# DEVELOPMENT AND THERMOMECHANICAL PROPERTIES OF $B_4C$, TiC AND GRAPHITE/ EPOXY COMPOSITES

S. Gioti[1], S.G. Stavropoulos[2], A. Sanida[3], G.C Psarras[4]

Smart Materials and Nanodielectrics Laboratory, Department of Materials Science, University of Patras, 26504 Patras, Greece
[1]Email: msci1547@upnet.gr, [2]Email: stavropoulos@upatras.gr,
[3]Email: ksanida@upatras.gr, [4]Email: G.C.Psarras@upatras.gr

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## Abstract

In the present study, three series of epoxy composite systems were manufactured varying the filler type and content. The thermomechanical properties were investigated by dynamic mechanical analysis (DMA). The experimental results led to the conclusions: (i) storage modulus increase systematically with the addition of filler, (ii) the transition from glassy to rubbery state moves to higher temperatures and glass transition temperature shifts, in general, to higher values in composite systems. Optimum performance varies not only with filler content but also with filler type, at low, medium and high reinforcing phase content.

## 1. Introduction

Polymer matrix composites form an important class of technologically important materials mainly because of their thermomechanical and dynamic mechanical behavior [1,2]. Composites can be classified according to the aspect ratio of the used filler and their performance can be tailored by suitably selecting the type and the amount of inclusions. Thermosetting-polymer based composites undergo a transition, known as glass to rubber transition, signifying the transition from the stiff and rigid performance to the soft and weak response. In the present study three series of composite systems were manufactured varying the type and the content of filler. Dynamic mechanical properties were determined via dynamic mechanical analysis (DMA) [3-5]. The aim of the present work is to examine the reinforcing ability, on the dynamic mechanical properties, of $B_4C$, TiC, and graphite micro-particles.

## 2. Experimental

The polymer matrix used in this research was a low viscosity epoxy resin with the trade name Renlam LY 5138-2 and curing agent Ren HY 5138 provided by Huntsman advanced materials. Boron Carbide ($B_4C$) particles with diameter less than 10 μm, Titanium Carbide (TiC) particles with diameter less than 4 μm and Graphite (C) particles with diameter less than 20 μm, acted as filler. All three reinforcing materials were provided by Sigma-Aldrich.

Resin and curing agent were mixed, at a 100:23 per weight mixing ratio, at ambient temperature for 5 min. Subsequently, predetermined amounts of each powder were added and the mixture was stirred at a low rate using a sonicator for 10 min in order to avoid agglomeration. Then, the mixture was poured into silicone molds and cured at ambient temperature for 7 days followed by post curing at T = 120ºC for 4 h. Filler content is expressed in parts per hundred resin per weight (phr).

The dynamic mechanical properties of the produced were investigated using a dynamic mechanical analysis device, Q800 TA instruments. More specifically, viscoelastic properties including, phase transition, storage modulus, loss modulus and loss tangent (tan delta) of the composites were determined. DMA scans were conducted in the temperature range from room temperature to 120ºC, at a scanning rate 5ºC /min, and at a constant applied frequency of 1 Hz. The employed type of test was three-point bending.

## 3. Results

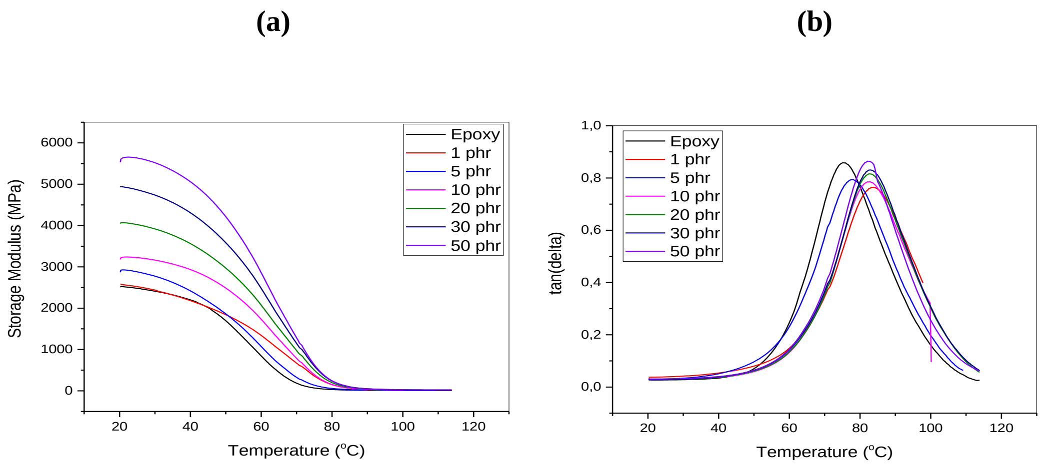


**Figure 1.** (a) Storage Modulus and (b) tanδ as a function of temperature, for the epoxy/$B_4C$ composites.

**(a)** **(b)**

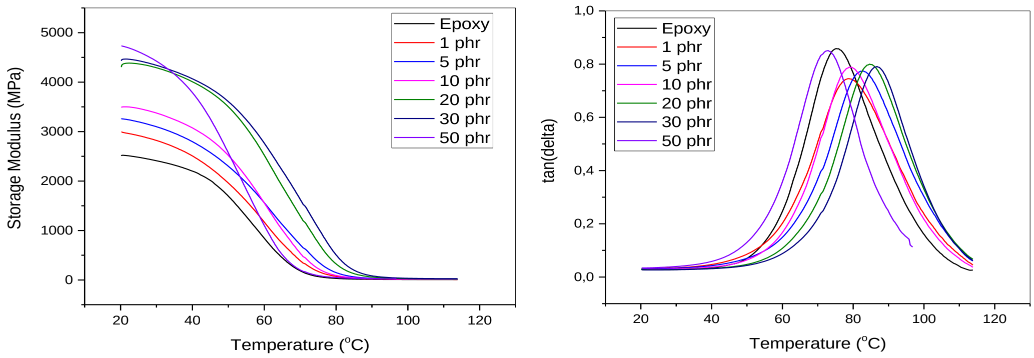


**Figure 2.** (a) Storage Modulus and (b) tanδ as a function of temperature, for the epoxy/graphite composites.

**(a)** **(b)**

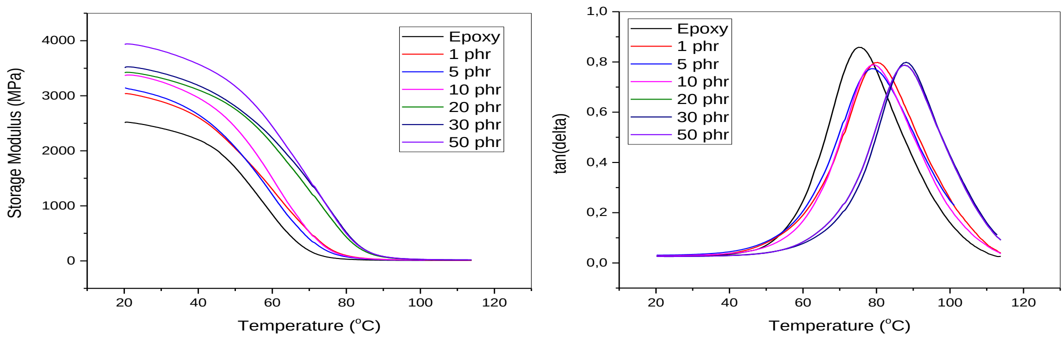


**Figure 3.** (a) Storage Modulus and (b) tanδ as a function of temperature, for the epoxy/TiC composites.

In figures 1, 2, 3 storage modulus and tanδ are represented as a function of temperature for all examined systems. In all cases, storage modulus starts from high values, followed by a decrease in the form of a step-like transition [6-8]. This is attributed to the transition from the glass to the rubber state of the polymer matrix and is demonstrated by forming peaks in the tanδ spectra. Experimental data show a systematic behavior with filler concentration. Storage modulus increases with the addition of filler implying mechanical reinforcement of the composites. Furthermore, the transition from glassy to rubbery state shifts to higher temperatures. This behavior showcases increased $T_g$ implying strong interactions between matrix and reinforcement due to the strong adhesion between matrix and filler as well. Recorder peaks in Tanδ spectra indicate glass transition temperature ($T_g$) being in accordance with the aforementioned behavior.

(a) (b) (c)

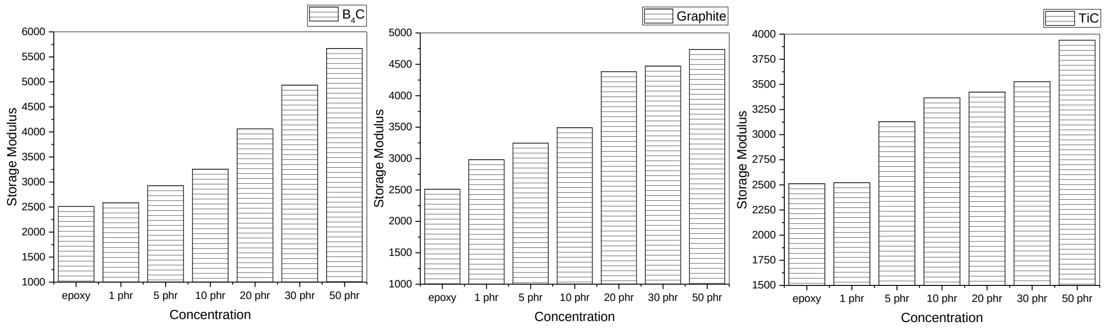


**Figure 4.** Bar diagrams of storage modulus as a function of filler concentration, for (a) epoxy/$B_4C$, (b) epoxy/graphite, and (c) epoxy/TiC composites.

(a) (b) (c)

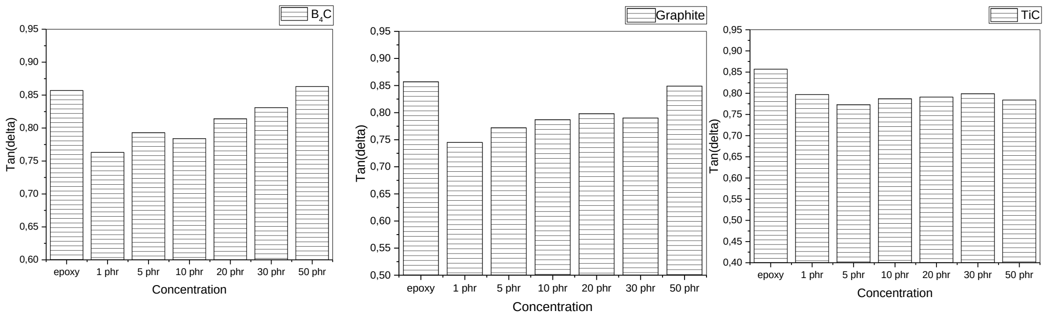


**Figure 5.** Bar diagrams Tanδ as a function of filler concentration, for (a) epoxy/$B_4C$, (b) epoxy/graphite, and (c) epoxy/TiC composites.

In case of composites with $B_4C$ reinforcement is observed a systematic increase of E΄ with filler concentration. Storage Modulus of the 50 phr specimen over doubles its value compared to the epoxy. Composites with graphite reinforcement shows optimum behavior at a lower filler level.

Tanδ bar diagrams show variation of damping coefficient with filler's concentration in all materials. First, is observed a decrease of maximum values followed by a progressive increase, in the case of composites with $B_4C$/ graphite reinforcement and a stabilization in case of composites with TiC reinforcement. These changes in tanδ could be attributed to macromolecules- particles and particles-particles interactions [9].

**(a)** **(b)** **(c)**

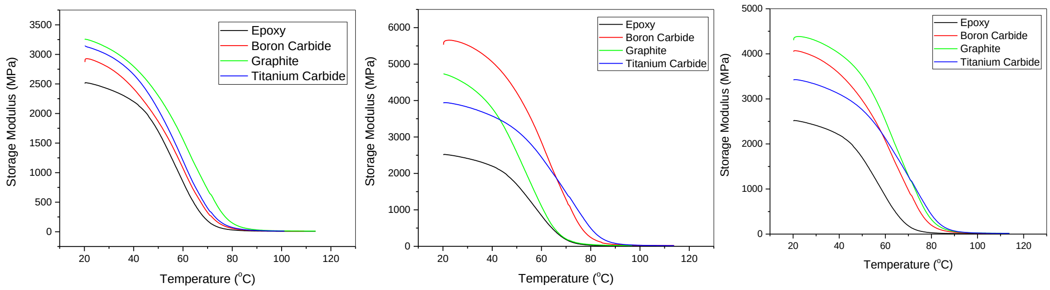


**Figure 6.** Comparative plots of Storage Modulus as a function of temperature, for composites with constant filler content: (a) 5 phr, (b) 20 phr, (c) 50 phr, varying the filler type.

**(a)** **(b)** **(c)**

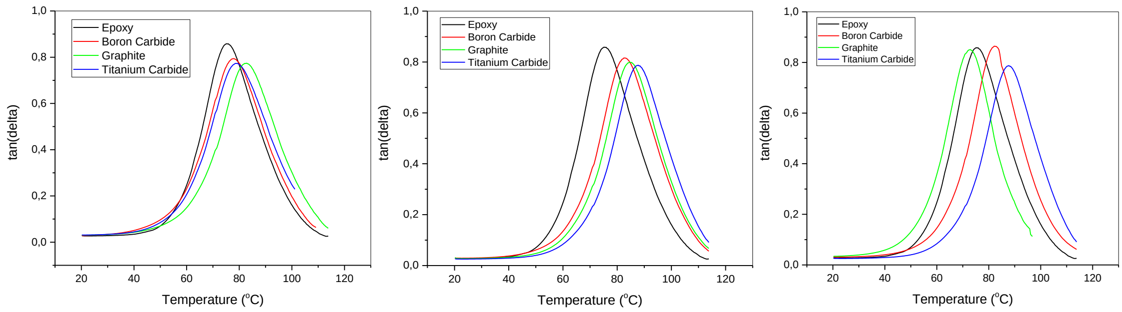


**Figure 7.** Comparative plots of Tanδ as a function of temperature, for composites with constant filler content: (a) 5 phr, (b) 20 phr, (c) 50 phr, varying the filler type.

In Figs. 6, 7 comparative diagrams of storage modulus and tanδ as a function of temperature are showed, for three different concentrations (5, 20, 50 phr) of the various reinforcing phases. It needs to be noted that the optimal behavior does not always correspond to the same reinforcing material. More specific, composites with TiC microparticles, exhibit higher storage modulus values at lower concentrations, while composites with graphite prevail at intermediate concentrations. Finally, it seems that the optimal behavior corresponds to composites with $B_4C$ reinforcement which have the higher E΄ at the maximum concentrations.

As tanδ is concerned, at lower concentrations (up to 10 phr) composites with $B_4C$ reinforcement shift their peak to higher temperatures. Shifting peaks to higher temperature indicate strong attractive forces between matrix and filler. In the rest of concentrations composites with TiC reinforcement show the highest glass transition temperature.

## 4. Conclusions

Summarizing, in this study three series of composites materials were successfully manufactured. The mechanical properties of these composites materials were investigated via DMA. Experimental data show that the addition of the filler reinforced significantly the mechanical properties of the composite systems. Different systems exhibit the optimum performance at different concentration regimes. The highest storage modulus value was achieved by the 50 phr Boron Carbide specimen, which exhibited a value over 5.5 GPa, corresponding to an increase of over 100% compared to the neat epoxy. Damping coefficient or tanδ decreases in nanocomposites at low and moderate filler content compare to the polymer matrix.

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