

# HYGROTHERMAL AGEING EFFECT ON GLASS FIBER COMPOSITES: A REVIEW

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## ABSTRACT

*A Composite material is a class of advanced material in which two materials work together to give the composite unique properties. The hygrothermal ageing effects (combination of high temperatures and moisture ingress) on glass epoxy composites studied in the literature which results in the degradation in mechanical properties of glass fiber composites. It was observing that in the GF composite degradation is not only a function of water uptake but is also time-dependent. In GF Composites shear strength reduction was observed up to 36% whereas in resin degradation approximately 17%. Fatigue life was obtaining up to three order shorter magnitude.*

**Keywords:** *Hygrothermal ageing, Glass epoxy composite, Mechanical properties*

## I. INTRODUCTION

A composite material is made by combining two or more chemically different constituents combined macroscopically to yield useful materials often ones that have very different properties. The two materials work together to give the composite unique properties. However, within the composite you can easily tell the different materials apart as they do not dissolve or blend into each other. And whose mechanical performance and properties are designed to be superior to those of the constituent materials acting independently.

Glass as a structural material was introduced early in the seventeenth century and became widely used during the twentieth century. Glass fibrous usage for reinforcement is pioneer in replacement of metals and used for both commercial and military uses with the advent of formulation control and molten material which is die or bushing pulled into continuous filaments. These events lead to a wide range of aerospace and commercial high performance structural applications still in use today. Over 95% of the fibres used in reinforced plastics are glass fibres, as they are inexpensive, easy to manufacture and possess high strength and stiffness with respect to the plastics with which they are reinforced. Their low density, resistance to chemicals, insulation capacity are other bonus characteristics, although the one major disadvantage in glass is that it is prone to break when subjected to high tensile stress for a long time.

The hygrothermal ageing is a combination of high temperatures and moisture ingress, on the mechanical behavior of composite material, which are require to with stand extreme service environments. Hygrothermal

stresses in the material as a function of location and time and changes in material properties as a function of time (e.g., strength, stiffness, glass transition temperature, thermal conductivity, and thermal expansion).

## **II. HYGROTHERMAL AGEING**

Hygrothermal ageing is an accelerated ageing by moisture absorption and temperature change on the mechanical behaviour of composite material, which are require to with stand extreme service environments. Hygrothermal ageing is a complex combination of physical and chemical phenomena that operate at different time and spatial scales. The incurred degradation strongly depends on the material components involved and on the exposure environment. The ageing process is drive by water ingresson, it is also important to understand the process of water diffusion across scales. For fiber reinforced polymers, the material micro structure brings complexity to the diffusion phenomenon since water molecules have to go around the fibers for diffusion in transverse direction, giving rise to orthotropic diffusion behaviour. Chemical interaction between the epoxy and fiber sizing creates an inter-phase region around the fibers. Composite structures operate in a variety of thermal and moisture environments that may have a pronounced impact on their performance.

## **III. RESEARCH WORK CARRIED OUT ON HYGROTHERMAL AGEING OF GLASS EPOXY COMPOSITES**

Hydrothermal ageing behaviour of glass/epoxy composite (with respect to neat epoxy) was studied by Rocha et al. (2017). Both type of specimens were conditioned in de-mineralized water at 50° C for 4800 h and tested quasi-statically and in fatigue. After immersion, specimens were dried and tested again. The damage due to moisture ingresson and the subsequent changes in failure behaviour were further investigated through thermal analysis (DSC, DMA) and optical microscopy. Significant degradation was found in composite specimens; with up to 36% lower static shear strength and three orders of magnitude shorter fatigue life due to high differential swelling stresses and damage to the fibre/matrix interface in composites. A lower degree of degradation was observed for neat epoxy specimens, with up to 17% lower tensile and bending module and strength. After drying, recovery of shear stiffness and strength of composites was incomplete, while stiffness and bending strength were completely recovered for neat resin.

Figure 1, shows stress-displacement curves for specimens test after each immersion time. The stress-displacement curves suggests that the failure behaviour becomes more ductile as immersion time increases, with gradual load drops instead of the more sudden drops observed in reference specimens.

Regarding the evolution of strength with immersion time, it can be seen that the degradation keeps increasing well beyond the apparent saturation phase between 1000 h and 2000 h. This suggests that degradation is not only a function of water uptake but is also time-dependent. Two distinct mechanisms may be suggested as being responsible for such behaviour. Firstly, hydrolytic chemical reactions and subsequent leaching of material at the fibre/matrix interfaces lead to a weakening of the interfacial bond and creation of additional spaces for water uptake. Secondly, differential swelling stresses at the weakened interfaces promote crack initiation and

propagation, further increasing the available space for water uptake. It is important to note that such mechanisms may act in isolation or in combination, synergistically reinforcing one another.

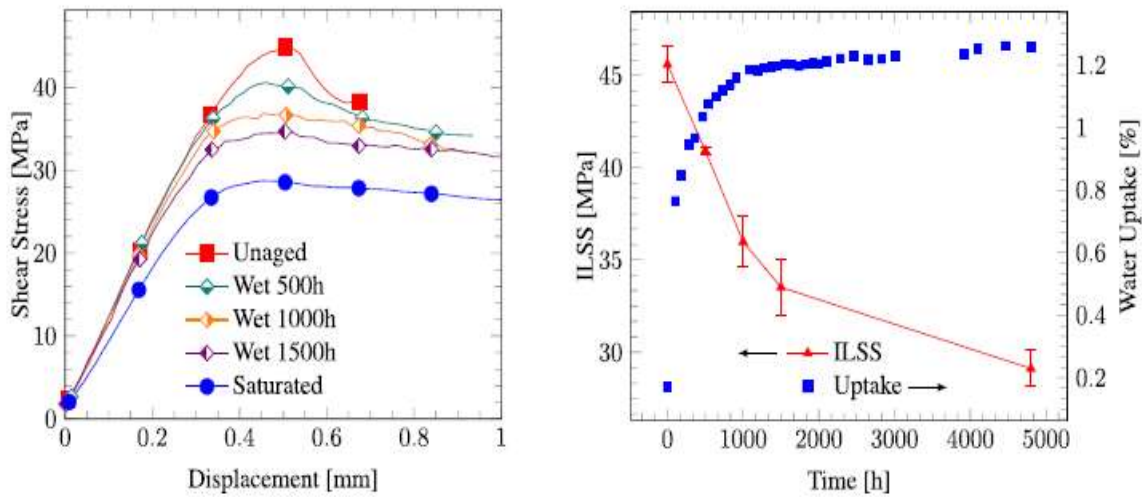


Figure 1: Static Inter laminar shear strength results for specimens immersed for multiple durations (4-ply composites) (Rocha et. al, 2017)

In Figure 2 shows that for neat epoxy, final moisture uptake is approximately 3.94%. Whereas in composite specimen final uptake of approximately 1.25%. And composite shows an apparent saturation at 1.19% between 1000h and 2000h.

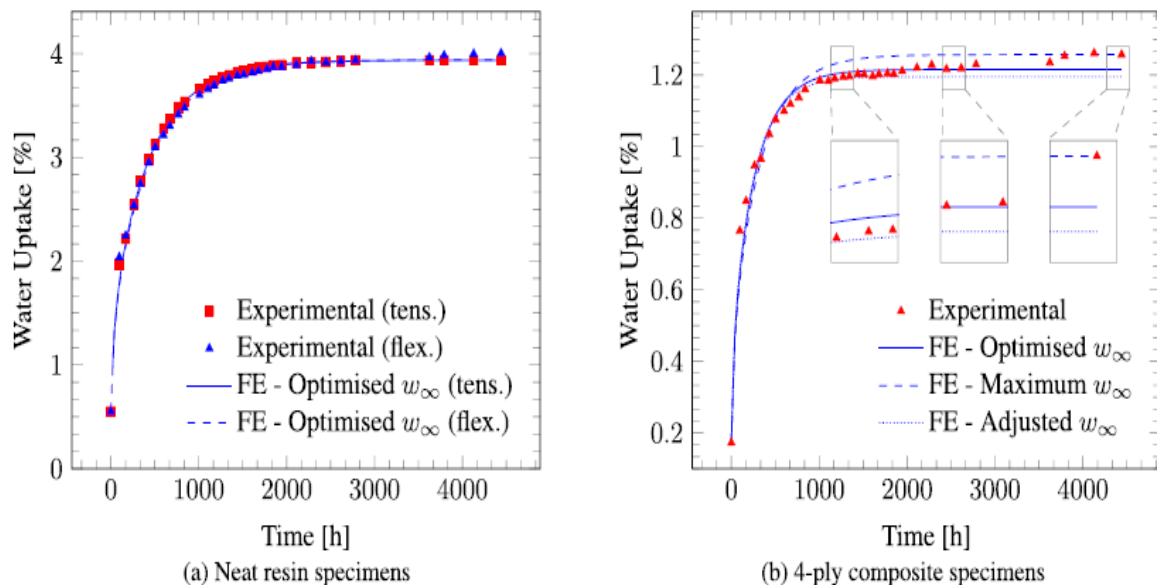


Figure 2: Water uptake values. (Rocha et. al, 2017)

Hygrothermal aging effects on flexural behaviour of pultruded glass fibre reinforced polymer laminates in bridge applications was studied by Xin et al. (2016). Specimens immersed in fresh water and artificial seawater

environments at 40°, 60° and 80° temperatures. The degradation in both average flexural strength and modulus increases at higher temperatures. And the degradation in transverse flexural strength and modulus is relatively higher as compared to those observed for longitudinal flexural strength and modulus.

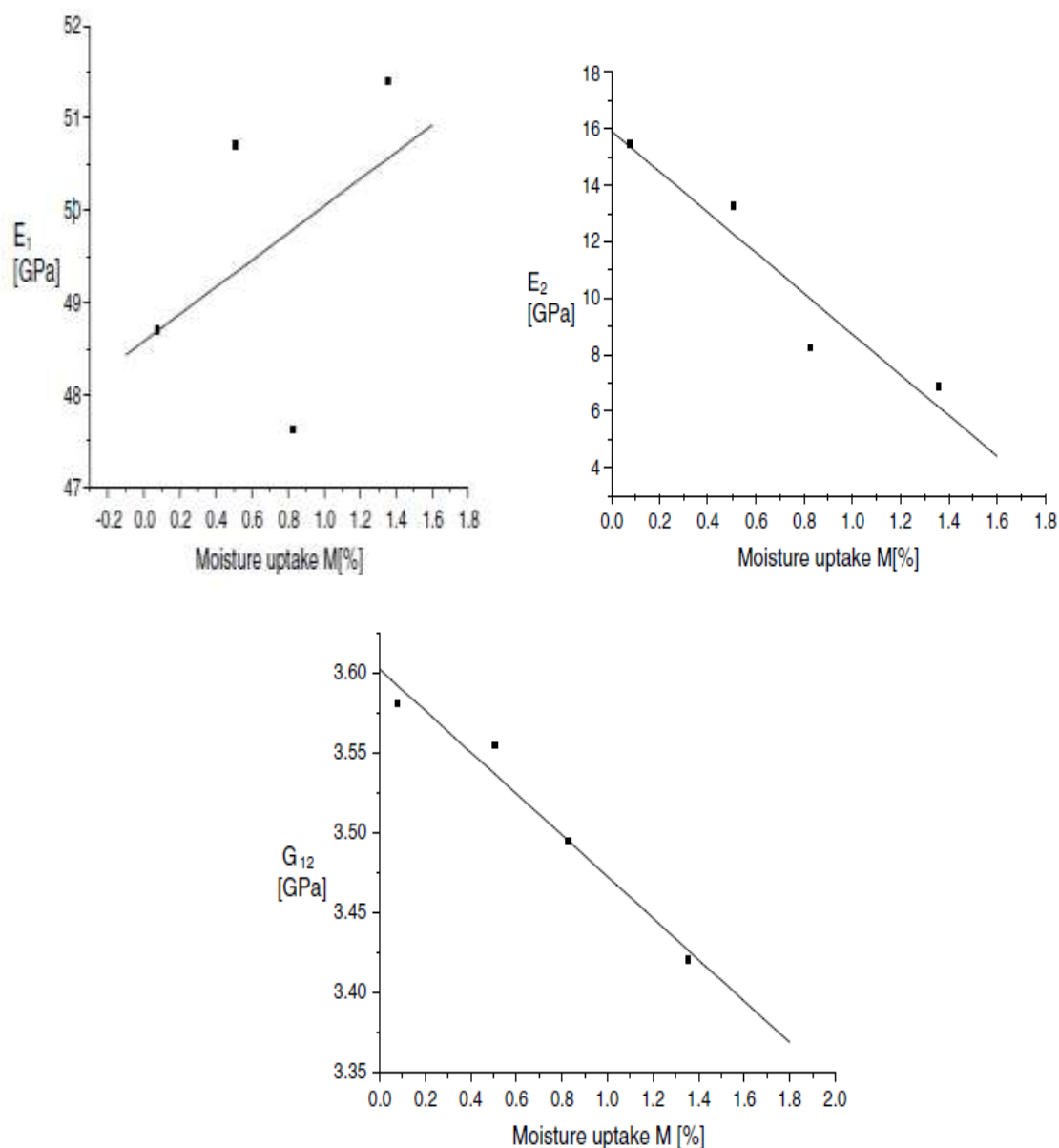
In hygrothermal condition mechanical performances of pCBT (poly butylenes Terephthalate), glass fiber reinforced pCBT (GF/pCBT), and nano-silica modified glass fiber/pCBT composites (nano-GF/pCBT) were studied by Yang et al. (2015). DSC, DMA and fiber pull-out tests were adopted to obtain the experimental results. Relative humidity was set at 60% and 90%, while the temperature was respectively set at 50 °C, 60 °C, 70 °C, and 90 °C. It was observing that all the SDR (Strength degeneration ratio) with time shows the linear relationship in hygrothermal environment, while SDR-temperature curves shows a bilinear relationship due to the effect of glass transition temperature ( $T_g$ ) of the matrix. Fibers modified by coating of nano-silica on the surface could decrease SDR of the composites. This was due to the fillers on the fiber surface could resist the movement of pCBT molecular chain and diffusion of water molecules in aging conditions.

The effects on the microstructure and the impact behavior of thermo set polyurethane composite material, exposed to an accelerated aging environment with hygrothermal and ultraviolet (UV) chamber was studied by Nicholas et. al (2015). The low velocity impact test was carried out at 250, 500, 750 and 1000 h for evaluation of impact properties of the material. By Impact energy test observed that at  $700 \pm 170$  N for all aged samples and found that  $F_{max}$ ,  $E_{max}$ , total absorbed energy and deflection is less than  $\pm 10\%$  compared to the virgin sample. The impact properties did not measurably vary as the exposure time increased.

Hygrothermal ageing effect on mechanical properties of two composite materials (carbon fiber / epoxy and glass fiber E/ vinyl ester) was studied by Larbi et. al (2015). Mechanical bending properties were obtaining by three point bending tests in the sea water and distilled water at a temperature of 40°C. The absorption in distilled water was greater than in seawater due to the harmfulness of the high pH of seawater and the presence of metal salts that cause degradation of the matrix, hence the continuous absorption.

A study of the effects of environmental conditions and fiber orientation angle on saturation values and ageing degradation of E-glass/epoxy composites exposed to hygrothermal conditions was studied by Boukhoulda et. al (2015)

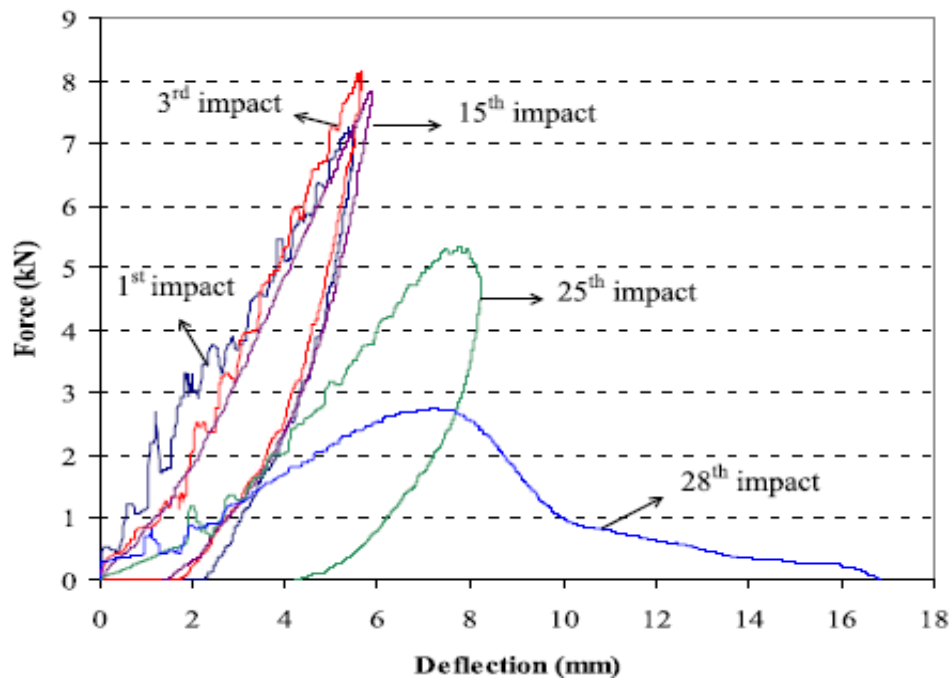
The results of the variation of Young's modulus (longitudinal and transverse modulus) and the in plane shear modulus versus weight water absorbed are shown in Figure 3. A considerable rigidity loss for  $E_2$  and  $G_{12}$  is noticed. The decreasing trend of the relative rigidity of  $E_2$  is more pronounced than that of  $G_{12}$ . The Young's modulus  $E_1$  of the unidirectional laminate is not apparently affected by the absorption of water. The same observation is made for Poisson's ratio  $m_{12}$ .



**Figure 3: Variation of Youngs modulus  $E_1$ ,  $E_2$  and  $G_{12}$  according to weight water absorbed. (Boukhoulda et. al, 2015)**

An experimental investigation on the repeated impact response of glass/epoxy composites subjected to thermal ageing was studied by Atas and Dogan (2015). Impact energies were chosen as 20 J, 40 J, 60 J, 80 J and 100 J and for single impact tests 20 J was chosen. The samples were exposed to ageing durations of 100, 400, 700, 1000 and 1300 h by using a climatic test cabin. Damage area increases with ageing duration, especially for smaller impact energies. The total energy absorbed until perforation of un-aged samples is over three times of perforation energy of samples exposed to temperature and humidity during 1300 h.

The contact force deflection curves of aged samples for ageing durations of 100 h, 700 h and 1300 h, respectively. In Figure 4, it shows that the successive impacts reduced the bending stiffness, slope of the curve, and contact force.



**Figure 4: Force deflection curves of an aged sample (700 h) for various impact numbers. (Atas and Dogan, 2015)**

Hygrothermal aging effects on fatigue behavior of glass fiber/ polydicyclopentadiene (pDCPD) composites were studied by Hu et. al (2014) by using acoustic emission monitoring. Samples were immersed in de-ionized (DI) water and salt water. The pDCPD composite was showed less water uptake than the glass/epoxy composites because of the intrinsic hydrophobicity of pDCPD matrix. Thus, pDCPD composites showed superior mechanical performance under dynamic and quasi-static loading in both 0° and 90° directions.

The weight change of 4-ply pDCPD (polydicyclopentadiene) and epoxy laminates after aging shows in Figure 5. The pDCPD composites showed less water uptake than epoxy composites after aging. The pDCPD composites showed a roughly linear increase, while epoxy composites showed a Fickian approach to saturation with accelerated kinetics in salt water.

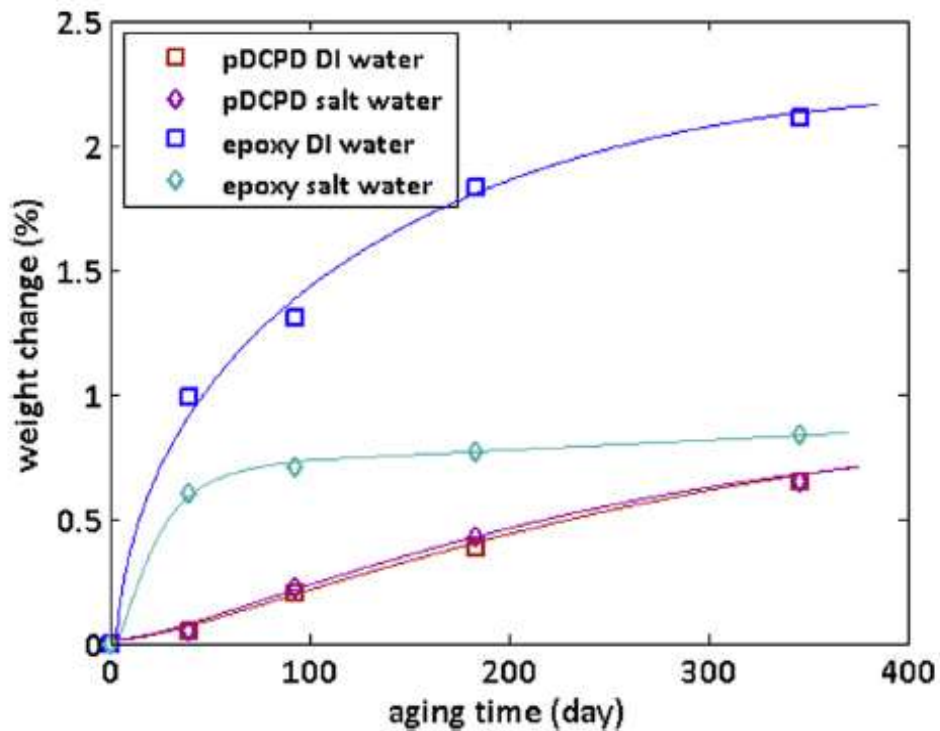


Figure 5: Weight changes after aging.(Hu et. al, 2014)

The effect of hygrothermal ageing on the flexural properties of glass/epoxy, glass/linseed oil and glass/castor oil composites was studied by Malmstein et. al (2013). Glass/castor oil and glass/linseed oil were absorbed more water and had not reached a moisture equilibrium condition after 46 weeks. The glass/epoxy failure became less brittle after ageing resulting in less cracking inside the material.

Figure 6 shows that after only 2 weeks of immersion glass/linseed oil specimens had reached the maximum degradation in the flexural strength and modulus indicating that water combined with elevated temperature had an extensive and rapid damaging effect on glass/linseed oil composite. While with glass/epoxy the properties kept decreasing with uniform moisture content, the moisture content of glass/linseed oil kept increasing while the properties remained at the same (albeit low) level. This suggests that the chemical degradation of glass/linseed oil composites was limited and the effects of water involved extensive interface and matrix degradation. However, as glass/linseed oil reaches the maximum degradation rapidly, the performance of the composite is highly predictable. Unlike glass/epoxy, the flexural modulus of glass/linseed oil was significantly affected by ageing. After 26 weeks, the modulus had reduced by 59.8 %. Glass/castor oil specimens showed lowest reduction (21%) in flexural strength after 22 weeks of ageing resulting in a comparable strength to glass/epoxy.

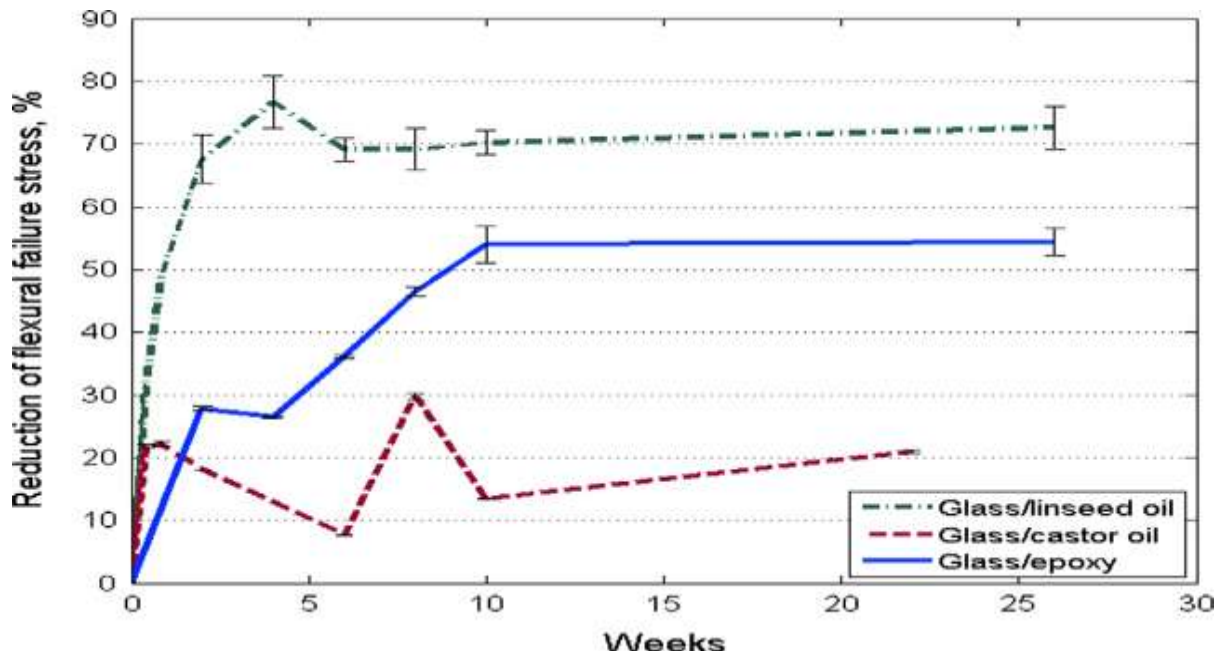


Figure 6: Reduction in flexural strength of glass/epoxy, glass/linseed oil and glass/castor oil composite over a 20 + week ageing period. (Malmstein et. al, 2013)

Hygrothermal aging and moisture absorption in glass-fiber-reinforced polymer laminates and a structural adhesive of FRP composite bridge were studied by Jiang et. al (2013). The moisture diffusion in GFRP laminates and adhesive was characterised in four environmental aging conditions (20 °C–50% RH (relative humidity), 20°C water, 40°C 96% RH and 40 °C water). In FRP laminates specimens aged in the 40°C water condition absorb more than six times of moisture content than that of the 20°C 50% aging condition due to fickian diffusion was humidity-dominant.

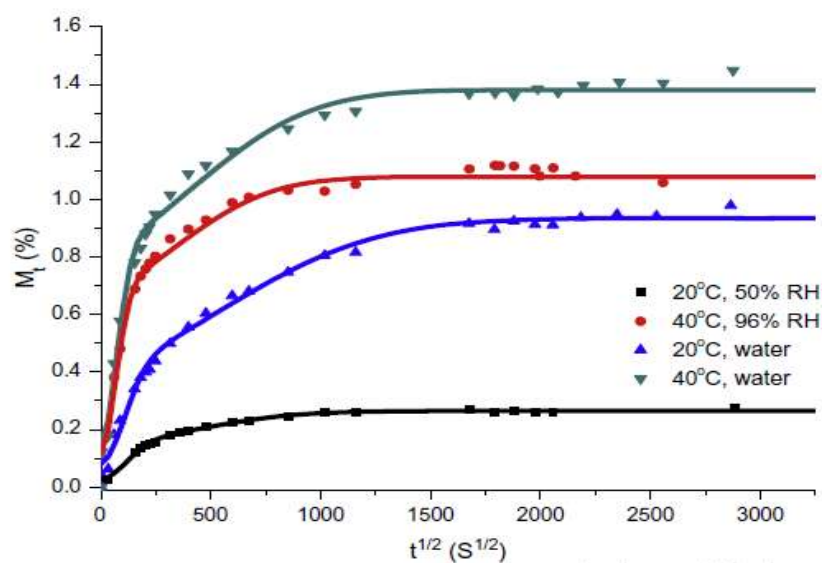


Figure 7 Moisture uptake curves of FRP composite specimens aged in four aging conditions.



Dynamic mechanical thermal analysis of hygrothermal aged pultruded glass fiber reinforced polymer rebar was studied by Swapan et. al (2012). GFRP rebars were accelerated aged in an alkaline aqueous environment at 60°C for 1, 2, 3, 4 and 6 months. There was no evidence of degradation of the GFRP rebars after 6 months exposure in the alkaline environment at 60° as analyzed by DMTA.

In hygrothermal environment aging-impact coupling upon the polyester matrix E-glass fibers composite material was studied by Boukhoulda et. al (2012). Composite plates were aged in an artificial hygrothermal environment using a climatic chamber at 50°C temperature and 95% of relative humidity. And low energy impacts test were also carried out on both aged and non-aged composite plates. Decrease in the impact force, increase in plate deflection and the contact duration, increased flexibility of the material compared to unaged and it showed that non-Fickian character of the moisture absorption kinetics.

The effect of voids formed in both the glass/epoxy and GLARE laminates on their long-term hygrothermal behaviors was studied by Park et. al (2009). Specimens immersed in distilled water and thermal cyclic fatigue up to 1500 cycles for Ilss tests were carried out. Specimens were obtained void contents ranging from 0.5% to 2.0% for glass/epoxy and 0.7% to 1.3% for GLARE. The ILSS for glass/epoxy was reduced to 8–13% and 9–18% for GLARE after 1500 thermal cycles.

The effect of the nature of glass fiber surface in the water absorption of glass fibers/epoxy composites use of fluorescence was studied by Olmos et. al (2006). Gravimetry and FTNIR measurements were used to complementary study of water absorption process. The glass fiber surface yields a change in the structure of the epoxy matrix in comparison with that of the polymer without reinforcement. The increase of the inter-phase (tens of micrometers) crosslink density the lower the functionality of the silane was a higher cross linking density should yield less free volume to be occupied by water molecules during the water absorption process.

Hygrothermal ageing on glass fiber reinforced polyethylene tere phthalate (PET) composites degradation was studied by Foulc et. al (2005). Composites degradation result was obtained by DSC and SAXS experiments. DMTA test of the plasticization of the PET matrix also carried out. After long ageing times, hydrolysis may affect even the crystalline zone of PET since the laminate thickness decreases. And interfacial de-cohesion occurs leading to the formation of cracks or voids that induce additional uptake of water and the final osmotic cracking responsible for the material fracture.

The hygrothermal aging on the thermo-mechanical properties, molar mass and microstructure of recycled poly(ethylene tere phthalate) (rPET) and its short glass fibers composite were studied by Pegoretti and Penati (2004). Hygrothermal aging at 70° in water and at 80% relative humidity. The elastic mechanical properties were determined at low strain levels. The occurrence of a chemi crystallization process that causes an increase of the cristallinity content and a consequent reduction of the mobility of the amorphous phase temperature of the loss factor peak was increasing.

The hygrothermal ageing on mechanical properties (fiber/matrix interfacial and/ or inter-physical chemistry) of glass fiber/epoxy composite experimentally were studied by B.C. Ray (2003). Hygrothermal conditioned laminated composites were treated at -6 °C temperature to freeze the absorbed moisture. It was also investigated the effect of variation of loading speeds on the degradation behaviour of polymer composites and found that it



was sensitive to loading speed. The strain-rate sensitivity was less pronounced at higher conditioning times. The degradative effect of further freezing treatment was more evident at lower loading speed. The more degradative effect because of frozen state of absorbed moisture was permanent.

Hygrothermal ageing on the fatigue behavior of a unidirectional glass/epoxy composite was studied by Vauthier et. al. (1997). Hygrothermal ageing was done at the most elevated temperature (70°C). At the most elevated temperature (70°C) significant losses in life times compared to unaged due to local interactions between composite and surrounding moisture.

#### IV. CONCLUSIONS

The hygrothermal ageing effects on glass epoxy composites studied in the literature which results in the degradation in mechanical properties of glass fiber composites. Based on the literature review following concluding remarks may be drawn:

- We are finding that in the GF composite degradation is not only a function of water uptake but is also time-dependent. We saw that water diffuses through the material and promotes resin plasticization it degrades interfacial strength.
- In GF Composites shear strength reduction was observed up to 36% whereas in resin degradation approximately 17%. After re-drying, an irreversible shear strength and shear stiffness in GF composite reduction found 17% whereas in resin fully recovered.
- For neat epoxy, final uptake is approximately 3.94%. Whereas in composite specimen final uptake is approximately 1.25%. Composite shows an apparent saturation at 1.19% between 1000h and 2000h.
- The absorption in distilled water was greater than in seawater due to the harmfulness of the high pH of seawater and the presence of metal salts that cause degradation of the matrix, hence the continuous absorption.
- The pDCPD composites showed less water uptake than the epoxy composites because of the intrinsic hydrophobicity of pDCPD matrix. The pDCPD also had greater interface bond strength to glass fibers than the epoxy and pDCPD composites exhibit exceptional resistance to hygrothermal environments and as a result, superior retention of static and dynamic mechanical properties.
- Impact energy difference was found to be about 10% for sample with ageing duration of 1300 h compared to un-aged one. The total energy absorbed until perforation of un-aged samples was over three times of perforation energy of samples exposed to temperature and humidity during 1300 h.
- For the E-glass/epoxy, the prediction of saturation exposed to the environment is estimated at around 1.95 years.
- The degradation in transverse flexural strength and modulus is relatively higher as compared to those observed for longitudinal flexural strength and modulus.
- The interfacial bonding strength of glass fiber with nano-fillers on the surface enhancement is effecting anti-aging property of composites.

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