Role of using some Hybrid Nano-Fillers on the Strength and Creep Resistance of Fiberglass/Epoxy Composit

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Abstract. The effect of adding hybrid nanomaterials (multi-walled carbon nanotubes (MWCNT) with calcium carbonate (CaCO\(_3\))) was studied and compared with the effect of adding only one of those nanomaterials and without any addition on creep behavior at 40 °C and constant applied load. Two different additives weight ratios were studied (1% and 2%) from the weight of epoxy. Three layers of Bi-directional (0-90) glass fibers used as reinforce material the epoxy matrix. vacuum bag technology and sound wave device were used for samples preparation according to the standards. The Samples were tested with crawl tester designed and manufactured according to engineering specifications and standards. The results showed that there was an improvement in the creep behavior and a high creep resistance of the samples with the hybrid nanocomposites compared to the neat samples.

Keywords: Creep Behavior; Hybrid Nano materials; multi-walled carbon tubes; calcium carbonate; Fiberglass; Epoxy

1. Introduction

Multi-domain composites have been of recent interest because of biocomposite materials with hierarchical levels of reinforcement. Man-made multiscale micro and nanocomposites offer a route by which multifunctionality, it is Take from nature as thermal stability, thermal and electrical conductivity, and low coefficient of thermal expansion, the additional level of hierarchy through transferred to the fiber-reinforced composites. The dominant properties of fibers of nanocomposites of hybrids may benefit greatly from the incorporation of nanomaterials composites. It is expected the resin-dominated properties to benefit. the utilization of nanotechnology has been developed in different domains and applications because of its high-performance [1]. The purpose of hybridization in composites is to extend the concept of tailoring properties to suit requirements and to offset the disadvantages of one component by the addition of another [2]. Mahrolz et al [3] reported a significant enhancement in mechanical performance including improvements in stiffness and tensile strength, delamination resistance, and safety factors of their epoxy-based multiscale composites. Yi-Luen Li,et [4], observed improvement in mechanical properties and creep behaviors by adding CNTs to epoxy/carbon fiber(CF) under various states of loading, temperature, fiber orientation, contents (CNT), and humidity, showing them as focal factors which have their great effect on creep behavior; this is because of the fact that creep strain and creep strain’s level of CF / epoxy matrix laminates increase following loading increment, angle degree θ between the fiber orientation and the applied stress direction, temperature and humidity; also, creep strain rate of CNT added composite plummets with the amounts of CNTs increment. CNTs can supply high mechanical
strengthening, and the addition of a suitable amount of CNTs also could diminish creep strain rate. Obuka Nnaemeka Sylvester P, et al [5], studied the new hybrid epoxy resin composite consists of [5, 10, 15, 20, 25 and 30] %wt of fills (alumina and calcium silicate) in nanoscale. Creep tests were conducted at temperatures of 50C, 70C, 90C, 110C, and 130C, at constant loading of 14 MPa. Tensile test’s results explain that such composite materials possess an excellent resistance compared to the resistance of neat epoxy or the baseline epoxy / alumina composite materials. New materials brought out a good rate of creep resistance particularly at temperature between 50C and 90C, whilst the 15 %wt. and 20 %wt. filler component showed such a good creep resistance when temperatures were rather higher. Hence, the hybrid composite with 15%wt of components was unique in that it could resist the temperature of 130C. Zhen Yao, et al [6], studied the creep behavior to Polyurethane (PU) nanocomposites included carbon nanotubes (CNT) were prepared during polymerization. The results explained that the existence of (CNT) leads to enhanced creep resistance to (PU) and that the resistance does not increase with increasing (CNT)because they depend on the dispersal of (CNTs). the nanocomposite included 1.0%wt CNTs shows the lowermost creep level. The as-obtained parameters moreover emphasize the handicapping influence on the creep by the presence of CNTs. Xiao Zhang, et al [7], studied the status of graphene to get better the mechanical properties of polymer composites. The literature has explained that graphene can significantly enhance the static mechanical properties, dynamic mechanical properties and creep properties of polymer composites. At the same time, the production process and the dispersion state of graphene have an important impact on the enhancement of mechanical properties.

2. Experimental Work

Materials and Preparation Of Samples

Epoxy resin produced by (SWISS CHEM) with a density of 1.05g / cm³ was used. The base of the liquid epoxy (Euxit 50KI) mixed with the hardener (Euxit 50KII) with a mixing ratio (3: 1) by weight. Figure (1) shows the Epoxy. Type E fiberglass formed as two-way simulated fiber mat (0-90) (Woven Roving) with density of 2.45g/cm³, and elongation 4.88% [8] as shown in Figure 2. The nano materials which were used as fillers are multi-walled carbon nanotubes (MWCNTs) supplied from USA Cheap tubs company, shown in figure 3. Calcium Carbonate Nano particles (CaCO₃) supplied by Sky Spring Nanomaterials, Inc. USA, shown in figure 4. The samples casting process gone through the following steps. First, the amount of nano materials prepared according to the sample’s weight fraction (1 or 2%wt) of the epoxy resin. Then, the nanomaterials were added to the epoxy and mixed within a magnetic hotplate stirrer for 10 minutes at a speed of 1000 r.p.m until the mixture is homogeneous and without bubbles. A vibrator of ultrasonic (Ultrasonic 1200W MTI Corporation) is utilized for 25 minutes to disperse nanoparticles uniformly. After the vibrating operation, the hardener was added to the mixture with gentle mixing. All samples were molded by vacuum bagging techniques with 35% volume fraction (volume of the fiberglass to composites volume). Figure 5. illustrates the vacuum bagging. After solidification the produced composite sheet have been cut to specimens with standard dimensions according to ASTM-D3039 [9, 10] and ASTM-D2990[11]. The geometry and dimensions of the tensile and creep specimens are shown in figures 6and 7.
Figure 1. SWISS CHEM RESIN.

Figure 2. Woven Roving Fiber glass

Figure 3. MWCNTs

Figure 4. (CaCO₃) Nano particles

Figure 5. Casting with Vacuum bag

Figure 6. The standard samples for the tensile test.
3. Tensile Test

This test was performed according to the (ASTM-D3039) standard [9] for composite with fibers (fiberglass) in their composition. Large (50) KN tensile test device is used and the test has been achieved at room temperature in the lab. The most important mechanical properties, including yield strength, Ultimate tensile strength and modulus of elasticity have been drawn from the tensile test. The tests were applied to three specimens from each combination of materials evaluating the average value to satisfy further accurate results. Load has been applied at fixed head velocity of (2mm/minutes) till specimens have been fractured. Some specimens (before and after test) are shown in Figure (8).

![Figure 8](image)

(a) Before the test  
(b) After the test

Figure 8. The samples before and after the tensile test.

Creep Test

The creep test is based on a reading of the deformation (elongation) of the sample. Then the strain during the predetermined period of time is calculated as follows:

\[ \varepsilon = \Delta L / L \]  
\[ \varepsilon(t) = \Delta L(t)/L \]  
\[ \varepsilon_o = L_o / L \]
where $\Delta L$ is the increasing in the sample length after applying the load during the time. 
$L$ is the real length of the sample.

The creep test device required for experimental work is designed and manufactured according to laboratory specifications and standards as shown Figure 9.

![Figure 9. The creep test rig.](image)

The creep test applied to all specimens in sequence (the specimens are fixed as figure 10)

![Figure 10. Sample in the fixation](image)

A constant load is applied causing a tensile stress of 147.15 MPa, the deformation was measured by means of strain dial gauge as illustrated in figure 11.

![Figure 11. Dial gauge.](image)
Table 1, shows the maximum values from the creep test for 12 hours at 40ºC temperature. As it is shown, when adding (MWCNTs + CaCO$_3$) to the epoxy, the values of the creep strain decreased. According to this data, it is clear that the addition of nanoparticles maintains the structural bonding of epoxy resins as it gives them hardness and sufficient strength for the fiberglass used in the composite.

| Weight fraction of hybrid Nano | Max. creep strain | Time (hr) |
|--------------------------------|------------------|-----------|
| With out                       | 0.137            | 12        |
| 1%                             | 0.123            | 12        |
| 2%                             | 0.1115           | 12        |

Results & Discussion
The experimental results to be discussed include the results of the tensile and creep tests for different types of laminated composites with nanocomposite materials.

Tensile Results
The ultimate tensile strength and Young's modulus were obtained from the tensile tests for all samples. Table (2) gives those mechanical properties of the composite specimens. As noticed, the studied hybrid nanomaterials give better mechanical properties than any type of nanomaterial alone (MWCNTs or CaCO$_3$), as shown in figures (12 & 13).

| Sample components                  | Additives Weight Fraction (%wt.) | Mechanical Properties |
|------------------------------------|---------------------------------|-----------------------|
| Epoxy / glass fiber Without additive | 0                               | $\sigma_{ult}$ (MPa)  |
|                                    |                                 | $E$ (GPa)             |
| Epoxy / glass fiber + CaCO$_3$     | 1                               | 213.867               |
|                                    |                                 | 1.6574                |
| Epoxy / glass fiber + MCNTs        | 1                               | 220.4                 |
|                                    |                                 | 1.5854                |
| Epoxy / glass fiber + MCNTs + CaCO$_3$ | 1                              | 224.267               |
|                                    |                                 | 1.6821                |
|                                    | 2                               | 242.8                 |
|                                    |                                 | 1.449                 |
|                                    | 2                               | 257.3                 |
|                                    |                                 | 2.1237                |

Figure 12. stress – strain curves of the samples of neat Fiberglass / Epoxy (G) and samples filled with (1%wt) of MWCNTs (GM) and CaCO$_3$(GC)
Creep Results

The behavior of strain vs. time traced for the neat fiberglass/Epoxy and the samples with 1%wt and 2%wt, checking the stages of deformation in which it occurs, where a decrease in the area of the cross-section and a continuous increase in the length of the sample due to the increase in stress until the failure occurs. Creep depends not only on the value of the applied load but depends on the time duration of load application too. Figures 11 and 12 explain the influence of adding the hybrid nanocomposite (MWCNTs, CaCO$_3$) on the creep behavior (strain-time) relation. Based on the experimental results it is showed that the addition of nanomaterials improved the creep behavior of composite material (G/MWCNTs+ CaCO$_3$) this is apparent by the low values of strain for the epoxy resin reinforced woven glass fiber - with hybrid nanocomposite contrasting with epoxy resin reinforced woven glass fiber without hybrid nanocomposites. Because hybrid nanocomposites MWCNTs+CaCO$_3$ (GMC) resist slipping, re-orientation, and polymer chain motion in nanocomposites. That is, it restricts creep deformation because the strength of the Nano fillings composites is highly dependent on the interfacial adhesion between the polymer matrix and the Nano filler that helps transfer a small portion of the stress to the filling particles through deformation [12, 13].

Figure 13. stress – strain curves of the samples of neat Fiberglass / Epoxy (G) and samples filled with (2%wt) of MWCNTs (GM) and CaCO$_3$ (GC)

Figure 14. Comparative Creep strain (1%wt) hybrid nanocomposite [0.5MWCNTs%+ 0.5% CaCO$_3$ (1%wt GMC)]. at 147.15Mpa (ambient temperature 40°C)
### Conclusions

The following are concluded from this work:

1. the improvement in the creep strain behavior is about (10.22 %) & (18.61 %) for the samples with (MWCNTs + CaCO₃) wt. respectively.
2. Creep resistance is very high for hybrid nanocomposites materials (GMC) compared to neat samples or with single nano material (MWCNT, CaCO₃).
3. The results showed that the hybrid nanocomposite gave better mechanical properties than any type of the single nano filled samples.
4. Adding the fillers to the base matrix material leads to an increase in the strength of the matrix, reducing the elongation,
5. It has been observed that the elongation properties decreased by adding the filling and this is due to the change in particles movement and stress concentration and the development and spread of cracks.

As, stated in other works, the creep strain is steeper and faster when the samples exposed to high temperatures for long periods, because the stress reduces the barrier of activating the bonds and disassembles them, and thus allows the partial chains to move more easily with increasing temperature, and these chains increase the creep acceleration, However, the creep strain values increase with increasing the temperatures for the same time interval [14] so, it is recommended to continue this work with samples exposed to temperatures more than 50 or 60°C.

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