

ELECTRICAL AND MECHANICAL PROPERTIES OF EPOXY-CLAY NANOCOMPOSITES

Xavier Kornmann*, Per Hjalmarsson *, Andrej Krivda*

* ABB Switzerland Ltd, Corporate research, CH-5405 Baden-Dättwil,

Abstract

Epoxy-layered silicate nanocomposites were prepared using a DGEBA resin cured with methylhexahydrophthalic anhydride curing agent and an amine catalyst and the addition of up to 10 wt% of organoclay. The nanostructured morphology of the nanocomposite was put in evidence using transmission electron microscopy and X-ray diffraction. At 10 wt% of organoclay content, the percolation threshold is reached. It has a detrimental effect on both mechanical and dielectrical properties and induces a sudden increase in thermal conductivity. With the addition of only 2.5wt% of organoclay content, the fracture energy is increased by more than 230% compared with that of the pristine resin. Up to 5 wt% of organoclay content, the dielectric properties of the nanocomposites are comparable with that of the pristine resin.

1 Introduction

Nanocomposites were first investigated and reported by Toyota researchers in 1987^[1]. Using polyamide 6 they achieved to double tensile modulus and strength compared to the pristine polymer at loadings of only 2 wt% of layered silicates. In addition the heat distortion temperature was elevated to almost 100 °C, enabling the polymer to be used in new areas such as under hood parts in vehicles. This novel advancement paved the way for other researchers and nanocomposites have been extensively studied over the past decade. Further achievements have included increased barrier properties [2],[3] increased flame retardancy [4] and improved thermal and mechanical properties [5],[6],[7]. To date however limited number of investigations has been made on applicability of these new materials to electrical insulation purposes even

though epoxies are widely used in this field as potting material.

2. Experimental

The organoclay was swollen in the DGEBA epoxy at 80°C prior to the addition of the methylhexahydrophthalic anhydride curing agent and amine catalyst. The mixture was cured 4 hours at 120°C. The nanostructure of the resulting nanocomposites was analyzed by XRD and TEM.

A Siemens D5000 X-ray diffractometer (20kV, 5 mA, Anode: Cu) was employed to determine the nanostructure of the different materials. Samples from 1 mm thin plates were cut to fit a circular holder and rinsed with ethanol. 2θ was set to range from 2.5 to 10 ° when analyzing the pristine clay and from 1 to 10 ° during the screening phase. When examining the samples more thoroughly in the later stage of experiments the scan ranged from 1 to 25 °. The step size was set to 0.020 ° and step time to 15 seconds. Raw data was retrieved by an evaluation software program (EVA).

TEM specimens were cut to a thickness of about 40 nm with an ultramicrotome Reichnert & Jung Ultracut E using a diamond knife. The specimens were collected on a through of water and positioned on a copper grid. The transmission electron micrographs were taken with a LEO 912 apparatus at an acceleration voltage of 12 kV. The interlayer spacing was measured using software program called ESI-Vision (SIS). Tensile and fracture properties as well as dielectric properties measurements were also carried out.

Tensile specimens were cast with a thickness and depth in the middle cross section of 4 and 10 mm respectively (NORM = DIN EN 1B) and were tested at a speed of 2mm/min. Fracture toughness was measured with the double torsion test with a testing speed of 0.5 mm/min.. The samples were cut from 4mm plates and had a width and length of 34 and 80 mm respectively.

Circular samples with a diameter of 50 mm were cut with water-jet for the evaluation of thermal conductivity. The samples were placed in a Holometrix TCA and the sample thickness was accurately measured before analysis. Thermal conductivity was then measured at 70°C, 80°C, 90°C, 100°C, 120°C.

Square shaped samples with a dimension of 35*35 mm were cut with a saw from 1 mm plates for dielectric measurements. After deposition of Chromium/Gold electrodes, all samples were tested with a Novocontrol high resolution dielectric analyzer with a frequency range of 1 to $1 \cdot 10^6$ Hz and for the temperatures; 25, 40, 60, 80, 100, 120, 140 °C. $\tan \delta$ and permittivity were retrieved from the measurements.

3. Results & Discussion

3.1 Morphology

Fig 1 shows a TEM picture of an epoxy-clay nanocomposite with 5 wt% organoclay. The silicate layers are partially exfoliated with an average interlamellar distance 5.5 nm. This value was confirmed by XRD measurements.

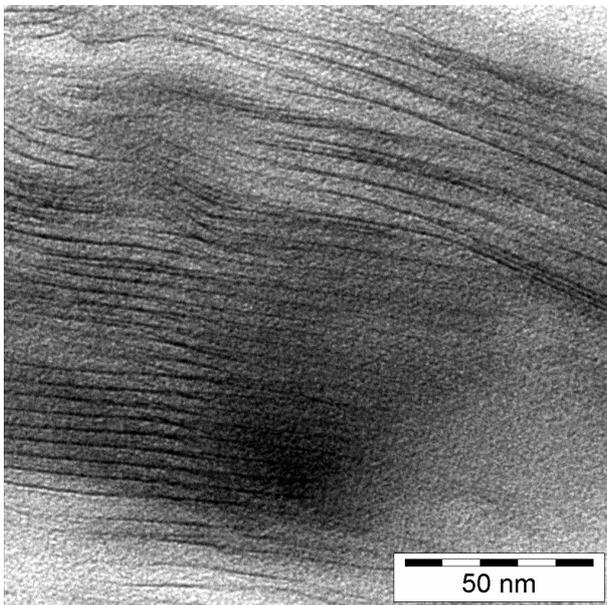


Fig. 1. Transmission electron microscopy of epoxy-clay nanocomposites

The pyrolysis of the nanocomposites revealed that at 10 wt% of organoclay content, the layers occupy the full volume suggesting that percolation is already reached at this nanofiller level.



Fig. 2. Nanocomposites after pyrolysis containing 2.5, 5, and 10 wt% of organoclay.

3.2 Mechanical properties

Table 1 presents the tensile properties of the pristine epoxy and of nanocomposites containing various amount of organoclay.

Table 1. Tensile properties of the pristine epoxy and of nanocomposites containing 2.5, 5, and 10 wt% of organoclay.

Material	σ_B	E	ϵ
SI Units	MPa	MPa	%
Epoxy	55.8 ± 9.5	3002 ± 119	1.78 ± 0.37
2.5 wt% clay	44.9 ± 3.9	3058 ± 119	1.58 ± 0.12
5 wt% clay	48.9 ± 3.4	3205 ± 120	1.60 ± 0.14
10 wt% clay	34.6 ± 4.3	4100 ± 387	0.85 ± 0.08

Tensile strength σ_B and elongation at break ϵ decrease slightly up to 5 wt% of organoclay content but are greatly affected at 10 wt% organoclay content. This is probably link to the fact that the percolation threshold is reached at 10 wt% organoclay content.

The reinforcing effect of the organoclay on the Young's modulus E of the nanocomposite is only clearly visible at an organoclay content of 10 wt% with a modulus increase of ca. 36%. Again, this is probably related to the fact that the percolation threshold has been reached.

Fig. 3. presents the fracture energy of epoxy-clay nanocomposites as a function of organoclay content. Interestingly, the facture energy is increased by ca. 230% at an organoclay content between 2.5 to 5 wt% compare to the pristine epoxy. However, at higher organoclay content (i.e. 10 wt%), the fracture energy decreases significantly, suggesting that

percolation of the clay layers has a negative effect on fracture properties.

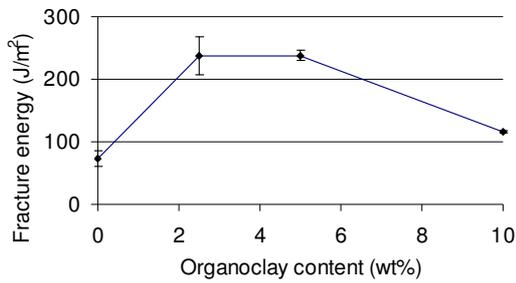


Fig. 3. Fracture energy of epoxy-clay nanocomposites as a function of organoclay content

3.2 Electrical properties

Fig. 4. presents the dielectric properties of epoxy-clay nanocomposites at different organoclay content.

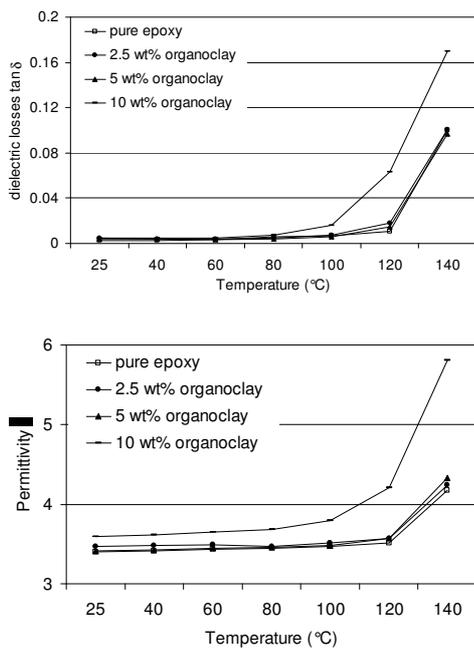


Fig. 4. Dielectric properties of epoxy-clay nanocomposites: permittivity (top) and dielectric losses (bottom)

Up to 5 wt% of organoclay content, the presence of the clay appears to have a limited influence on the dielectric properties. However, higher organoclay content has a detrimental effect on the dielectric properties from an electrical insulation point of view. Both the permittivity and the dielectric losses increase significantly especially above 100°C. Here again, the percolation threshold reached at 10wt%

organoclay appears to have a negative effect on the dielectric properties of the epoxy from an electrical insulation point of view.

Fig. 5 shows the increase of thermal conductivity as a function of the organoclay content. The sudden increase of thermal conductivity observed at 10wt% organoclay suggests that percolation has been reached. Indeed, it is likely that the silicate layers form a continuous network in the epoxy so that heat can be more easily dissipated through the nanocomposite.

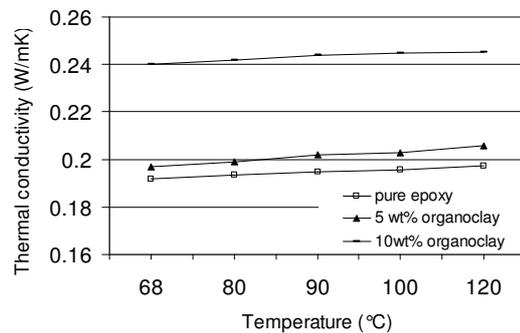


Fig. 5. Thermal conductivity measurement of epoxy nanocomposite at different organoclay content.

4. Conclusions

Epoxy-clay nanocomposites present greatly improved fracture toughness at low clay content (5 wt% and below) without affecting the dielectric properties of the resin which are important for electrical insulation. Addition of 10 wt% of organoclay has detrimental effects both on the mechanical and the electrical properties of the resulting nanocomposites. This is probably due to the fact that 10wt% of organoclay are sufficient to reach the percolation threshold.

5. References

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