– *Abstract* –

In this thesis two main concepts were used for the synthesis of polybenzoxazine/graphene oxide (GO) nanocomposites. Firstly, polybenzoxazine nanocomposites containing 1% wt. of different GO structures functionalized with organic groups were prepared using the solvent dispersion method. It was shown that the nature of organic functionalities from the surface of graphene sheets has a major role in the polymerization process of the monomer. From the DSC results on the synthesized materials, a decrease in the polymerization temperature was observed for the BA-a monomer from 241.5ºC to 237.7ºC in case of the nanocomposites containing graphene oxide with numerous oxidized groups (GO) and up to 239ºC for the nanocomposites containing graphene oxide with carboxyl groups (GO-COOH). In the case of the use of rGO-NH_2 and rGO-NH, no major contributions were observed in accelerating the polymerization, but it was shown that the amine groups are consumed during the reaction with the BA-a monomer, contributing to the crosslinking of polybenzoxazine materials. Secondly, the covalent functionalization method was used by developing new strategies to directly synthesize the benzoxazine monomers onto different GO layers. GO-COOH, aminated reduced graphene oxide (rGO) and GO were used as main reagents for the benzoxazine formation and the successful formation of the hybrid monomers was proved by FT-IR, XPS and ^{1}H-NMR analyses. Moreover, the final materials showed improved thermal stability and the polymerization temperature was reduced with ~10°C for GO functionalized with poly(amidoamines) and benzoxazine structures. The Raman spectrometry, XRD analysis and TEM images of the final polybenzoxazine/GO nanocomposites were employed for better understanding of the exfoliation and polymerization process occurring for each type of benzoxazine-functionalized GO synthesized material and the final balance between *in-graphene polymerization* and *out-graphene polymerization* that significantly influences the mechanical properties of the final materials is discussed.