Preparation and Dynamic Mechanical Analysis of Glass or carbon Fiber/Polymer Composites

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Abstract. Nowadays, glass fiber reinforced polymer (GFRP) and carbon fiber reinforced polymer (CFRP) composites are broadly used so it is essential their properties to be explored in depth. In this study, GFRPs and CFRPs were produced by vacuum bag oven method and their viscoelastic behaviour at elevated temperatures was investigated. In particular, by Dynamic Mechanical Analysis experiments properties such as storage modulus, loss modulus, tanδ and glass transition temperature were determined. The results showed that, apart from a very small increment in the low temperatures in some of the compounds, as the temperature increases the storage modulus of the composites decrease while the composite containing unidirectional glass fibers in longitudinal direction and 34% percentage of matrix achieved the higher storage and loss modulus. Furthermore, as the unidirectional fibers change direction from transverse to longitudinal the GFRPs exhibit much higher storage and loss modulus.

1. Introduction
Composites are being used by humans from ancient times. Their great advantage is that they combine the properties of their individual substances so the mechanical behaviour of the final composite is better than the one of the initial materials. A great category of them is the fiber reinforced polymer (FRP) composites which are composed of fibers, such as glass or carbon, embedded in a polymer matrix. Types of them are the glass fiber reinforced polymers (GFRPs) and carbon fiber reinforced polymers (CFRPs) while both of them are familiar for their low weight, high strength and modulus. The applications of these compounds involve a broad range such as in automotive, aircraft, marine and space industry.

Furthermore, prepregs materials can be used in order to achieve maximum performance of the composites. Prepregs are materials which have been pre impregnated with the resin and they are ready to be cured under heat and pressure combination. Advantages such as excellent quality of the final product, high fibers percentage, determination of the exact ratio of matrix-reinforcement percentage and many other have done the prepregs very popular in the composites industry while their high cost is a restrictive factor for their extensive use.

Generally, the production of the composites by prepregs can be separated in autoclave and out of autoclave (OOA) methods. By autoclave use, high quality compounds are produced. It should be noted that, especially for big parts, autoclaves imply considerable costs for function and tooling [1].
Nowadays, scientists are trying to develop new techniques such as the vacuum bag oven process, without the increased cost of the autoclave’s use. During the vacuum bag oven technique, the composite is placed in a mould under a vacuum bag, the air is evacuated from the system and the cure of the compound takes place in an oven. An important factor which must be taken into account in OOA methods is to avoid the porosity and form of voids in the final composite due to the present of air during its production. As a result of the broadly use of GFRPs and CFRPs, it is essential their properties to be entirely investigated so as the mechanical behaviour of them to be absolutely predictable. An important point in the investigation of the materials is how their properties are influenced by fluctuations of the temperature. An efficient way for the exploration of the materials’ mechanical behaviour is the Dynamic Mechanical Analysis (DMA) which offers study of the rheology under different temperatures, frequencies or loadings. In particular, by these experiments properties such as storage modulus, loss modulus and tanδ can be determined. Moreover, DMA offers the ability for the determination of the glass transition temperature (Tg). Below this temperature many amorphous polymer are hard and stiff while above it they are transformed to soft.

By DMA tests, researchers have investigated various properties of materials such as epoxy based glass/carbon hybrid composite laminates [2] epoxy/poly(methylmethacrylate)/ glass fiber composites [3], glass fiber reinforced polymer composites [4,5,6,7] and carbon fiber reinforced polymer composites [8,9,10]. Furthermore, Gupta and Rohit [11] explored the dynamic mechanical properties of glass fibres reinforced epoxy composites with varying numbers of layers and it was found that the highest storage modulus value was achieved by the composite with the highest number of layers. By dynamic mechanical analyser method, Kister and Dossi [12] monitored the cure characteristics of carbon/epoxy composite prepreg. Goertzen and Kessler [13] evaluated carbon fiber/epoxy matrix composite through dynamic mechanical analysis. Katunin et al. [14] studied the ageing of GFRP laminates in deionized and seawater and investigated them by various tests including DMA. However, a broad research study on the dynamic mechanical properties of polymer matrix composites containing carbon or glass fibers in different orientations while the thickness of the compounds and the percentage of the matrix are variable seems to be missing. Specifically, it would be useful a comparison between the viscoelastic properties of GFRP composites containing unidirectional fibers in longitudinal or transverse direction.

In our study, GFRPs and CFRPs were produced and by DMA tests their dynamic mechanical properties were investigated in order to determine the optimal fibers’ type and orientation under elevated temperatures. The basic scope of this study is for the GFRPs to compare unidirectional fabrics when they are placed in longitudinal or transverse direction and for the CFRPs to provide some details for their viscoelastic behaviour because these compounds have different thicknesses so the results of the tests cannot be compared directly. Furthermore, two different methods were used for the determination of composites’ Tg temperature. Particularly, the compounds were designed and tested for transport industry applications.

2. Experimental

2.1. Materials

For the production of the composites prepreg in various layers numbers were used so as the thickness of the compounds to be variable. Both carbon and glass fibers were in unidirectional or woven fabric type. The glass fiber epoxy prepregs, both the unidirectional and woven types, were fabricated by Delta-Preg S.p.A. by the resin DT 806R. The carbon fiber prepregs were prepared by Impregnatex Compositi S.r.l. using the resin IMP 503Z. In Table I, the specifications of the prepregs and the fibers’ type which were used are presented. The unidirectional were placed in longitudinal (Fig. 1) or transverse direction and for the CFRPs to provide some details for their viscoelastic behaviour because these compounds have different thicknesses so the results of the tests cannot be compared directly.
GFRPs consisted of unidirectional fibers in longitudinal direction
GFRPs consisted of unidirectional fibers in transverse direction
GFRPs consisted of woven fabrics at 0°, (fibers 0°/90°)
GFRPs consisted of woven fabrics at 45°, (-45°/+45°)
CFRPs consisted of unidirectional fibers in longitudinal direction
CFRPs consisted of unidirectional fibers in transverse direction
CFRPs consisted of woven fabrics 0°, (0°/90°)
CFRPs consisted of woven fabrics 45°, (-45°/45°)

Furthermore, two more GFRPs (UD fibers in longitudinal or transverse direction) with different percentage of matrix were produced. These composites are presented analytically in Table I.

![Figure 1. Base of orientation: unidirectional fibers in longitudinal direction (0°)](image)

### Table 1. Types of prepregs—fibers and specifications of composites

| Symbol of composite | Type of prepreg-fabric name | Type of fibers and orientation | Percentage of matrix | Thickness of composite (mm) |
|---------------------|-----------------------------|-------------------------------|----------------------|-----------------------------|
| GF-UD-L-34%         | VV430U-DT806R-34            | Glass fiber, unidirectional, longitudinal (0°) | 34%                  | 2.35                        |
| GF-UD-TR-34%        | VV430U-DT806R-34            | Glass fiber, unidirectional transverse (90°) | 34%                  | 2.35                        |
| GF-UD-L-34%         | VV430U-DT806R-39            | Glass fiber, unidirectional longitudinal (0°) | 39%                  | 2.85                        |
| GF-UD-TR-39%        | VV430U-DT806R-39            | Glass fiber, unidirectional transverse (90°) | 39%                  | 2.85                        |
| GF-W-O°-37%         | VV320P-DT806R-37            | Glass fiber, woven0° (0°/90°) | 37%                  | 2.35                        |
| GF-W-45°-37%        | VV320P-DT806R-37            | Glass fiber, woven, 45° (-45°/+45°) | 37%                  | 2.35                        |
| CF-UD-L-38%         | IMP503Z/GV420U              | Carbon fiber, unidirectional longitudinal (0°) | 38%, +1%             | 3.7                         |
| CF-UD-TR-38%        | IMP503Z/GV420U              | Carbon fiber, unidirectional transverse (90°) | 38%, +1%             | 2.85                        |
| CF-W-O°-38%         | IMP503Z/GG630T              | Carbon fiber, woven 0° (0°/90°) | 38%, +1%             | 3.15                        |
| CF-W-45°-38%        | IMP503Z/GG630T              | Carbon fiber, woven 45° (-45°/+45°) | 38%, +1%             | 1.3                         |

#### 2.2. Production method

By vacuum bag oven method GFRPs and CFRPs were fabricated. The base of this process is the construction of the vacuum bag system.
Specifically, the steps described bellows were followed. Firstly, the prepreg laminates were cut in the desired dimensions and they were put, in various layers numbers, in the moulds. Over them, a release film and a breath cloth were placed. Then, the system was covered by a vacuum bag which was stuck in the perimeter of the mould by sticky tape. Also, the bag was connected with the vacuum pump by the vacuum valve. After the sealing of the system, air leak was tested as it is one of the most critical problems in these processes. Finally, the system was placed in the oven where the cure process took place by the following steps. Firstly, the temperature was increased slowly up to 65 °C where it was held for 30 minutes, subsequently the temperature was raised to 120 °C where it was maintained for 60 minutes and finally it was slowly reduced up to ambient temperature. Totally, this process lasted about 4 hours. The composites were let to solidify for one day at room temperature and then samples were mechanically cut in the desired dimensions.

![Figure 2. Production of composites.](image1)

2.3. Experimental details

The DMA experiments were carried out by METTLER TOLEDO instrument over a temperature range of 30-200 °C, at a steady frequency of 1Hz while the heating rate was 2 °C/min. The displacement amplitude adjustment was 20 μm whereas a force of 1 N was used as preload. The samples were tested under a 3 point bending configuration mode which is suggested by METTLER TOLEDO for composite and stiff materials while the span length between the supports was 30 mm. The specimens were in rectangular shape with dimensions of (40x7) mm whereas their thickness was variable and it is presented in table I.

![Figure 3. Plates of composites.](image2)

![Figure 4. Specimens after the experiments.](image3)
3. Results

3.1. Storage modulus
The storage modulus (E’) is correlative with the stored energy. Figures 5 and 6 depict the effect of the temperature on the storage modulus of GFRPs and CFRPs, respectively. As it can be seen, except for a slightly increase in the low temperatures in some of the composites, the storage modulus decreases as the temperature increases. A remarkable point is that in all of the curves there is a sharp decrease which is associated with the glass transition temperature (Tg). Usually, this steep fall determines the maximum operating temperature of the materials. Despite the fact of its relative small thickness, the GFRP compound with 34% percentage of matrix exhibited much higher storage modulus than the GFRP with 39% matrix. This can be attributed to the positive effect which the glass fibers offer to the composite and it is indicative of the fact that a small increment in the percentage of them can give a high positive change in the measured values. In all cases of GFRPs, the placement of the unidirectional fibers in the longitudinal direction seems to be much more effective than if they are placed in the transverse direction. Furthermore, the GFRP sample with woven fabric at 0° succeeded higher storage modulus than the GFRP with woven fabric at 45°.

![Figure 5. Storage modulus of GFRPs as a function of temperature.](image1)

![Figure 6. Storage modulus of CFRPs as a function of temperature.](image2)

3.2. Loss modulus
The loss modulus (E'') is related with the energy which was dissipated as heat under dynamic mechanical analysis experiments. The values of the loss modulus at elevated temperatures of GFRPs and CFRPs are
presented in figures 7 and 8. As it can be revealed from these graphs, the values are almost stable at low temperatures while after about the 70-80 °C a steep increase can be observed until a peak value. Then a sharp decrease is obtained until very low values which are maintained until 200 °C. As in storage modulus graphs, GFRPs composed of unidirectional fibers in longitudinal direction have higher loss modulus than the same composites with the fibers in the transverse direction.

![Figure 7. Loss modulus of GFRPs as function of temperature.](image1)

![Figure 8. Loss modulus of CFRPs as a function of temperature.](image2)

3.3. $\tan\delta$

The loss factor ($\tan\delta$) is defined as the ratio of loss modulus to storage modulus:

$$\tan\delta = \frac{E''}{E'}$$

and it is independent of geometry effects.

Figures 9 and 10 show the $\tan\delta$ values of GFRPs and CFRPs as temperature increases. From these figures, it is observed that after about the 80 °C there is a rapid raise in the values of the $\tan\delta$ until a peak point while then a steep decrease is observed.

In figure 10, a remarkable observation is that the curve of the CF-W-45°-38% after the peak follows a non-linear decrease. This can be attributed to degradation of the specimen at high temperatures under the 3
point bending mode due to its small thickness. Moreover, a second specimen of this composite was tested and the same nonlinear curve was observed.

![Figure 9. tanδ of GFRPs as function of temperature.](image)

![Figure 10. tanδ of CFRPs as a function of temperature.](image)

3.4. Glass transition temperature (Tg)

The Tg temperature is a point for disagreement between the scientists because there are many methods for its determination. As a result, it is essential for the method which was followed for its specification to be mentioned. In this research, the Tg temperatures were obtained by two different ways; peak of loss modulus and tanδ curves respectively. These results are presented in table II. As it can be seen from a comparison between these two methods, the Tg temperatures obtained by the peak of tanδ curves are a little higher than these revealed from the loss modulus curves.

| Material          | Tg values obtained by loss modulus peak | Tg values obtained by tanδ peak |
|-------------------|----------------------------------------|---------------------------------|
| GF-UD-L-34%       | 116,91                                 | 120,54                          |
| GF-UD-TR-34%      | 111,69                                 | 119,85                          |
| GF-UD-L-39%       | 117,47                                 | 121,27                          |
| GF-UD-TR-39%      | 111,68                                 | 121,83                          |
| GF-W-0°-37%       | 120,79                                 | 123,66                          |
| GF-W-45°-37%      | 110,46                                 | 120,01                          |
| CF-UD-L-38%       | 103,08                                 | 107,54                          |
| CF-UD-TR-38%      | 96,27                                  | 104,96                          |
| CF-W-0°-38%       | 100,97                                 | 106,56                          |
| CF-W-45°-38%      | 88,93                                  | 101,42                          |
4. Conclusions
During this study, GFRPs and CFRPs were produced by vacuum bag oven method and their dynamic mechanical properties were explored. Various types of fibers’ architecture were used such as unidirectional fibers placed in longitudinal or transverse direction and woven fabrics oriented at 0° (fibers 0°/90°) or 45° (fibers -45°/+45°). From this viewpoint the results indicate that as the percentage of glass fibers in GFRPs increases the composites exhibit higher storage and loss modulus. Furthermore, in terms of storage and loss modulus the use of the fibers in the longitudinal direction is much more effective than if they are used in the transverse direction. In addition, very low thickness of CFRPs seems to be a restrictive factor for the successful completion of DMA temperature sweeps experiments under three point bending mode. Generally, the vacuum bag oven method is a cheap and very efficient method for the production of FRP composites by using prepreg materials.

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6. References
[1] T. Centea, L.K. Grunenfelder and S.R. Nutt, “A review of out-of-autoclave prepregs – Material properties, process phenomena, and manufacturing considerations,” Composites: Part A: Applied Science and Manufacturing, Volume 70, March 2015, pp. 132-154, https://doi.org/10.1016/j.compositesa.2014.09.029
[2] R. Murugan, R. Ramesh and K. Padmanabhan. “Investigation on static and dynamic mechanical properties of epoxy based woven fabric Glass/Carbon hybrid composite laminates,” Procedia Engineering, Volume 97, 2014, pp. 459-468, https://doi.org/10.1016/j.proeng.2014.12.270
[3] D. Olmos , K. Bagdi , J. Mózcó, B. Pukánszky and J. González- Benito, “Morphology and interphase formation in epoxy/PMMA/glass fiber composites: effect of the molecular weight of the PMMA,” Journal of Colloid and Interface Science, Volume 360, Issue 1, August 2011, pp. 289-299, https://doi.org/10.1016/j.jcis.2011.04.028
[4] João R.Correia, Marco M. Gomes, José M. Pires and Fernando A. Branco, “Mechanical behaviour of pultruded glass fibre reinforced polymer composites at elevated temperature: experiments and model assessment,” Composite Structures, Volume 98, April 2013, pp. 303-313, https://doi.org/10.1016/j.compstruct.2012.10.051
[5] Yu Bai, Nathan L. Post, John J. Lesko and Thomas Keller “Experimental investigations on temperature-dependent thermo-physical and mechanical properties of pultruded GFRP composites,” Thermochimica Acta, Volume 469, Issues 1-2, March 2008, pp. 28-35, https://doi.org/10.1016/j.tca.2008.01.002
[6] R. Steigmann, A. Savin, V. Goanta. P. D. Barsanescu, B. Leitou, N. Itimie, M. D. Stanciu and I. Curtu, “Determination of mechanical properties of some glass fiber reinforced plastics suitable to wind turbine blade construction,” IOP Conference Series: Materials Science and Engineering, Volume 147, Issue 1, 2016, doi:10.1088/1757-899X/147/1/012140
[7] S. Cabral-Fonseca, J. R. Correia, M. P. Rodrigues and F. A. Branco, “Artificial accelerated ageing of GFRP pultruded profiles made of Polyester and Vinylester resins: characterisation of physical-chemical and mechanical damage,” strain, 2012, Volume 48, Issue 2, pp. 162-173, doi: 10.1111/j.1475-1305.2011.00810.x
[8] Vasileios M. Drakonakis, James C. Seferis and Charalambos C. Doumanidis, “Curing pressure influence of out-of-Autoclave processing on structural composites for commercial aviation,” Advances in Materials Science and Engineering, Volume 2013, Article ID 356824, 14 pages, doi:10.1155/2013/356824
[9] D. Kaka, J. Rongong, A. Hodzic and C. Lord, “Dynamic mechanical properties of woven carbon fibre reinforced thermoplastic composite materials,” Proc. of the 20th International Conference on Composite Materials (ICCM), July 2015
[10] Ana X. H. Yong, Graham D. Sims, Samuel J. P. Gnaniah, Stephen L. Ogin and Paul A. Smith, “Heating rate effects on thermal analysis measurement of Tg in composite materials,” Advanced Manufacturing: Polymer and Composites Science, Vol 3, pp. 43-51, 2017, doi:10.1080/20550340.2017.1315908

[11] M. K. Gupta and Kunwar Rohit, “Multi layers glass fibres reinforced epoxy composite: dynamic mechanical analysis,” Advanced Materials Proceedings, Volume 2, Issue 8, pp.518-520, 2017, doi: 10.5185/amp.2017/810

[12] Guillaume Kister and Eleftheria Dossi, “Cure monitoring of CFRP composites by dynamic mechanical analyser,” Polymer Testing, Volume 47, October 2015, pp. 71-78, https://doi.org/10.1016/j.polymertesting.2015.08.009

[13] W.K. Goertzen and M.R. Kessler, “Dynamic mechanical analysis of carbon/epoxy composites for structural pipeline repair,” Composites Part B: Engineering, Volume 38, Issue 1, January 2007, pp. 1-9, https://doi.org/10.1016/j.compositesb.2006.06.002

[14] Andrzej Katunin, Adam Gnatowski and Wojciech Kajzer, “Evolution of static and dynamic properties of GFRP laminates during ageing in deionized and seawater,” Advanced Composites Letters, Vol. 24, July 2015, pp. 38-43