For participation in ISyDMA'5 : Virtual Conference 15-17 April 2020

ISyDMA

Investigating the electrical transport mechanisms of pentafunctional epoxy resin/natural phosphate composites

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Abstract: An investigation on electrical transport mechanisms of composites materials based on novel pentafunctional epoxy resin matrix reinforced with different concentrations of natural phosphate (NPh) particles is presented. The electrical properties were measured in the frequency domain 100 Hz - 1 MHz and temperature range from 200 to 400 K. The frequency-dependence of the electrical conductivity is analyzed using the Jonscher power law and the Nyquist representation of the electrical modulus spectra is modeled using the Cole-Cole model. The calculated relaxation parameters show that, with the increasing of the NPh particle concentrations, the composite becomes more heterogeneous. Furthermore, the relaxation time and the DC electrical conductivity were analyzed using the Arrhenius equation.

Composites preparation

The pentafunctional epoxy resin matrix namely pentaglycidyl ether pentabisphenol A of phosphorus (PGEPBAP) was synthesized in two steps according to Hsissou et al. [1].
The composition of natural phosphate (NPh) particles are $Ca_5(PO_4)_3(OH)$, $Ca_5(PO_4)_3F$ and $Ca_5(PO_4)_3Cl$ with average grain size 0.06 µm.
Crosslinking reaction of the PGEPBAP epoxy resin in the presence of methylene dianiline as a hardener and NPh particles at foor concentrations (0 %, 5 %, 10 % and 20 %) to get a (PGEPBAP/MDA/NPh) composites [2].

Electrical measurements

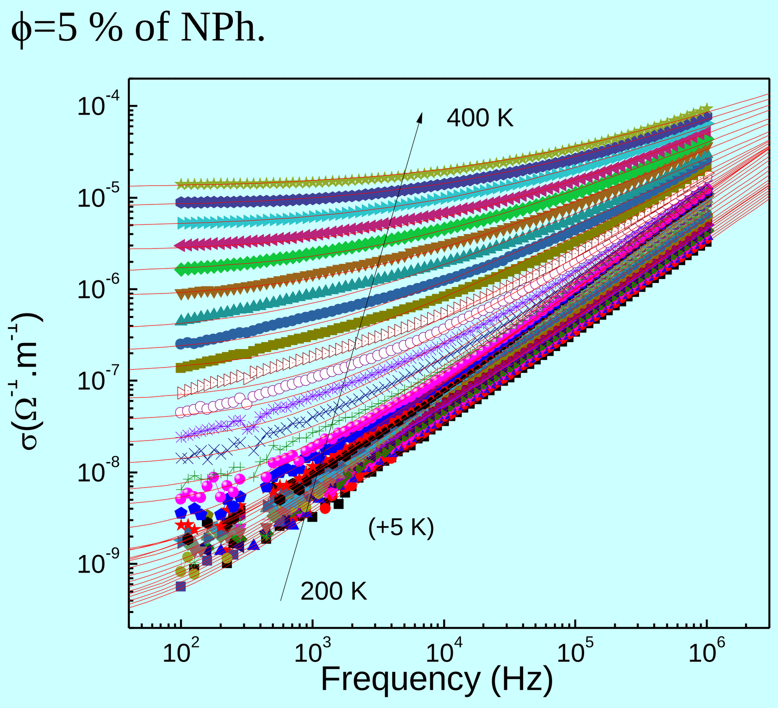
The samples were prepared as discs of thickness about 1 mm, with aluminium electrodes of 10 mm diameter on the opposite sites of the samples. The electrical contacts were formed by silver paint. Electrical measurements were performed using an Agilent 4294A Impedance Analyzer. The measured impedance is equivalent to a capacitor C_p in parallel with a resistance R_p. The electrical conductivity, σ, and both real, M', and imaginary, M'', parts of the complex modulus were deduced from the measured values of C_p and R_p using the following equations:

$$\sigma = \frac{d}{A}\frac{1}{R_p} \quad ; \quad M' = \frac{A}{d}\frac{\varepsilon_o C_p (\omega R_p)^2}{1+(\omega R_p C_p)^2} \quad \text{and} \quad M'' = \frac{A}{d}\frac{\varepsilon_o R_p \omega}{1+(\omega R_p C_p)^2}$$

where *d* and *A* are the sample thickness and electrode surface area, respectively.

Electrical conductivity analysis

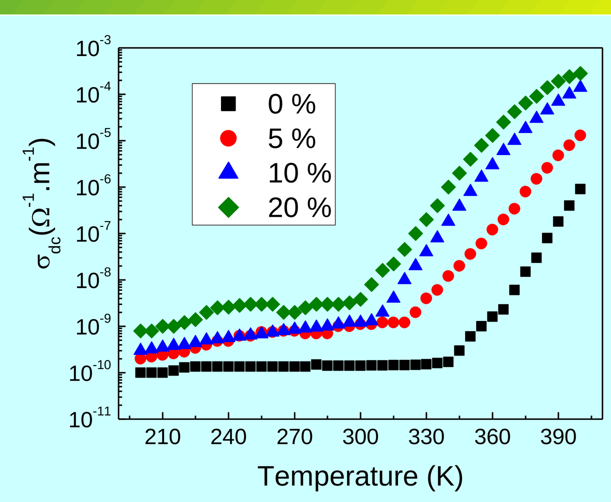
The electrical conducivity was analysed using the Jonscher power law. Figure below shows the case of composites with ϕ=5 % of NPh.

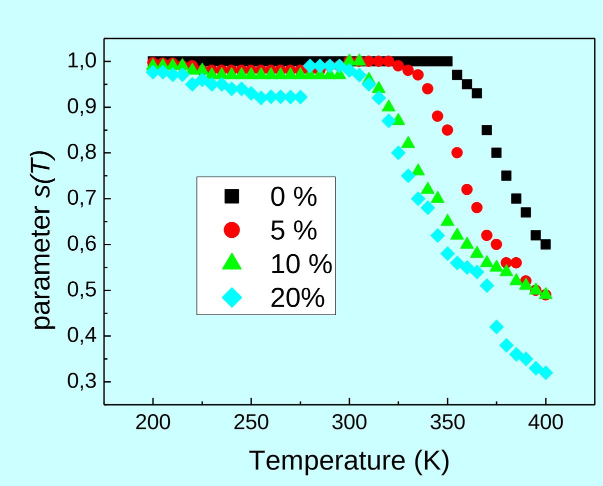


Frequency dependence of electrical conductivity of composite with ϕ=5 %, at different temperatures.

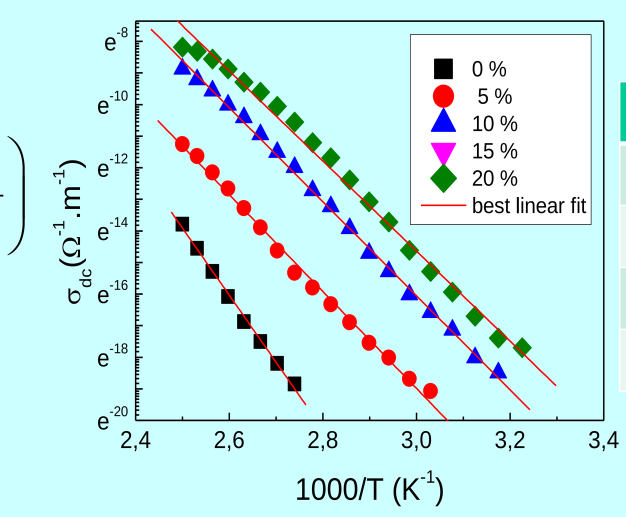
$$\sigma(T) = \sigma_{dc}\left(1 + k\left(\frac{\omega}{\omega_c}\right)^{s(T)}\right)$$

σ_{dc} is the DC electrical conductivity
ω_c is the crossover frequency
s is a parameter between 0 and 1





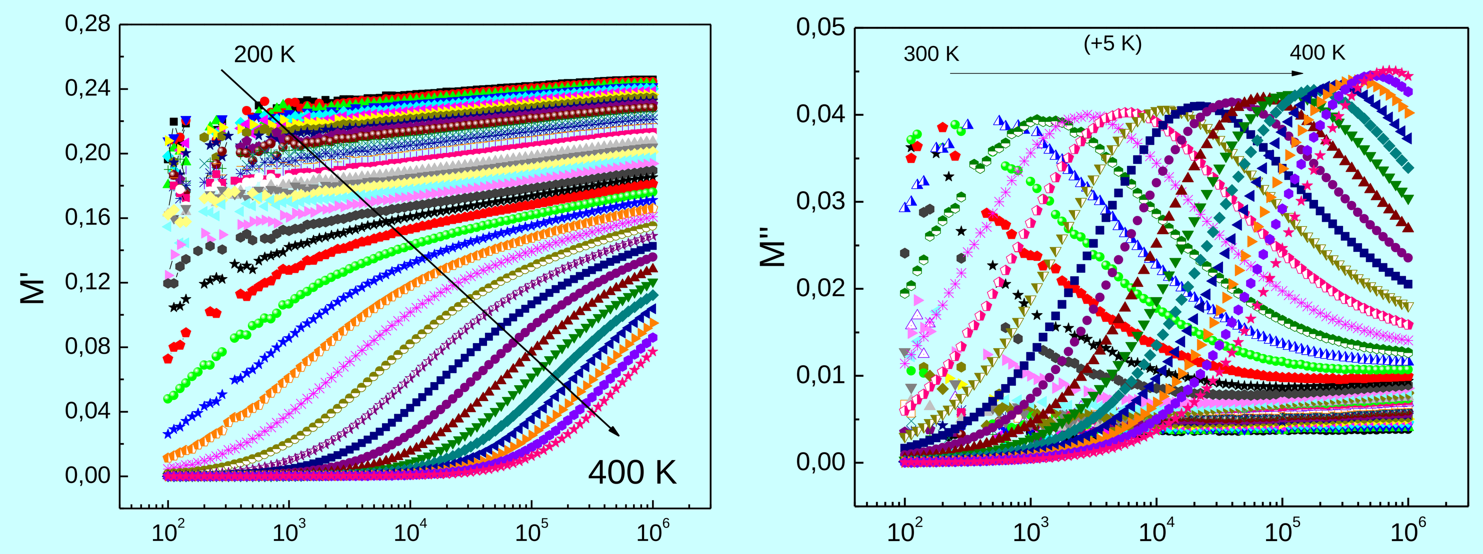
$$\sigma_{dc}(T) = \sigma_o \exp\left(\frac{-E_a}{k_b T}\right)$$



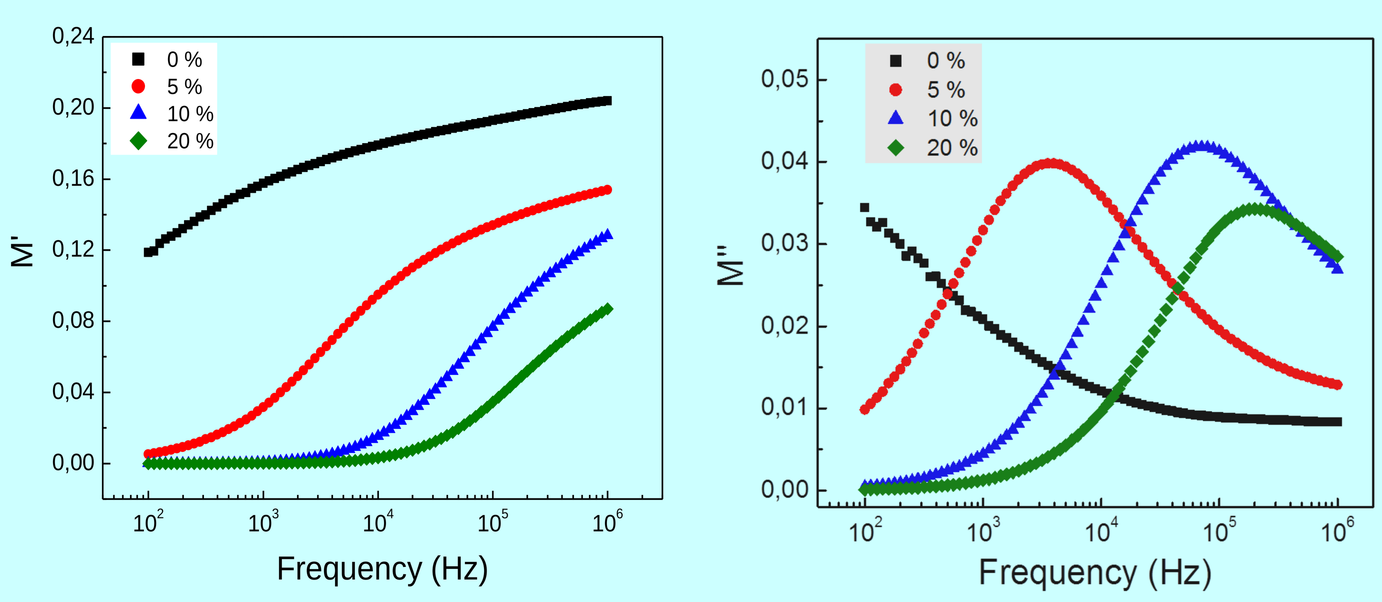
Φ(%)	E_a (eV)	ΔE_a (eV)
0	1.82	0.04
5	1.31	0.02
10	1.27	0.01
20	1.22	0.02

- s(T) equal to 1 in the low temperature domain, showing a capacitive behavior of composite, and a linear relationship at high temperature domain.
- σ_{dc} increases with temperature, showing the thermal behavior of the prepared composites
- E_a decreases as filler concentration increases, this behavior may be due to an increase of polarization energy leading to the decrease of the domain boundary potential of NPh into the polymer matrix.

Electrical modulus analysis



Frequency dependence of the real, M', and imaginary , M'', parts of the electrical modulus of the composite (ϕ=10 %) at different temperatures.



Frequency dependence of the real, M', and imaginary , M'', parts of the electrical modulus of neat matrix and with all concentration of phosphate particules, at T=370 K.

=> The values of M' tends to zero at low frequency, indicating that the electrode polarization has a negligeably contribution.

⇒The relaxations peaks shifts to higher frequency as temperature increases, showing thermal behavior of these composites.

Conclusion

- Analysis of the electrical conductivity and dielectric response of composites materials based on novel pentafunctional epoxy resin matrix reinforced with different concentrations of natural phosphate (NPh) particles has been presented.
- The frequency-dependence of the electrical conductivity is analyzed using the Jonscher power law and the DC electrical conductivity using the Arrhenius equation.
- The frequency dependence of the electrical modulus revealed a relaxation peak, showing thermal behavior and indicating that the composites becomes more heterogeneous as the concentration of loading filler increases.

Acknowledgments

This work was conducted under collaboration between Ibn Tofail University (Morocco) and University of Aveiro (Portugal).

References

[1] R. Hsissou *et al.* Polymer Bulletin 76 (2019) 4859
[2] R. Hsissou *et al*. SN Applied Sciences 1 (2019) 869