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Study of the glass transition temperature and the mechanical properties of PET/modified silica nanocomposite by molecular dynamics simulation

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ABSTRACT

In this study, the glass transition behavior and mechanical properties of poly(ethylene terephthalate) (PET), PET/silica nanocomposite and PET/hydroxylated silica nanocomposite were studied through molecular dynamics (MD) simulations. It was found that the density and the specific volume of PET, PET/silica nanocomposite and PET/hydroxylated silica nanocomposite regularly changed along with the changes of temperature and the transition occurs at the Tg point, the simulation results show the addition of nanosilica decreased Tg of PET in PET/silica nanocomposite, but the addition of hydroxylated silica increased Tg of PET in PET/hydroxylated silica nanocomposite,and the underlying mechanism resulting in the change of the glass transition temperature of nanocomposite was discussed. Moreover, the thermal and mechanical properties of the systems were characterized, The MD simulations showed that the addition of silica particle increases Young's modulus, bulk modulus, Poisson's ratio, lame constants and compressibility of PET/silica nanocomposite in comparison with those in the pure PET system.

1. Introduction

As a highly crystalline polymer, polyethylene terephthalate (PET) has excellent mechanical properties, creep resistance, fatigue resistance, frictional resistance, and dimension stability in relatively wide temperature ranges. On the other hand, it also has a slow crystallization rate, processing difficulties, high molding temperature, and bad impact performance, restricting the applications of PET. Generally, the processability and physical properties of PET can be improved by enhancing, filling and blending ways [1–12]. For instance, silica films have been widely used in industry as coating materials due to their excellent electrical, optical and chemical properties. They are used as passivation layers [1], antireflection layer [2], gas barriers for food and medical packages [3,4] and recently developed organic flat panel displays [5,6]. It has also been forecasted that the use of metal oxide films coated on polyethylene terephthalate (PET) will increase rapidly in the near future [7]. In the study of modified PET, the glass transition temperature (Tg) is an important intrinsic character, which influences the material properties of a polymer and its potential applications. In the glass transition state, the mechanical properties (such as deformation modulus, etc.) and physical properties (such as volume, density and specific heat) of the polymer will change. Correspondingly, various properties after molding will also be affected.

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To date, the Tg of the polymer or polymer blends has been measured using the experimental method. However, the whole processes are time-consuming and expensive. Alternatively, computer simulation can be used as an effective means to predict the Tg, which has been applied to the glass transition of many polymers [13–16]. In the majority of studies using molecular dynamics simulation, the Tg of the polymer can be obtained by analyzing its physical properties [14–18].

Therefore, in this study, the Tg of PET, PET/silica nanocomposite and PET/hydroxylated silica nanocomposite were investigated employing the molecular dynamics method. Moreover, the mechanical properties of the systems were studied to find its capability to be used in industrial applications. At the same time, the main structural factors of the systems will be analyzed to better understand the nature of the improved properties of the systems.

2. Simulation details

2.1. Model buildings and simulation method

All molecular simulations were done using the Materials Studio6.0 software package of Accelrys, Inc. (San Diego, CA) [19]. Atomistic packing models were generated using the Amorphous Cell module and were subsequently equilibration using molecular dynamics (MD) simulations with the Forcite module, The COMPASS force field [20–22] was used during each stage of the simulations. In all cases, the non-bond interactions were calculated up to a cut-off distance of 18.5 Å, which was truncated using a cubic spline function with a width of 1 Å. For this, a group-based representation for the summation of the electrostatic interactions, and an atom-based representation for the summation of the van der Waals interactions were used during packing model generation, equilibrium and subsequent production runs. The respective Andersen thermostat [23] temperatures, and Berendsen [24] pressure control algorithms were used throughout.

In this research, the repeat unit of PET structure was constructed followed by the single chain of the studied polyesters with a degree of polymerization 70. The initial macromolecular conformations of the simulated polymers were minimized. Then, energy minimized single chain of the polymer was used to construct the periodic unit cell of the studied polymer. In order to reduce the chain-terminal effect [25], an amorphous molecular model with a molecular chain was built employing the Amorphous Cell module. To create some roughly spherical silica particles, a a-quartz supercell crystal structure was first created, then all Si and O atoms outside a radius of 5 Å from the center were discarded [26] (Fig. 1(a)), hydroxylated silica

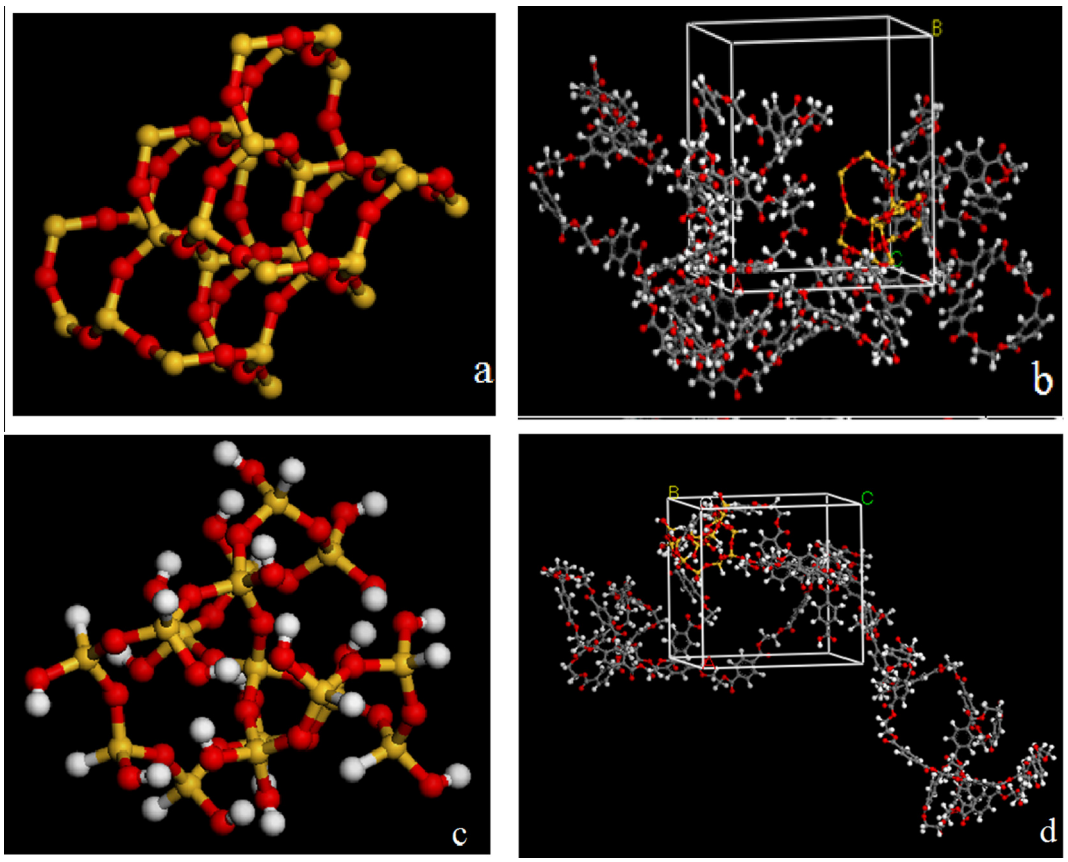

Fig. 1. Molecular model of silica nanoparticle (a), PET/silica nanocomposite (b), hydroxylated silica (c) and PET/hydroxylated silica nanocomposite (d).