Quantifying Porosity Influence on Metallic Particle Reinforced Composite Properties

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***Abstract*— The paper aims to present a comparative study on the theoretical predicted and experimentally retrieved elastic moduli and CTEs in case of different particle reinforced epoxy composite samples. Attempts on sizing the porosity influence on these material properties, as one of the influencing factors due to the manufacturing technology used, are done based on a well known micromechanics models from the literature. Once fully characterized, the results can be used in further composite design processes and/or engineering applications such is mechatronics, electronics, automotive or space industries.**

***Index Terms*— Composite, properties, polymer, porosity.**

I. 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], [4], [6].

Elastic properties are employed in the structure design and further analyses and simulations from mechanical or combined influences point of view. Knowing this important material property may help other developments in material characterization. With respect to this composite class, a higher elastic modulus and reduced coefficient of thermal expansion (CTE) is highly desirable and can be attain by incorporating the different types of particles into the matrix.

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 fiber-optic Bragg grating, pressure/force sensors, automotive engine parts, space applications etc.

Theoretical studies have shown that the effective elastic moduli and coefficient of thermal expansion of composites is strongly dependent on the volume fraction, the distribution, inclusion size and the elastic and thermal properties of constituents.

Many theoretical studies have used micromechanics models, such as dilute, self-consistent method, Mori-Tanaka approach, composite spheres (cylinders) model and differential method or bounds expressions to predict the effective properties of the composite material [2], [5].

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 behavior and material property at macroscopic scale.

A problem arises when the microstructure has 3 or more individual phases, case in which the micromechanical approach imply a multi-step or an overall description of the effective material property. Either the method chosen, neither give the same result.

Porosity is an unavoidable part of all particle reinforced composite materials due to the mixing and consolidation of two different materials phases, the particles and the matrix. Usually, during the manufacturing steps the porosity content is being kept under a certain volume fraction, but it happens to make a noteworthy contribution to the overall composite volume. A number of factors are responsible for these: complex surface chemistry of particles which complicates particle/matrix bonding, irregular form and size of particles, applied processing techniques, etc. [7], [8-10].

The aim of present study is to develop a framework for further approaches of mechanical and thermal effective properties in case of particle reinforced composite materials that integrate the presence of a supplementary phase due to manufacturing conditions with these material properties.

II. Experimental Research

A. *Sample manufacturing*

Composite sample was manufactured using a self developed manufacturing technology, the phases – particles (Al particles) and the polymeric matrix (epoxy 040T resin) being chosen after several trials on different types of metallic particles (e.g. Al – 80%, SiC – 70% and Fe – 60%, 70% and 80%, etc.) and different polymeric resins (e.g. polyvinyl, other types of epoxies). The epoxy 040T (Romania) was chosen due to its good adherence at the particles surfaces. The additives used were chosen as being chemical compounds showing compatibility with the other phases and allowing polymerization process initiation and development.

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B. Experimental investigation

The non-destructive ultrasonic testing was carried out using a classical configuration base on contact principles of the transducers with the sample's surface and came from Krautkramer-Branson. Calibration was done prior the measurements. Two different measurements frequencies, 1 MHz and 2 MHz were used in the longitudinal travelling velocity of the ultrasonic wave.

The coefficient of thermal expansion was retrieved using a dilatometer DIL 402 C from NETZSCH (Germany). For each sample were used 2 successive heating stages in order to size the influence of the thermal cycling, with temperature range between 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.

III. THEORETICAL APPROACH

A two-step homogenization scheme, based on a micromechanical approach, is applied in order to retrieve the effective Young modulus and coefficient of thermal expansion of the particle reinforced polymeric composite samples considered herein. The first step takes accounts for the presence of porosities and will lead to the so called *equivalent matrix* as a consequence of homogenization between the random porosities and the polyester matrix while the second step will lead to the effective material properties considered for the composite structure and was done by a homogenization approach applied to the fillers as a second phase (e.g. ceramic and metallic particles embedded into the equivalent matrix).

A. Elastic moduli prediction

The bulk modulus and elastic longitudinal modulus of the equivalent matrix was retrieved using a Mori-Tanaka based micromechanical approach, being the theoretical model that approaches better the dilute behavior of the porosities within the composite structure. According to the model, the elastic moduli can be predicted using:

$$K_{me} = G_m \frac{4 \cdot \left(1 - V_{por}\right)}{3V_{por} + 4\frac{Gm}{Km}} \quad (1)$$

and

$$E_{me} = \frac{E_m\left(1 - V_{por}\right)}{1 + 2V_{por}} \quad (2)$$

where G_m, K_m and E_m are the shear, bulk and Young moduli of the matrix material, K_{me} and E_{me} the bulk moduli of the equivalent matrix, all in GPa, and V_{por} accounts for the porosities volume fraction.

In the second homogenization step, the classical Mori-Tanaka expressions were used to predict the effective bulk and effective shear moduli of the composite structure. According to the theoretical model, the relationships between the constitutive material mechanical properties are as follows:

$$\frac{K_c}{K_{me}} = 1 + \frac{V_p\left(K_p - K_{me}\right)}{K_{me} + \zeta\left(1 - V_p\right)\left(K_p - K_{me}\right)} \quad (3)$$

and

$$\frac{G_c}{G_{me}} = 1 + \frac{V_p\left(G_p - G_{me}\right)}{G_{me} + \xi\left(1 - V_p\right)\left(G_p - G_{me}\right)} \quad (4)$$

where

$$\zeta = \frac{1 + \nu_m}{3\left(1 - \nu_m\right)} \quad (5)$$

respectively,

$$\xi = \frac{2\left(4 - 5\nu_m\right)}{15\left(1 - \nu_m\right)} \quad (6)$$

In the previous expressions K_c and G_c stands for the effective bulk and effective shear moduli of the particle reinforced composite sample, the K_p and G_p are the bulk and shear moduli of the particles, all in GPa, ν_m is the Poisson ratio of the matrix material, V_p being the particle volume fraction.

The effective Young modulus of the composite can be retrieved indirectly using the well known general expression between the shear modulus, Young modulus and Poisson ratio:

$$G = \frac{E}{2\left(1 + \nu\right)} \quad (7)$$

B. Coefficient of thermal expansion prediction

The CTE of particle reinforced composite materials is not easy to predict precisely especially when the porosities influence has to be sized. Technical literature provides many expressions for predicting CTEs of the composites and among them the Levin models shown a good correlation with the experimental data and widely used.

The Levin model was derived by its author who believed that the effect of CTE of two-phase composites depends strongly on the overall bulk modulus of the composite. He has derived an expression for the effect of the CTE of composite as:

$$\alpha_c = \left(1 - V_p\right)\alpha_m + V_p\alpha_p + \frac{\alpha_p - \alpha_m}{\frac{1}{K_p} - \frac{1}{K_m}}\left[\frac{1}{K_c} - \left(\frac{1 - V_p}{K_m} + \frac{V_p}{K_p}\right)\right] \quad (8)$$

where α_c, α_m and α_p are the CTEs of the composite, matrix and particles, respectively, in $1/^0$K, the other material properties being as were defined previously.

The Levin theoretical model was used in both homogenization steps, firstly to retrieve the CTE of equivalent matrix and next to retrieve the effective CTE of the overall composite.

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IV. RESULTS AND DISCUSSION

Among the particle reinforced composite polymeric composites tested herein, the most interesting behavior, especially sized in its thermal property, was encountered for the 80% Al particles embedded into the epoxy matrix selected. The experimental data, as will be shown next, were influenced by the presence of porosities within the structure.

To aid the theoretical predictions different porosity volume factions were taken into account with the aim of sizing of their influence on the elastic and thermal properties – 0.5%, 1% and 2.5%, respectively.

In Fig.1 is being presented the theoretical variation with the particle volume fraction of the effective elastic longitudinal modulus, prediction made accordingly with the two-step homogenization scheme based on Mori-Tanaka theoretical model, the phases being porosities, Al particles and the epoxy matrix. As it can be seen, the variation is asymptotically increasing with the increase of the particle content and for small variations in porosities volume fraction, at a first sight there are no huge differences in the overall elastic modulus.

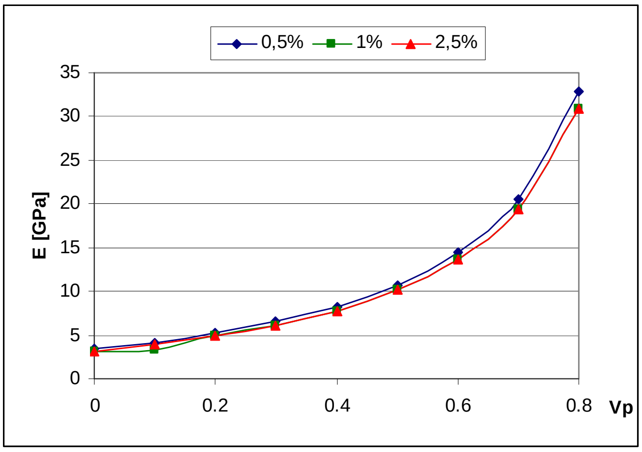

Fig. 1 Effective Young modulus variation with the particle volume fraction, different porosities volume fraction and Mori-Tanaka based prediction in the homogenization

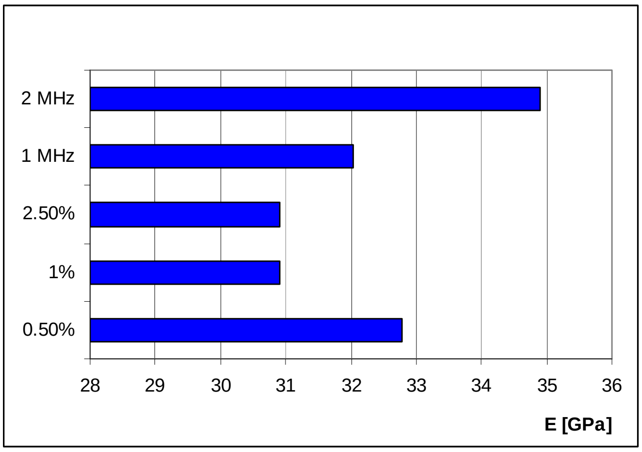

Fig. 2 Theoretical – experimental data comparison in case of the 80% Al samples, different ultrasonic frequencies and porosities content

Fig. 2 is associated to the theoretical-experimental data comparison for an 80% Al reinforced epoxy composite sample, the Young moduli being retrieved for 2 different frequencies of ultrasonic pulses. As it can be seen the theoretical predicted longitudinal moduli are strongly influenced by the porosity content, the smaller the latter (<0.5%) the higher the previous. In this case the predicted longitudinal modulus is closely to the experimentally retrieved data, the ultrasonic frequency being another factor that has to be taken into account in case of further approaches.

The CTE variation with the particle content has an opposite behavior. Fig. 3 reveals a decreasing in the CTE values with the increase of the particle content, the porosities influencing this variations in large extent only when their volume fraction are <0.5%. The variation was predicted applying the two-step procedure using the Levin theoretical model, the mechanical and thermal parameters corresponding to the 80% Al reinforced sample considered. For the other type of samples developed herein, the general trend resemble, differences relying only in the CTE values for each volume fraction.

In Fig. 4 is being shown the thermal strain and CTE variation with temperature, from 20^0 to 250^0 C, for a 70% SiC particles embedded into the selected epoxy matrix, after one thermal cycle. As it can be seen, up to 65^0 C the major influencing factor can be associated with the polymer behavior under thermal changes, actually regarding to the polymerization process that is on due. As it acknowledged, the CTE variation is linearly over a large range of temperature interval.

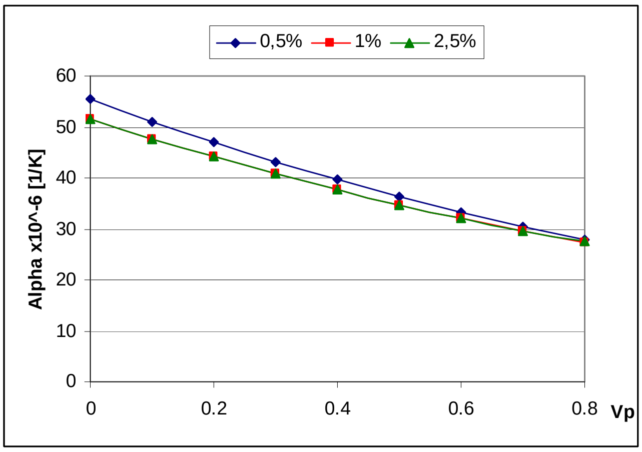

Fig. 3 Effective CTE variation with the particle volume fraction, different porosities volume fraction and Levin based prediction in the homogenization

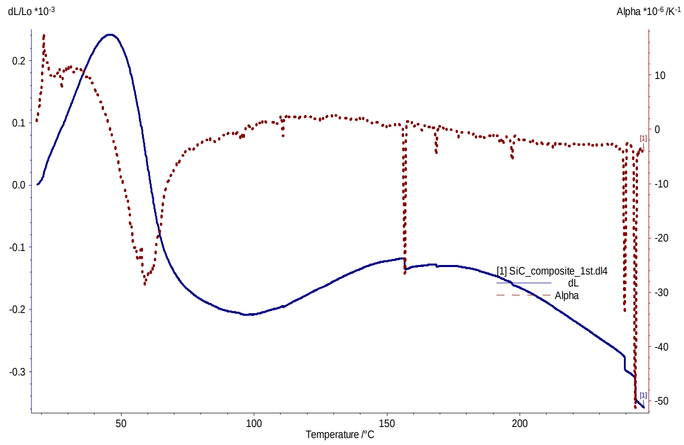

Fig. 4 Thermal strain and CTE temperature variation for a 70% SiC particle reinforced epoxy composite

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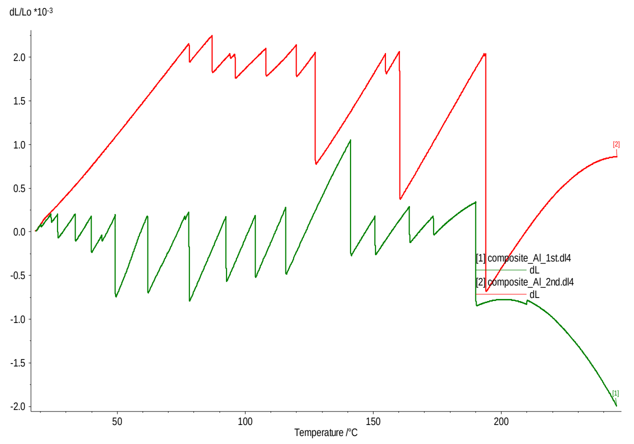

Fig. 5 Thermal strain variations with temperature for 2 different heating cycles for an 80% Al reinforced composite

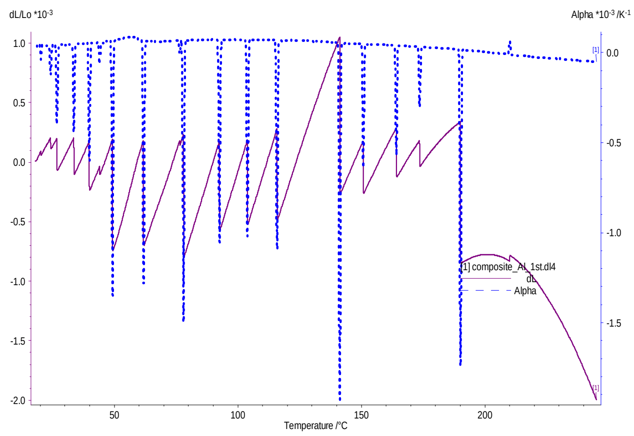

Fig. 6 Thermal strain and CTE variation with temperature for an 80% Al reinforced epoxy sample – 1 heating cycle

In Fig. 5 is being plotted the experimentally retrieved instantaneous CTE or the thermal strain field corresponding to an 80% Al reinforced epoxy composite for two different thermal cycles. As it can be seen there are differences in the thermal strain fields revealing again the polymer influence in the thermal general behavior of the composite. In the second cycle the polymerization process are considered completed, the variation being regarded mostly to the surface at the interface between phases – particles and matrix.

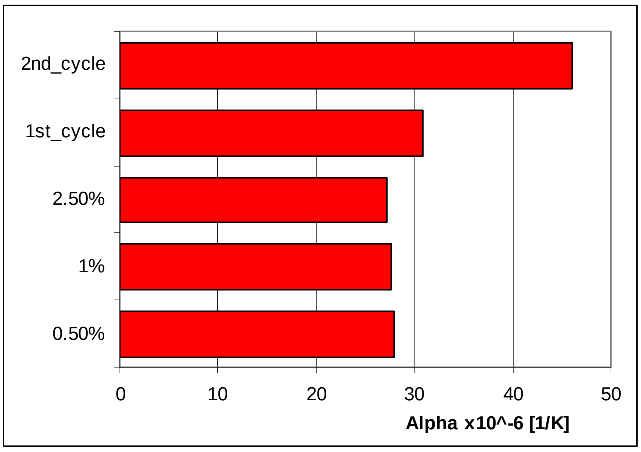

Fig. 7 Theoretical – experimental data comparison in case of the 80% Al samples, different thermal cycles and porosities

As it can be seen from the Fig. 6, the awkward variation in the CTE can be associated with the porosities within this composite sample. The CTE has an approximately linear variation over the temperature range selected, with spikes due to the gas accumulation within the internal closed cells accounting for porosity shapes.

Fig. 7 represents a theoretical and experimental data comparison for the CTE's predicted and retrieved in case of the 80% Al reinforced epoxy composite sample analyzed. As it can be seen, the Levin theoretical model developed herein for effective CTE of the particle reinforced composite, as a consequence of a two step homogenization process approaches the experimental retrieved value in case of composite's CTE. Typical curves for any particle reinforced composites, even fiber reinforced polymeric composites resemble the curve from Fig. 4.

V. CONCLUSION

The subject developed herein has a more general purpose, not only to fully characterize a particular class of polymeric reinforced composite materials, namely the two-phase particle reinforced ones, 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 or particle-fibers combinations with the aim of retrieving and predicting their mechanical, electrical or thermal properties, with or without any other external influences of extreme environment conditions. All these approaches were done within the frame of revealing the multifunctional character of these polymeric composite structures.

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