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NEW DEVELOPMENTS IN HARD-PHASE REINFORCED ELASTOMERS: A STUDY OF COPOLYURETHANES DERIVED FROM MIXTURES OF DIISOCYANATES

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New insight has been gained into structure-property relations in thermoplastic polyurethane elastomers (TPUs) by preparing them using mixtures of isocyanates. The role of hard segment structure on the polyurethanes properties was investigated, when the phase segregation is no more perturbed by any potential crosslinking. A family of TPUs was synthesized with ethylene glycol (EG) as chain extender. Two isocyanates were employed: 4,4-diphenyl methane diisocyanate (MDI), and 4,4'-dibenzyl diisocyanate (DBDI), alone or as mixtures. Materials derived from DBDI are based on hard segments of variable geometry. In such polymers it observed a remarkable tendency of the EG-DBDI couple to crystallize by self-association. Materials were characterized by WAXS, DMA and mechanical measurements. Changes induced by varying the type and the number of isocyanates and the order of their introduction were followed. Results of the mechanical tests showed systematic differences between the materials, revealing differences in the mechanical contribution of the hard phase, since the matrix was formed from the same macrodiol polytetrahydrofuran (PTHF) in each case. Polymers with DBDI hard segments, showed higher stiffness and strength than the conventional MDI based polymers. The dominant feature was a hysteresis loop with a width that varied between the polymers. Polymers with MDI hard segments gave the lowest stresses and least hysteresis, whereas polymers with DBDI hard segments gave the highest stresses and most hysteresis. Polymers with mixed MDI/DBDI hard segments) occupied an intermediate position. Crystallinity was strongly reduced when DBDI was mixed with MDI. The general improvement in properties with DBDI was retained when the diisocyanates DBDI and MDI were included, especially when reacted together in a random fashion, rather than sequentially, in the prepolymer stage. The degree of crystallinity was found to be dictated by the diisocyanate introduced first during synthesis.