Abstract

The purpose of this thesis is the detailed study of the structure and the properties of certain materials. To be more specified, we study the thermal transitions (e.g. glass transition), the dielectric relaxation mechanisms (main and secondary) and the influence of the material's composition in general, on the molecular mobility of polymer/clay nanocomposites. The nanocomposites include the polymer matrix which is Polyurethane with the addition of montmorillonite (MMT).

Two series of nanocomposite materials were studied on the basis of ionic oligourethaneacrylate and modified montmorillonite (MMT). The first series contains one sample which is the matrix and three more that all have montmorillonite concentration 2.5% and have different types of montmorillonite modification. The second series contains one sample which is the matrix and three more with increasing concentration of montmorillonite (1.5% - 3.5%). Three experimental techniques were employed, in order to study the samples. These were: Differential Scanning Calorimetry (DSC), Thermally Stimulated Depolarization Currents (TSDC) and Dielectric Relaxation Spectroscopy (DRS). All measurements were carried out to the Dielectric Spectroscopy lab in the National Technical University of Athens. Some experiments were also carried out with the addition of water to the samples in order to study the amount of water absorbed under certain temperature and humidity levels. In addition the diffusion parameter was also calculated.

At first, the glass transition temperature T_g was calculated using all three experimental techniques DSC, TSDC and DRS. In general, concerning the first series of samples, an increase on the glass transition temperature was observed regarding the sample with the most powerful type of montmorillonite modification. Concerning the second series of samples, an increase on the glass transition temperature was observed as the concentration of montmorillonite was increased. The dielectric relaxation mechanisms were thoroughly studied during the dielectric measurements and as a result these materials could be characterized as highly homogeneous systems, as far as thermal transitions and dielectric relaxation mechanisms are concerned.

As regards to the first series of samples, the dielectric experiments resulted in a series of conclusions: The matrix sample has the higher mobility in contrast with the mobility of the samples that have the other types of montmorillonite modification. The sample with the most powerful type of montmorillonite modification has the lowest conductivity in contrast with the other samples. The concentration of the charge carriers is higher regarding the samples with higher conductivity and as a result they affect the amount of conductivity.

As regards to the second series of samples, the dielectric experiments resulted in a series of conclusions: The matrix sample has the higher mobility in contrast with the mobility of the samples that have higher concentrations of montmorillonite. The mobility of the sample with montmorillonite concentration 2.5%, is really low and it is assumed that the morphology of exfoliation is obtained. The low mobility that is observed for the samples with higher concentrations of montmorillonite, leads to the conclusion that we have improved mechanical properties. This conclusion is confirmed by the elasticity values provided. The sample with the highest concentration of montmorillonite has the lowest conductivity.

Moreover, the experiments concerning the addition of water to the samples, resulted to the conclusion that the water concentration of the samples is not modified systematically with the concentration of montmorillonite. In addition, the diffusion

parameter was also calculated and significant alterations in the values of the diffusion parameter were observed from sample to sample (20%-25%). A correlation between the diffusion parameter and the results of the conductivity was expected but not observed. A possible explanation is the probability of exfoliation morphology of the samples that have low values of the diffusion parameter. Concerning the other samples, there is a need for further investigation, of the nanocomposite's molecular mobility and morphology in order to result to some certain conclusions.