MODELLING AND CHARACTERIZING NEW MULTIPHASE POLYMERIC COMPOSITES AS MATERIALS FOR HEATING SYSTEMS

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ABSTRACT. The paper aims to present sequences from a DOE conducted approach on a self-developed and manufactured multiphase particle-fiber type polymeric composite materials with the purpose of using as alternative materials for heating system configurations. The herein approach will focus on the modelling issues and experimental research done with respect to one of the major influecing factors in engineering materials design from thermal point of view , namely the coefficient of thermal expansion (i.e. CTE). The latter is responsible for the dimensional stability of any mechanical structures and is becoming lower and constanly over a large temperature range by controlling the materials' constitutives and individual properties.

Keywords: micromechanics, heating systems, composite materials.

REZUMAT. Lucrarea prezintă o parte din rezultatele experimentale şi cele predicţionate, utilizând conceptele micromecanicii, obţinute şi vizate pentru una dintre proprietăţile de material importante în ceea ce priveşte dezvoltarea de materiale şi structuri destinate sistemelor de încălzire – coeficientul liniar de dilatare termică. Materialele compozite hibride analizate şi investigate au fost elaborate utilizând un concept şi o tehnologie proprie şi sunt de tipul particulă-fibre armate într-o matrice de material polimeric. Analiza se efectuează din punct de vedere al influenţei constituenţilor asupra acestei proprietăţi de material, abordându-se aspecte cu privire a morfologia structurilor, procentul de armare cu incluziuni şi regimul termic aplicat.

Cuvinte cheie: micromecanica, sisteme de incalzire, materilae compozite.

1. INTRODUCTION

Many applications of the emerging class of multiphase polymeric composite materials require controlled thermal expansion characteristics in order to match those of other components and low values of the property to attain a good dimensional stability. With respect to the stability issue, this can be view from two perspectives: a change in the geometrical form – materials' CTE (i.e. CTE - coefficient of thermal expansion) is playing a key role and a change in mechanical properties – a mismatch between the constitutive have a dominant effect [1]-[2], [5].

Technical literature provides numerous experimentally related papers on the effective CTE for composite materials, manufactured as metal or polymeric based matrices. Most of these papers are approaching the 2-phase combination, either with fibres or particle reinforced as inclusions of the composite structure leaving an unexplored field with respect to the multiphase composite structures [10-11]. The previous holds for the theoretical based micromechanical approaches as well, the papers focusing on different methods developed to predict the effective properties of the composite structures, at the representative volume element or representative unit cell levels, based on double or multi-inclusion homogenization schemes.

These approaches are aiming the general objective of predicting the interaction between the microstructure and the effective (overall) properties [8]-[9]. In order to design a proper multiphase composite material for the previous mentioned engineering applications, to withstand different environmental condition, a lower CTE is desired. Following this objective, an attempt has been made to study and present the CTE behaviour for self-made multiphase polymeric composite materials combinations, to identify, to size and to tailor the major influencing factors in order to address thermal management issues.

The paper aims to present a comparison approach, from theoretical and experimental perspectives, of the CTE and the influencing factors on the thermal behaviour (e.g. manufacturing conditions, material morphology, material structure etc.) in case of particle-fibre type multiphase polymeric composite samples. The samples were manufactured using self-developed technology [7]. The fillers used were different materials (e.g. metallic, ceramics) with different particle sizes and were embedded in different volume fraction, solely or in combinations into a polymeric matrix.

2. THEORETICAL APPROACHES

Homogenization techniques were widely used to simulate the mechanical behaviour of composites and multiphase materials. These were naturally extended to predict the thermal properties or thermal related dependencies of the mechanical properties as a result of the simplicity of their basic assumptions and of their accuracy to describe the overall material properties. The Mori-Tanaka homogenization scheme is one of the approaches developed based on the mean-field approximations and covers the full range of phase volume fractions, from 0 to 1. The model proved to be one of the best estimates for prediction of the effective two-phase composite materials. Literature provides numerous other theoretical models developed to aid the prediction of the overall CTE of composite materials structures, from random/aligned long fibres to short ones, and last for particle reinforced structures [3].

Herein, a specialized computer program commercially was employed to retrieve the overall CTE of particle-fibre reinforced polymeric composite materials, based on the Mori-Tanaka scheme and double inclusion method. The later can lead to the direct version and inverse version of the previous and in some cases to more accurate results and was developed by replacing the real composite with a model composite made of a fictitious reference matrix in which are embedded the inclusions coated with a material having the properties of the initial composite matrix. The RVEs (i.e. RVE – Representative Volume Element) were generated by setting a 3D random configuration of the fillers within the polymer matrix. Authors were presenting the overall CTE filler volume fraction dependencies based on other homogenization schemes in other papers along with their observations related to the „best predictor” and experimentally related comparison [4]-[7].

3. EXPERIMENTAL RESEARCH

3.1. Materials. manufacturing issues

The multiphase composite samples were manufactured as having three phases – random fibers and particles - embedded in different volume fraction into a polymeric matrix. The matrix material is commercially known as Synolite 8388 P2 from DSM Composite Resins (Switzerland), a polyester resin type. The particle inclusions considered were metallic inclusions (e.g. iron) or ceramic (e.g. alumina) mixed within the polyester resin mass in 5% and 10% volume fraction, respectively.

The 3rd phase chosen were E-glass type random fibers, commercially available under MultiStratTM Mat ES 33-0-25 trade name, from Johns Manville, SUA, mixed as having a 65% volume fraction in the overall composite volume. The reference sample was made without any particle content and used for comparison purpose.

3.2. Testing procedure and devices

The CTE measurements were performed using a DIL 420 PC differential dilatometer from NETZSCH GmbH (Germany). The particle-particle combinations of multiphase composites were shaped as cylinders of 10×25 mm^2 (width×length). For all samples the transversal external surfaces were polished to guarantee plan-parallel surfaces for precise positioning within the measuring head.

The samples are positioned horizontally on two quartz beds. The measured experimental data were sent to a PC via an USB cable, the acquisition software – Proteus Analysis (from the same manufacturer) – displaying information regarding the thermal strain variation with the imposed thermal range. Further experimental data manipulation allows linear or technical CTE retrieval vs. temperature range or time.

The temperature variation was set up having different trends in time, with 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. The thermal regimes imposed were having a linear variation up/down to/from 150^0C (heating followed by cooling).

4. RESULTS AND DISCUSSIONS

In Fig. 1 is being shown the microscopic view for the 10% ceramic & 65% E-glass fibres multiphase combination. Due to fillers size differences and dilute nature of the ceramic particles, the latter is barely distinguished within the composite mixture.

In Fig. 2 to Fig. 3 are being plotted the temperature variation of the instantaneous coefficient of thermal expansion along with the thermal strain developed during the heating within the composite structure. The thermal regime variation can be also sized in the same graphical representations.

Fig. 1. Microscopic view for the 10% ceramic & 65% E-glass fibres multiphase composite sample.

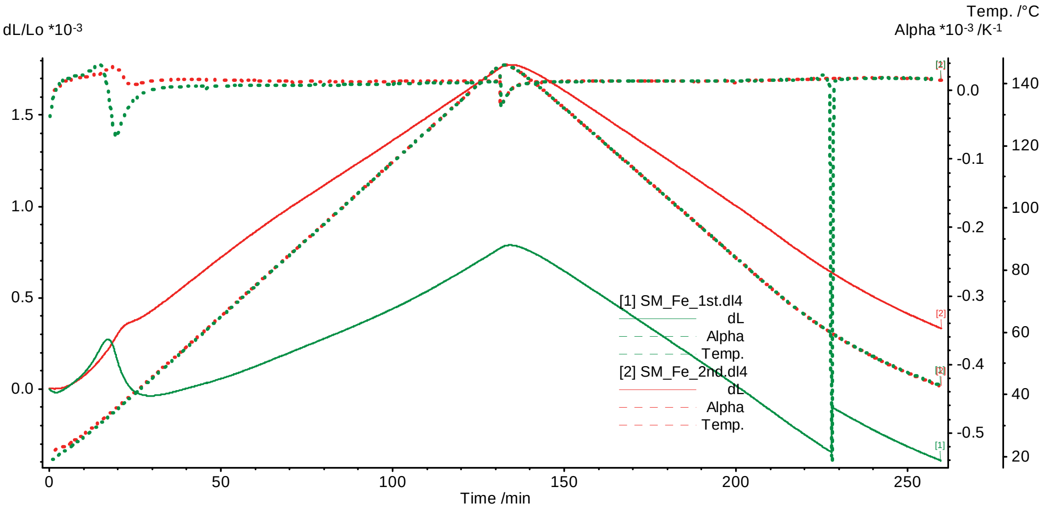

Fig. 2. Thermal strain and instantaneous coefficient of thermal expansion for the reference sample subjected to successive heating under a linear increase/decrease of temperature .

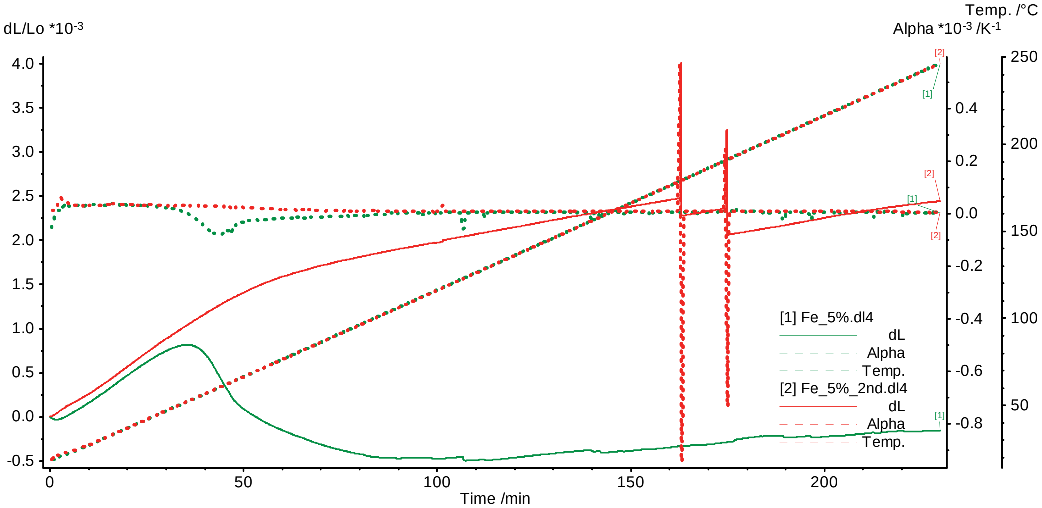

Fig. 3. Instantaneous coefficients of thermal expansion and thermal strain variation over the time for two successive thermal cycles applied to the 5% Fe & 65% E-glass multiphase composite..

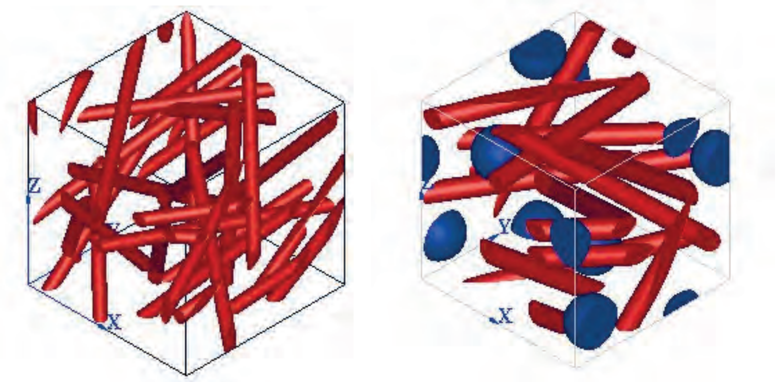

Fig. 4. Representative volume elements for random particle reinforced polymeric composite (left) and multiphase particle-particle reinforced polymeric composites (right).

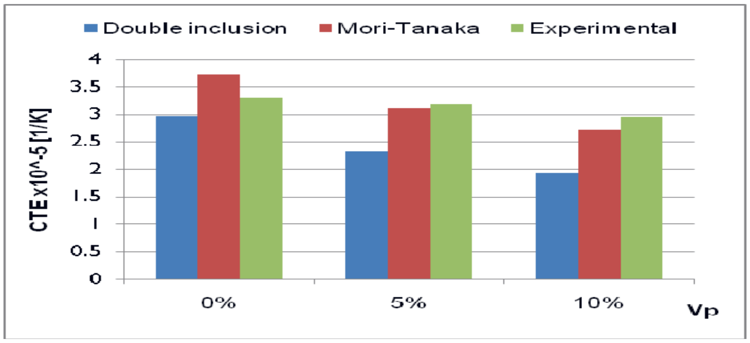

Fig. 5. Theoretical vs. experimentally recorded CTE values for the multiphase particle-fibre type polymeric composite materials (0% to 10% Fe particles, 65% E-glass particles).

The *particles volume fraction* is influencing the thermal behaviour of the overall multiphase composite structure. The higher the particle content the different vary the thermal strain fields developed within the composite structure. For all the composite samples an abrupt decrease in the instantaneous CTE variation it can be seen around the 35^{0}C in the heating step, changes that can be regarded to the polymeric matrix material whose chains it must be broken and reshaped differently. It is acknowledged from literature that increasing the particle volume fraction the overall linear CTE experience a slightly decrease. This behaviour is being used to address stability issues.

The *successive heating cycles* do have an influence on both thermal strains and instantaneous coefficient of thermal expansion of the composite materials. For all the particle-fibre multiphase composite polymeric samples the second thermal cycles lead to a smoothly variation in the instantaneous CTE over the temperature range. From *manufacturing* point of view, the first heating cycle can be viewed as a thermal treatment applied upon the composite samples, as a supplementary aid to the polymerization process.The predicted values based on the micromechanical approach were subjected to comparison with the experimental retrieved data. In Fig. 4 is being shown two RVEs corresponding to the reference composite sample (only E-glass random fibres) and for a 5% Fe & 65% E-glass fibre reinforced multiphase polymeric composite sample. The RVEs were generated using dedicated software to predict the CTE values based on the double inclusion and Mori-Tanaka homogenization concepts. In Fig. 5 were plotted the predicted values vs. the experimentally retrieved data for the multiphase particle-fibres reinforced composite materials at the reference temperature of 20^{0}C.

For all the particle-fibres multiphase composite samples in the cooling down step of the thermal regime imposed, the retrieved CTE values are higher than their counterparts from the heating step. This behaviour is somehow natural due to the fact that the temperature within the oven do not experience a suddenly temperature decrease. Also, it can be seen the fact that in the cooling step the instantaneous CTE values are almost the same over a large temperature range, denoting stability in the composites' behaviour.

This also can be sized on the peak values retrieved for all the multiphase polymeric composite samples. In the heating step of the first thermal cycle, from 20^{o}C up to 150°C, two different peaks at different temperature values are being registered whereas in the cooling down step, from 150°C to 20°C only one peak is being revealed. With respect to the second thermal regime applied a single peak is being registered for all the samples, in the cooling down step.

As it can be seen from Fig. 5 the predicted values based on the double inclusion method closely approaches the experimentally recorded values. This observation it cannot be generalized to hold for all multiphase combinations due to the lack of the experimentally values and composite structures subjected to study.

5. CONCLUSIONS

The instantaneous coefficient of linear thermal expansion in case of a multiphase polymeric composite material is not an invariant value being very sensitive to the heterogeneities and thermal regimes at which is subjected the measured sample. Its temperature dependence reflects phase changing, degree of polymerization, differences in internal structure, external environmental conditioning and the evolution of the internal thermal strain.

Inclusion type, volume fraction, size and distribution within the overall composite structure have their influences on the predicted and experimentally measured CTE values. Adding inclusions such as particles into a two phase composite material lead to a lower overall effective CTE, fact that sometimes is required in industrial applications such is case of materials for heating systems.

The herein analysis did not take into account the presence of voids or other micro-defects nor of the inclusions interaction within the multiphase composite structure even their presence or influence it can be sized in the experimentally retrieved values of the effective CTE material property.

Further studies are under development on multiphase composites made of different combinations, arrangements, particle sizes or fibre length. Different thermal regimes will be also considered as viable alternatives for developing a protocol for thermal residual stresses recovery. Supplementary, the presence of porosities, inclusions or interaction among the constitutive will be included into the modelling models. The opportunity to use recycled composite materials in order to develop thermal storage materials or alternative materials to be used as components for heating systems will be addressed in the near future.

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