Dielectric properties of Graphite/Carbon nanotubes/Polyester ternary composites

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Abstract

Ternary composites composed of a polyester matrix were synthesized with the combined inclusion of two types of fillers, namely, multi-walled carbon nanotubes and graphite. Electrical measurements were carried out in a frequency range from 100 Hz to 1 MHz and temperatures from -33 to 107 ºC. Temperature effect on dielectric permittivity showed a specific change at glass transition temperature. Two dielectric relaxations were identified using the Nyquist representation of electric modulus which were analyzed using the Cole-Cole model. Furthermore, the analysis of the temperature dependence of the relaxation frequency, using the Arrhenius equation, showed the existence of filler/matrix interaction.

Keywords: Ternary composite, polyester, carbon nanotube, graphite, Cole-Cole model.

Materials and samples preparation

In this study, we used as a matrix an unsaturated polyester resin 154TB, containing 31 wt% of styrene monomer, requiring 30 min for gelation at room temperature (Cray Valley/Total, USA). The two conducting fillers are: Natural graphite flakes (CRPP Center, Bordeaux University, France) with grain size of 20-100 µm, Density of 2.09-2.26 and electrical resistivity is about 0.1 to 10 ohms/square. Multiwalled carbon nanotubes (CNT) (Cheap-Tubes, USA), with average diameter is about 50 nm, length in the range from 10 to 20 µm, and purity above 95 wt%. The samples of composites were prepared with the 8% contents of CNT/Gt/Polyester. CNT/Gt were mixed with the polyester in different concentrations and stirred at room temperature. The CNT/Gt were first dispersed in methanol solution under magnetic agitation to reduce the maximum size of the aggregates. After complete evaporation of methanol, the obtained CNT/Gt powder was then directly added to the polyester. Finally, it was injected into sample moulds. After achieving gelation, the samples were unmolded after a few hours, then they were left in rest in order to reach the complete polymerization.

Results and discussion

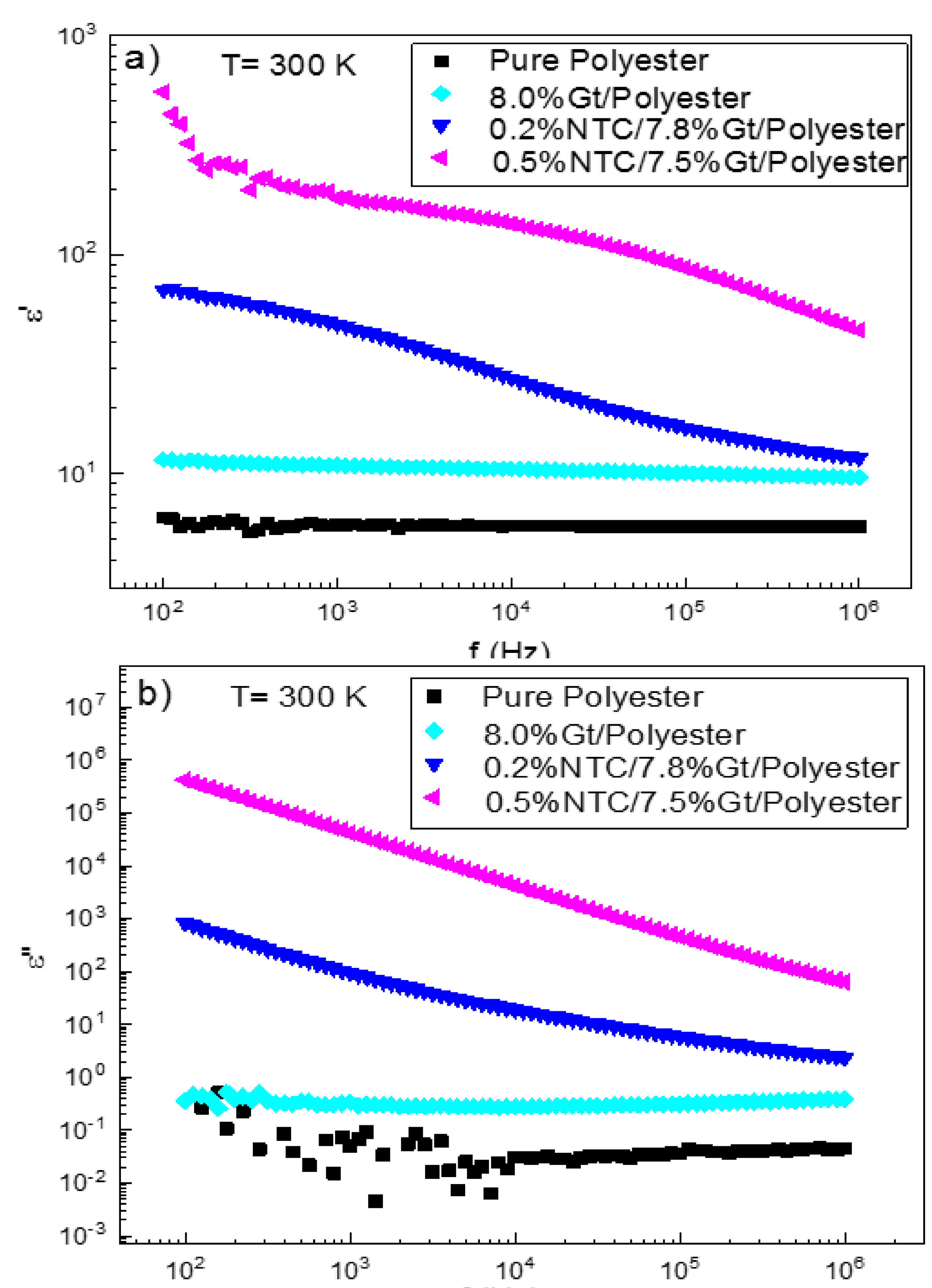

Fig. 1. Frequency dependence of dielectric permittivity (a) real part, and (b) imaginary part of Pure Polyester, Gt/Polyester and NTC/Gt/Polyester composites at room temperature.

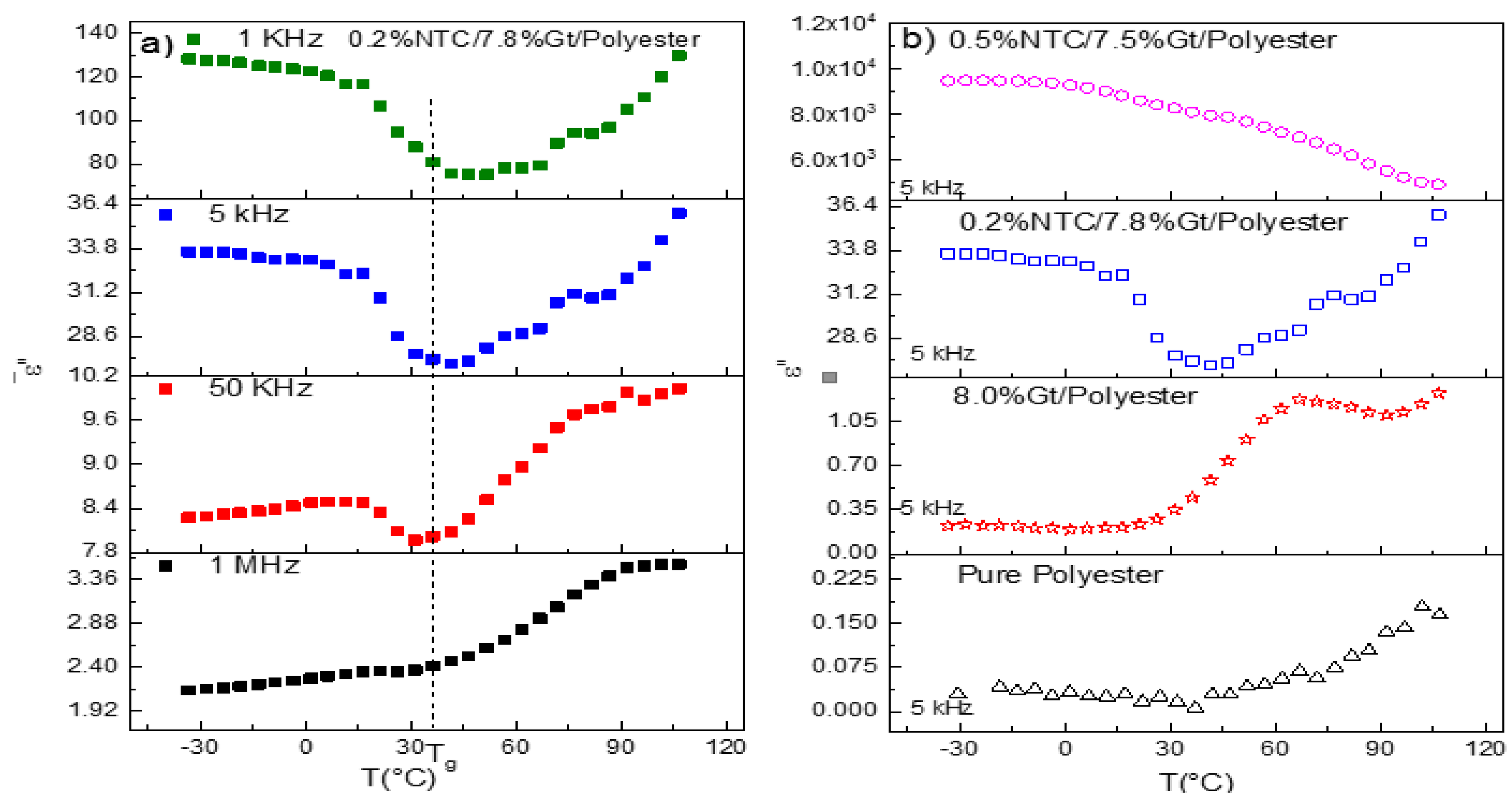

Fig. 2: Temperature dependence of imaginary part of dielectric permittivity: (a) for 0.2%CNT/7.8Gt/Polyester at different values of frequency, (b) comparative curves of each sample at 5 kHz.

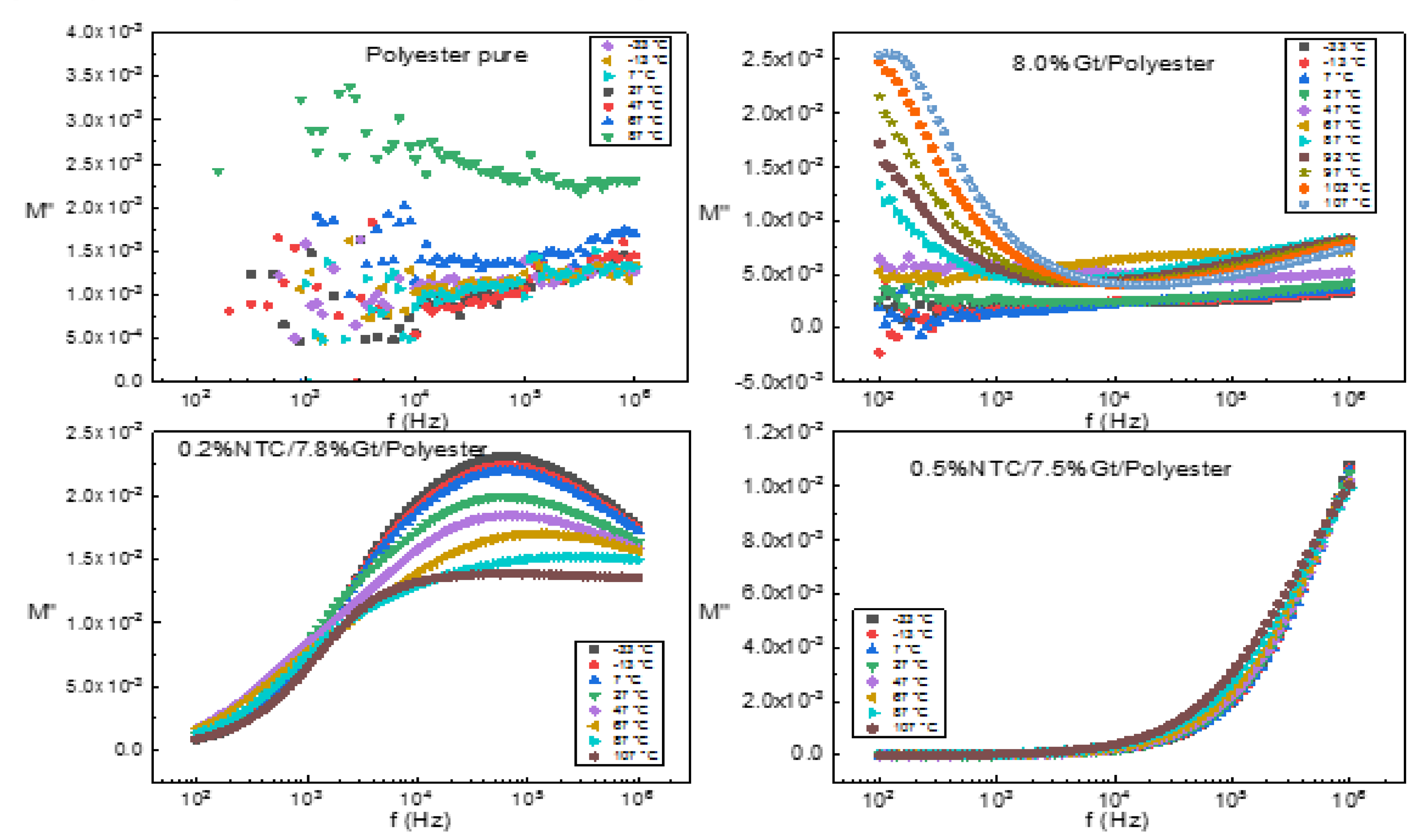

Fig. 3: Imaginary part of the electric modulus, M'', as a function of frequency for pure Polyester, GT/Polyester and CNT/GT/Polyester composites at different temperatures.

Cole-Cole model

$$M^*(\omega) = M_\infty + \frac{\Delta M}{1+(i\omega\tau)^{1-\alpha}}$$

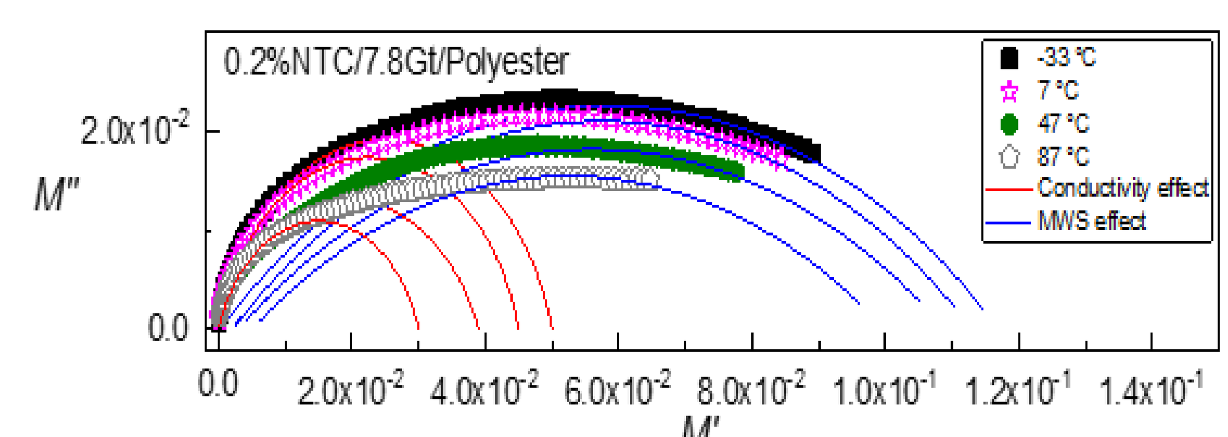

Fig. 4. Nyquist plot of the complex electric modulus of 0.2%CNT/7.8%Gt/Polyester composite. Solid lines are fits to the Cole-Cole model.

Arrhenius equation

$$f_r(T) = f_o \exp\left(-\frac{E_a}{k_b T}\right)$$

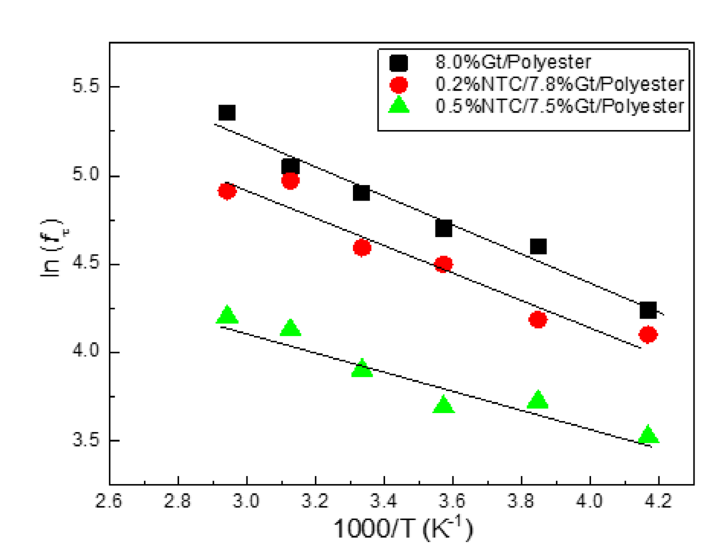

Fig. 5: ln (fr) vs. 1000/T for Gt/Polyester and CNT/Gt/Polyester composites. Solid lines are the best linear fits to the Arrhenius equation.

Table. 1. Values of the activation energy obtained using the Arrhenius equation.

ϕ (%)	E_a (meV)
8.0%Gt/Polyester	71.55
0.2%CNT/7.8%Gt/Polyester	65.50
0.5%CNT/7.5%Gt/Polyester	47.90

Conclusion

- The dispersion of the dielectric permittivity may be due to the fact that the dipole polarization and interfacial polarization within the composites cannot keep up with changes in the applied electric field frequency as the frequency increases, which resulted in the reduction of dielectric permittivity.
- The Cole-Cole model presents a very good adjustment for the experimental data of real and imaginary parts of the complex electric modulus.
- The obtained activation energies decrease with the volume fraction of CNT fraction. This decrease may be due to an increase of polarization energy and/or charge carrier density leading to decrease of the domain boundary potential of CNT into the polyester matrix.