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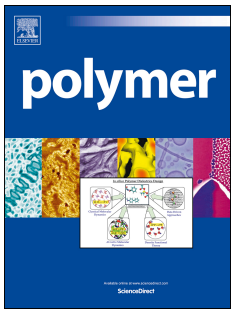

Molecular dynamics simulations of structural transitions of crystalline polystyrene in response to external stresses and temperatures

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Molecular dynamics simulations of structural transitions of crystalline polystyrene in response to external stresses and temperatures

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Abstract

The structural transitions of syndiotactic polystyrene in response to external stresses and temperature were investigated in detail by molecular dynamics simulations. Upon uniaxial compression along the *b*-axis, the nanoporous ε form was transformed into a lower-density porous "S-I" form, accompanied by marked elongation of the *a*-axis. The S-I form was transformed into the γ form following stress reduction. The transition from the ε to the β' form was also observed under tensile stress along the *c*-axis, which is associated with a conformational transition of the main chains. The stress responses and the reproducibility of the transitions were confirmed by extensive simulations. The thermal stability of the polymorphs was also examined *via* stepwise heating simulations. Experimentally observed thermal transitions of $\varepsilon \rightarrow \gamma$ and $\delta_{\mathrm{e}} \rightarrow \gamma$ were reproduced. It was found that the S-I form is more stable than the ε and δ_{e} forms under 0.27 GPa uniaxial stress along the *b*-axis. Considering the reproducibility of the stress-induced transitions and the thermal stability, the S-I form could be realized under an appropriate stress condition, though no experimental observation has been reported for now.

Keywords: molecular dynamics simulation, polystyrene, crystal, stress

PACS: 61.50.Ks, 81.40.-z

1. Introduction

The crystal structure prediction from first principles is recently much interested, not only in material design but also in fundamental sciences [1, 2]. Various methods [3–7] have been proposed and tested in combination with *ab initio* calculations and molecular dynamics (MD) simulations. These methods

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