## Abstract

Novel polyurethane (PU) nanocomposites based on toluene diisocyanate, poly(propylene glycol), various hyperbranched polymers (HBPs) and different nanoparticles were synthesized and characterized. The effect of the functionality of HBP and the loading of nanoparticles on the structure, mechanical, dynamic mechanical, thermal, adhesion and gas barrier properties of polyurethane and its nanocomposites was also determined. Structure-property relationship of these nanocomposites was established. With an increase of the number of functional groups of the hyperbranched polyol, the tensile strength and the modulus increased. Dynamic mechanical thermal analysis also suggested an increase of the storage modulus (E') and a decrease of peak tan  $\delta$ , when the number of functional groups increased. It was in line with phase morphology of cured polyurethane showing a decrease of size of nanophase with an increase of branching in hyperbranched polyol. Crosslinking kinetics of polyurethane from its pre-polymer stage with three hyperbranched polyol was studied using FTIR spectroscopy. The activation energy was lowest with fourth generation hyperbranched polyol. The microstructure of the novel nanocomposites was investigated by X-ray diffraction analysis, high-resolution transmission electron microscopy and atomic force microscopy. It was found that good dispersion was obtained up to 8 phr nanoclay and 4 phr of nanosilica loading. ~100% increment in the tensile strength,  $\sim$  2-fold increase in the lap shear strength,  $\sim$  200% increment in the peel strength, and 120% increment in the storage modulus along with a dramatic improvement in thermal stability were observed with the addition of 8 phr clay, over the pristine polyurethane. The presence of small concentration of nanoclay (Cloisite 30B) platelets in the highly crosslinked PU reduced the helium penetration rate to a great extent by creating torturous paths for gas molecules. There was ~76% decrease in permeability for 8 phr Cloisite 30B filled PU over the control pristine 3rd generation PU. In similar fashion, with the addition of nanosilica loading up to 4 phr, tensile strength and storage modulus at 25 °C increased by 52% and 40% respectively over the pristine polyurethane. Organo-treated nanosilica exhibited higher physico-mechanical properties than the untreated one. Further, the thermo-mechanical properties of hyperbranched polyurethane and its nanocomposites were compared with those of linear polyurethane and its nanocomposites.

Key words: polyurethane, nanocomposites, hyperbranched polyol