of the polymer [13]. It is clear from the measurements that the epoxy in the micro-filled system absorbs about the same amount of water as the pure epoxy for a given humidity and that, for the micro-filled system, virtually all the water is absorbed by the epoxy, because water has polar dipole when these molecule were diffused into Polymer, the polar dipole will act as electrostatic force between polymer back bone chains, so that the polymers become more strength [15, 17, 18]

Diffusion is the net movement of matter down a concentration gradient due to the random movement of individual particles. The process is dependent on the imposed boundary condition [11]. When a dry specimen is put in a humid environment, then, after some time, a steady state is obtained in which water diffusing through the plane per unit time is zero. Within the plane, the concentration gradient is not zero. However, once a steady state is established, the concentration at any location does not change with time. This defines a steady state, and Fick's First Law is most useful in dealing with this situation [14]

4. Conclusions

a) The results reveal that the diffusion coefficients of resin composites are less than that of net epoxy

b) Also the results shows that the diffusion coefficient of specimens of epoxy / metal oxide composites are lessthan epoxy resin and epoxy / metal composites

c) The results show that the immersed samples in water filled with metals has maximum diffusion coefficient (4.73E-07cm^2/sec) for EP/Al

d) the samples filled with metal oxides has maximum diffusion coefficient (3.41E-07cm^2/sec) for EP/Fe$_2$O$_3$

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