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DESIGNING PARTICLE REINFORCED POLYMERIC COMPOSITES WITH IMPROVED THERMAL PROPERTIES FOR SENSORS AND ACTUATORS

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Abstract: The herein paper approaches, theoretically and experimentally, the class of particle reinforced polymeric composites with the aim of designing a structure showing stability with temperature variation. A copper based, embedded in different volume fractions into an epoxy matrix (60%, 70%, 80%), composites samples were manufactured and subjected to dilatation measurements over a 20^0C to 250^0C temperature range to retrieve their CTE and size the major influencing factors on this thermal property. Several micromechanics based theoretical bounds from literature were used for data comparison and analysis.

1. INTRODUCTION

Particle reinforced polymeric composite materials are being used increasingly in a variety of modern engineering applications and this trend is likely to continue due to the fact that these materials possesses a number of highly desirable engineering properties that can be exploited to design structures with high demand on their performance.

To cope with the obvious limitations of polymers, for example, low stiffness and low strength, and to expand their applications in different engineering areas, different types of particulate fillers are often added to process polymer composites, which normally combine the advantages of their constituent phases. Particulate fillers modify the mechanical and thermal properties of polymers in many ways [1]-[5].

Coefficient of thermal expansion is an important property of composite's application for electronic packaging, thermal insulation and devices to control the influence of temperature on the operation of fibre-optic Bragg grating, pressure/force sensors, automotive engine parts, space applications etc.

Theoretical studies have shown that the coefficient of thermal linear expansion of composites is strongly dependent on the volume fraction, the distribution, inclusion size and the elastic and thermal properties of constituents. At the very fundamental level, micromechanical analysis facilitates understanding of how the individual material properties of constituent phases and their spatial distribution influence the structural behaviour and material property at macroscopic scale [5]-[10].

The aim of present study is to provide an understanding of the thermal properties in case of particle reinforced composite materials, the influencing factors on this property in order to develop engineered materials for novel configurations of sensors/actuators in mechatronics industry and not only.

2. EXPERIMENTAL RESEARCH

2.1 Sample manufacturing

Composite samples were manufactured using a self developed manufacturing technology, the phases – copper particles (Cu) and the polymeric matrix (epoxy 040T resin) being chosen out of other metallic particles (e.g. Al and Fe) and polymeric resins. The particles were embedded into the polymeric resin as having different volume fractions – 60%, 70% and 80%, respectively. The additives used were chosen as being chemical compounds showing compatibility with the other phases and allowing polymerization process initiation and development.

2.2 Experimental investigation

The thermal property under the research is the coefficient of thermal linear expansion (CTE). The CTE of each sample was retrieved using a dilatometer DIL 402 C from NETZSCH (Germany). A dedicated software - Proteus Analysis – was used to retrieve, compare and export thermal data in terms of thermal strains and linear coefficient of thermal expansion. Each sample was subjected to 2 successive heating stages in order to size the influence of the thermal cycling, with temperature rising linearly between the ranges 20^0C to 250^0C, at a heating rate of 1 K/min, into a static air atmosphere. To eliminate the systems errors, the dilatometer was calibrated by measuring a standard SiO_2 specimen under identical conditions.

3 THEORETICAL APPROACH

The CTE of particle reinforced composite materials is not easy to predict precisely especially when the materials of phase's influences have to be sized. Technical literature provides many expressions for predicting CTEs of the composites and among them the micromechanical based bounds models are imposing the lower/upper limits for the experimental retrieved data. One of the micromechanical based theoretical models used often in predicting the CTE bounds is known as *Levin-Hashin-Shtrikman*. The expressions for the upper and the lower bounds of CTE are derived based on the theoretical model developed by *Levin*, such as:

$$\alpha_c = (1 - V_p)\alpha_m + V_p\alpha_p + \frac{\alpha_p - \alpha_m}{\frac{1}{K_p} - \frac{1}{K_m}}\left[\frac{1}{K_c} - \left(\frac{1 - V_p}{K_m} + \frac{V_p}{K_p}\right)\right] \tag{1}$$

where the composite bulk modulus is considered bounded within:

$$K_c^{inf} \leq K_c \leq K_c^{sup} \tag{2}$$

the limits being known in the literature as the *Hashin-Shtrikman* bounds:

$$K_c^{inf} = K_m + \frac{(K_p - K_m)(3K_m + 4G_m)V_p}{(3K_p + 4G_p) + 3(1 - V_p)(K_p - K_m)} \tag{3}$$

$$K_c^{sup} = K_p + \frac{(K_m - K_p)(3K_p + 4G_p)(1 - V_p)}{(3K_p + 4G_p) + 3(K_m - K_p)V_p} \tag{4}$$

The other theoretical model that was used in the paper is known as the *Rosen-Hashin* bounds, such as:

$$\alpha_c = \alpha_p + (\alpha_m - \alpha_p)\frac{\frac{1}{K_c} - \frac{1}{K_p}}{\frac{1}{K_m} - \frac{1}{K_p}} \tag{5}$$

in which the composite's bulk modulus will be replaced with the *Hashin-Shtrikman* limits from expressions (3) and (4). In all the previous formulae the followings stands for: K_m, K_p and K_c are the bulk moduli of the matrix material, particles and the composite, respectively, in GPa, G_m and G_p are the shear moduli of the matrix material and particles, in GPa, α_m, α_p and α_c are the coefficients of linear expansion of the matrix material, particles and the composite, in K^{-1}, V_p being the particle volume fraction embedded within the polymeric matrix.

4 RESULTS AND DISCUSSION

Figure 1 represents the microscopic view, with an x500 magnification factor, for the 60% copper particles reinforced polymeric composite sample. The oxidation process is being present and can be sized at the particle-matrix interface as well as at the sample's surfaces.

In figure 2 is being shown the thermal strain variations for all the copper reinforced polymeric composites tested while in figure 3 is being shown both the thermal strain and instantaneous coefficient of thermal expansion variation with temperature for a 60% copper particles reinforced composite material. As it can be seen, the thermal strains increase with increasing of temperature, while the CTE decreases with increase in the particle content.

Every composite sample is characterized as showing a maximum value of its coefficient of thermal expansion over the temperature range at different temperatures, such as: for the 60% volume fraction of copper particles the peak is at 120.4^0C, for the 70% volume fraction of copper particles the peak is at 160.3^0C while in case of the 80% volume fraction of copper particles the peak is at 182.6^0C.

The instantaneous coefficient of thermal expansion, which is the thermal property retrieved experimentally, is not an invariant property over the temperature range. The associated curve, as it can be seen in Fig. 3, over a large temperature intervals has a linear variation but also few outside of range values. The later can be regarded, for all composite samples experimentally investigated, to the differences between the thermal properties of each constitutive, to the thermal barriers at the particles-matrix interfaces and to the behaviour of the polymer matrix at temperatures exceeding values higher than the glass transition temperature.

In Fig. 4 is being plotted the theoretically predicted and experimentally retrieved CTEs for all the copper reinforced composite samples under investigation. As it can be seen, the experimental values lies between the lower and the upper bounds predicted based on the *Rosen-Hashin* theoretical models, and are experiencing a decrease along with the increase in the particle content. This variation is normal and as information it has to be taken into account in designing the composite structures. It can be summarized that the higher the particle content the more stable become the composite structure but this fact may affect the other material properties such as mechanical or electrical properties. This stability at temperature variations or influence has to be tailored according to the other material property an optimum volume fraction for the particles being established within a min-max or max-min optimization approach.

The thermal cycling is another experimental related influencing factor. While in the first thermal cycling the composites are revealing a more up and down variation of their thermal strains over the entire temperature range, and this mostly due to the polymer chain rearrangements, broken and reconnection, a second thermal cycling lead to smoothly variations, the polymerization process ending. Supplementary thermal cycling does not lead to substantial modification in the composite behaviour with temperature variation. In

Fig. 5 is being plotted the thermal strains vs. temperature range for a 80% copper reinforced composite sample subjected to two successive heating cycles.

Fig. 1 Micrographic view of a 60% copper reinforced polymeric composite sample (x500 magnification)

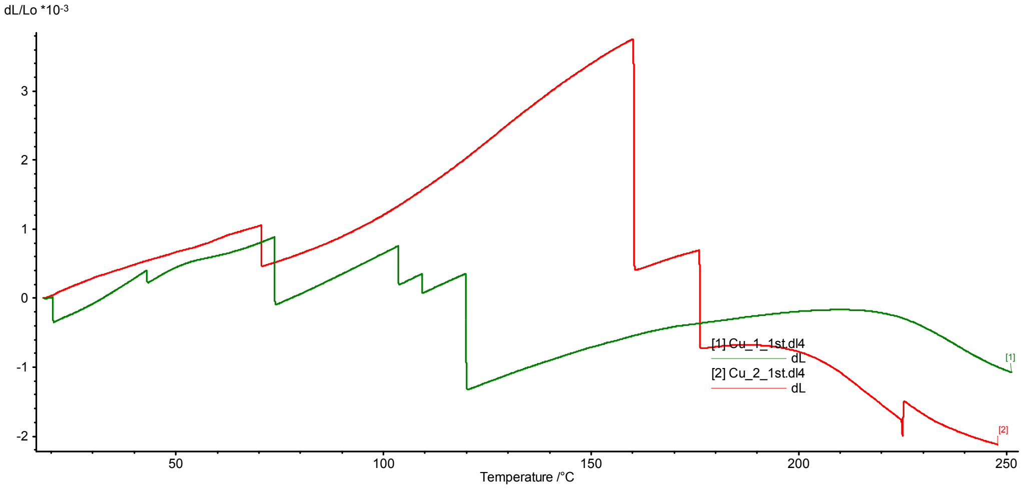

Fig. 2 Thermal strain variation with temperature for 60% and 70% Cu reinforced polymeric composites – first heating cycle

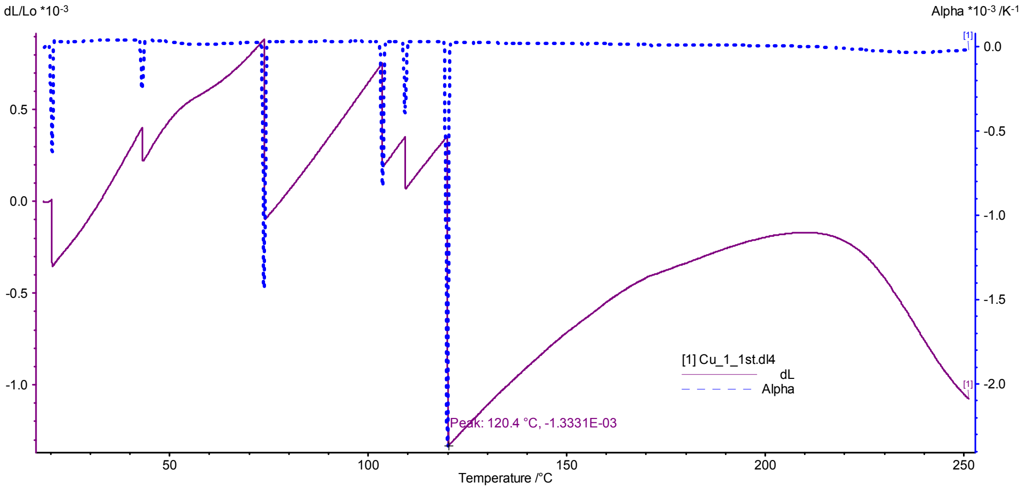

Fig. 3 Thermal strain and coefficient of themal expansion variations with temperatures for the 60% Cu polymeric composites – one heating cycle

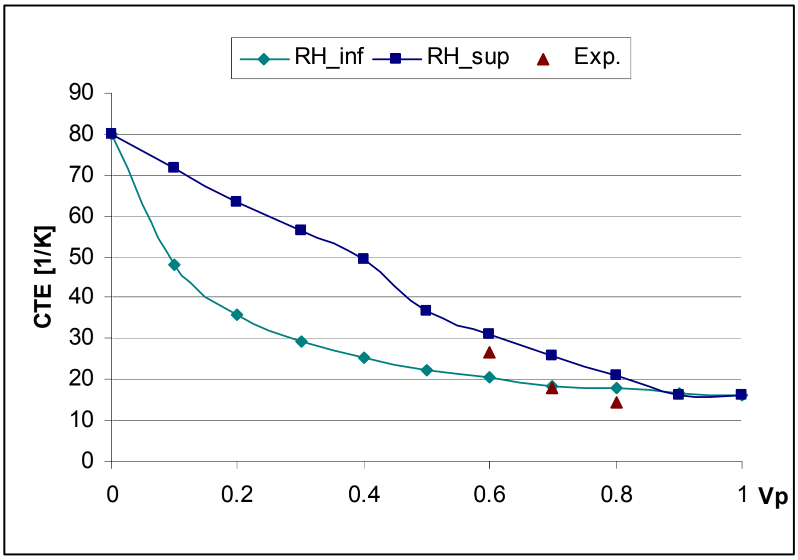

Fig. 4 Theoretical predicted (Rosen-Hashin models) and experimentally retrived CTE values for all the copper reinforced polymeric composites

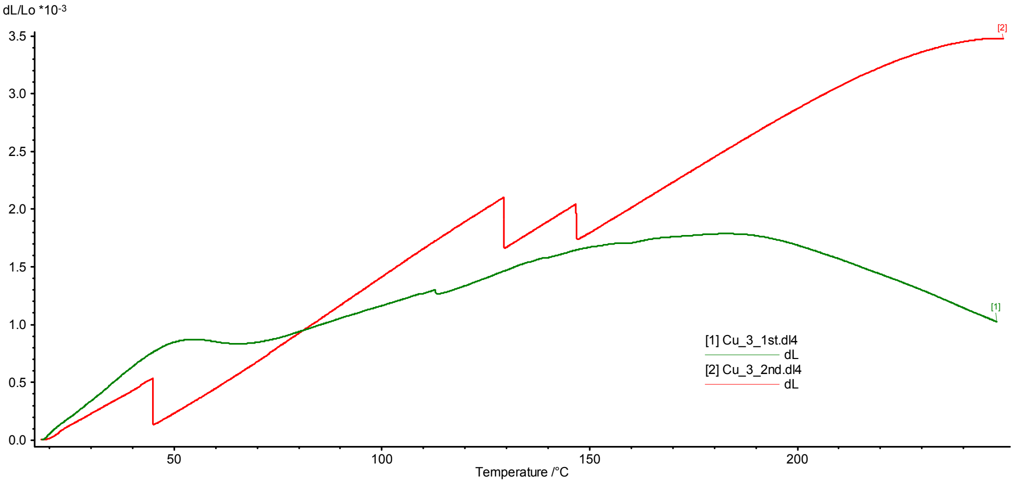

Fig. 5 Thermal strain variation with temperature for two succesive heating cycles applied upon the 80% copper reinforced composite sample

5 CONCLUSIONS

The subject developed herein has a more general purpose, not only to characterize a particular class of polymeric reinforced composite materials as potential material structures for sensors and actuators, but to develop, characterize and set up a data base for further comparisons for a novel class of advanced, multiphase composite materials. Several studies are under development with respect to the particle-particle combinations with the aim of retrieving and predicting their mechanical, electrical or thermal properties. All these approaches were done within the frame of revealing the multifunctional character of these polymeric composite structures, their wide range of applicability, especially while adressing thermal management issues.

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