DEVELOPMENT AND VALIDATION OF AN ATOMISTIC MOLECULAR DYNAMIC MODEL TO STUDY THE THERMO-MECHANICAL PROPERTIES OF LLDPE

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ABSTRACT

In a never-ending quest for cheaper polymeric materials with better properties, broader applications and processability for recycling, polymer composites and blends may be the answer. Blends are the product of physical blending of pure polymers or polymers and wax [1-4]. Even though Luyt and co-workers [4-5] have shown that blending of polyolefins and wax with improved properties is possible, the characterization of the exact morphology of the resulting polymers blends remains a challenge. This is due to lack of adequate understanding of the interaction between wax and polyolefin at molecular level and/or insufficient analytical techniques to describe phase morphology.

Morphology of blends is linked to the miscibility of blends. Miscible blends have a continuous one-phase morphology, which has one glass transition temperature while immiscible blends have two or more glass transition temperatures depending on the number of components in the blends [6]. Various properties of blends, including transition temperature, energy of mixing and solubility parameter, could be investigated using molecular dynamics [7-10]. However, before molecular dynamics of the blends could be used, an atomistic model of the different components of the blends needs to be developed and validated.

For the purpose of model development and validation, pure polymer (LLDPE) was investigated. The development of the model entailed building of monomers, namely ethylene and 1-hexene, and geometric optimisation of these monomers with the DMol^3 module [11] in Materials Studio software [12]. Using these monomers different lengths of LLDPE with different branch contents were constructed. Different combinations of LLDPE were then incorporated in three-dimensional periodic systems using Amorphous Cell module in Materials Studio software [12]. These three dimensional periodic systems were relaxed using various molecular dynamics calculation such as NVT at 298 K and NPT ensembles at 298 and 500K. The relaxed systems were analysed and thermo-mechanical properties, such as glass transition temperature, bulk modulus and Young's modulus were calculated in order to validate the model by comparison with experimental data.

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