

## THERMAL PROPERTIES OF SOME PARTICULATE COMPOSITES

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**Abstract:** Among composites, the polymer matrix ones are the cheapest and the easiest to form but they show major disadvantages such as poor electrical and thermal conductivity, low fire resistance etc. In the case of any composite, some of the properties may be designed, some of them may be obtained by using an appropriate forming technique and, at least, some of them may be improved by special treatments. This study is about identifying the changes in terms of thermal properties of filled polymers.

**Key words:** epoxy, clay, amyllum (starch)

### 1. INTRODUCTION

Fillers can be classified according to their source, function, composition, and/or morphology. No single classification scheme is entirely adequate due to the overlap and ambiguity of these categories. Extensive usage of particulate fillers in many commercial polymers is for the enhancement in stiffness, strength, dimensional stability, toughness, heat distortion temperature, damping, impermeability, and cost reduction, although not all of these desirable features are found in any single filled polymer. The properties of particulate-filled polymers are determined by the properties of the components, by the shape of the filler phase, by the morphology of the system, and by the polymer-filler interfacial interactions (Tsou & Waddel, 2004).

Particulate-filled polymers are widely used in many fields of application. With the exception of poly(vinyl chloride) (PVC) it is mostly technological reasons that justify the introduction of fillers, since a separate compounding step increases considerably the price of the composite. The combination of a filler with a polymer results in a new material with changed properties. Some of these changes are advantageous (improved stiffness, higher heat deflection temperature, better heat conductivity) while others are less favorable (decreased deformability and impact toughness, wear of the processing equipment) (Pukanszky, 2007).

Almost all fillers do not exist as the discrete individual particles of their primary structure. They form aggregates, secondary structures, which can agglomerate into tertiary structures in the material to be filled. An aggregate is a collection of primary particles that are chemically bonded together. The surface area of an aggregate is less than the sum of surface areas of all primary particles in that aggregate. In general, aggregates are extremely difficult to be broken down into individual primary particles by physical methods such as mechanical mixing. The union of aggregates, although weakly associated through non-bonded physical interactions, leads to a cluster. Filler materials often exist as agglomerates in their natural state. The total surface area of a cluster is similar to the sum of individual surface areas of aggregates in that cluster. The mixing and dispersion of fillers in a material involves primarily the incorporation and distribution of filler pellets or powders, and breakdown into agglomerates, and then into aggregate structures.

Polymer matrix nano-composites are a fairly new class of

engineered materials which offer for a broad range of properties, an interesting and even radical alternative to more conventional filled polymers, yet at much lower filler loadings. They can be defined as polymer matrix systems in which the dispersed inorganic reinforcing phase has at least one of its dimensions in the nanometer range, which is quite close to the scale of elementary phenomena at the molecular level. The resulting unique combination of large interfacial area and small inter-particle distance strongly influences nano-composite behavior (Lefebvre, 2004).

### 2. MATERIALS

The Epoxy system RE 4020 – DE 4020 was used as matrix to form particulate composites with Clay and Starch. Each of the fillers is used on a certain purpose connected to the final properties of the composite.

The use of Clay is recommended by its capacity to generate nanostructures inside the matrix each nano-plate of clay acting as a crystallization germ and changing, locally, the normal polymer structure with effects on mechanical properties of the final material due to the soft transfer of the loadings between the crystalline and amorphous phases (Winey & Vaia, 2007). Another aspect is about the fact that Clay presence in the pre-polymer mixture allows a better dispersion of other particles which generally tend to aggregate (such as CNT, Ferrites, carbon black).

Despite its low thermal stability the Starch might be used not only to disperse other fillers' particles but also because, as it is well known, it forms own structure when put in liquids. If a polymeric structure of starch appears inside the epoxy bulk it is possible to improve the mechanical properties of matrix material due a mechanism reinforcement like induced by the existence of starch chains (as a nano-reinforcement).

It is also well known that macroscopic properties of a particulate composite depend mostly on nature and quality of matrix-particle interface but they depend also on nature and dimensions of particles. This study was carried out using the same weight concentration of both fillers: clay powder for cosmetic use and corn starch.

Taken separately each of the fillers may be used to obtain a benefit but the problem is to use all of them to fill the same matrix. This type of filled polymer might be used as matrix for a reinforced composite (with long fibers or fabrics) such as, at least, one design problem to be solved in one layer of the hybrid composite. For instance the external layer has to show good electrical conductivity and good wear resistance, while the other properties of the structure might be set in the internal layers by using glass fibers – for strength, protected by aramide fibers for shock resistance. Using various polymers in various layers it is more possible to control the design of composite materials or composite structures. Such filled polymers might be used as films for insulating the buildings or pipes in civil engineering due to their high adherence and their low water absorption (Andrei et al. 2009).

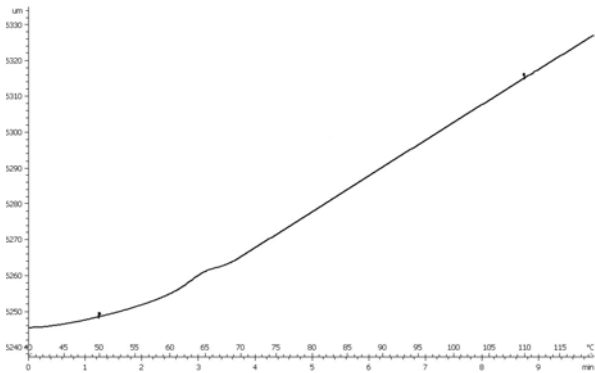


Fig. 1. Profile of dilatation curve for epoxy resin

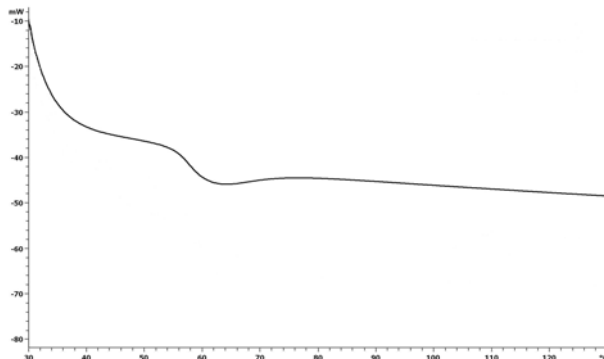


Fig. 2. Profile of heat absorption curve for epoxy resin

For this study 23 materials were formed to study thermal properties. The reference sample is Epoxy (0%) obtained by respecting all the recommendations of the producer, including the thermal cure. The plates of 150 mm x 150 mm x 4 mm were formed into glass moulds. The particulate composites were formed with the same technique but the pre-polymer was filled before molding. For both types of fillers weight ratios were set. The amounts of Clay or Starch were added in the A component of the Epoxy system (RE 4020) after mechanical dispersion for 30 minutes the right amount of polymerization agent (the B component of the Epoxy system – DE 4020) was added and the mixture was mechanically homogenized for other 15 minutes before molding (the RE 4020 – DE 4020 Epoxy system's gel time is about 45 minutes).

### 3. MEASUREMENTS AND RESULTS

Aiming to find solutions to improve epoxy's properties by filling it is obvious that all the properties of particulate composites have to be evaluated.

Coefficient of thermal expansion of each material was evaluated using TMA-SDTA 840 from *Mettler Toledo* and *Stare* dedicated software for the temperature interval of [50°C – 60°C] and [70°C – 110°C] The temperature interval (60°C – 70°C) was avoided because of a peak which appears in epoxy's case (**Fig. 1**) and which corresponds to a phase transition observable in heat absorption on DSC (**Fig. 2**). The specific heat of materials was evaluated using DSC1, also from *Mettler-Toledo* and the same software. The results are presented in **Fig. 3** for coefficient of thermal expansion and in **Fig. 4** for specific heat. Even if in the case of Starch filled epoxy, for the first temperature interval the variations are high it might be noticed that both particulate composites have almost the same variation profile connecting the results rather with the filler's concentration than its nature. The same observation is valuable for specific heat. Taking into account just the filler's concentration and not its nature is explainable the thermal dimensional stability of both particulate composites at elevated temperatures.

#### Coefficient of Thermal Expansion of Particulate Composites

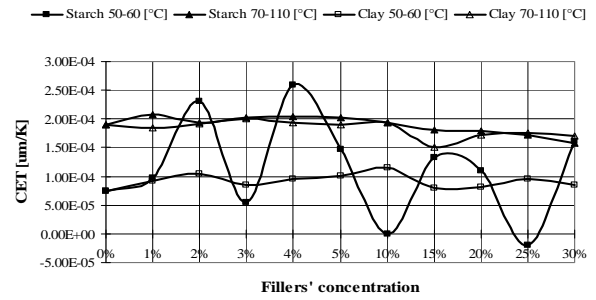


Fig. 3. Coefficient of Thermal Expansion of composites

#### Specific Heat of Particulate Composites

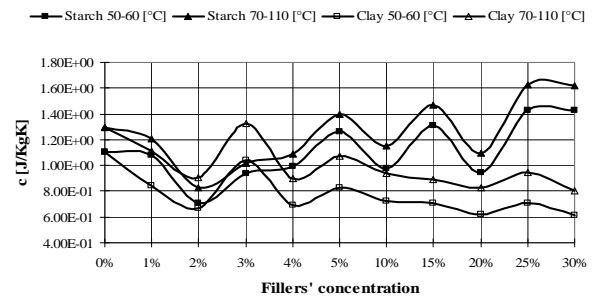


Fig. 4. Specific Heat of particulate composites

### 4. CONCLUSION

Thermal properties of particulate composites seem to depend stronger in fillers' concentration than in filler's nature. Further studies have to focus on influence of fillers' particles dimension over the thermal and other physical or mechanical properties. The presented materials are intended to be used to form stratified composites (the same matrix but various fillers in various layer) as a solution to design and control composites' properties.

### 5. ACKNOWLEDGEMENTS

The authors acknowledge CNCSIS – UEFISCSU, project number PNII – IDEI 519/2008 9/2008 and Project POSDRU /6/15/S/15, ID 6853 SIMBAD

### 6. REFERENCES

- Tsou, A.H., Waddell, W.H., (2004) Fillers, in Mark, H.F. (ed), *Encyclopedia of Polymer Science and Technology*, John Wiley & Sons, Inc., vol. 10, pp. 1 – 20, ISBN: 978-0-471-27507-7
- Pukanszky, B., (2007) Mineral-filled polymers, in Mortensen, A., *Concise Encyclopedia of Composite Materials*, Elsevier, ISBN 10: 0-08-042300-0
- Lefebvre, J.-M., (2004) Nanocomposites, Polymer-Clay, in Mark, H.F. (ed), *Encyclopedia of Polymer Science and Technology*, John Wiley & Sons, Inc., vol. 3, pp. 336 – 352, ISBN: 978-0-471-27507-7
- Winey, I. Karen, Vaia, A. R. (ed), (2007) Polymer Nanocomposites, *MRS Bulletin*, Vol. 32, No. 4, pp. 314 – 319, ISSN: 0883-7694
- Andrei, G., *et al.*, (2009), Selected mechanical properties of fabric reinforced composites with talc and carbon black filled matrix, in *Journal of Polish Society for Composite Materials*, Quaterly, 9, Nr.4/2009, 347-351 pp, ISSN 1641-8611