NUCLEATION OF CHAIN ENTANGLEMENTS AND NETWORK STRUCTURE IN AMORPHOUS POLYMER UNDER TENSION: A MOLECULAR DYNAMICS STUDY

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Polymeric materials show complicated deformation behavior depending on the internal microscopic phenomena such as the cross-link or the chain network structure. In order to clarify the fundamental mechanism of the deformation behavior of the irregular, long and massive molecular chains in polymeric materials, a molecular dynamics simulation is conducted on a nanoscopic specimen of amorphous polyethylene under uniaxial tension. The specimen involves 3,542 random coil molecular chains composed of 500-1500 methylene monomer with the total amount of about 2 million methylene groups. Fig.1 shows the stress-strain curve derived from the tensile simulation with the condition of $\dot{e}_{zz} = 5.0\text{x}10^{11}$/s. It exhibits linear elastic relation at the initial stage of e_{zz}<0.03, then the material "yields" by elongating without stress increase up to the strain of 1.5, where the strain hardening appears. Careful investigation of changes in dihedral angle and morphology of all molecular chains reveals that the *gauche*→*trans* transition dominantly takes place during yielding, generating new network-like structure composed of entangled molecular clusters and oriented chains bridging them. Fig.2 shows the entangled clusters, evaluated with the molecular density, in the network structure nucleated. The clusters keep the relative position each other, as the clusters A, B and C in the figure, expanding the distance between them in the tensile direction. The strain hardening is due to the directional orientation and stretch of molecular chains between entanglements in the nucleated structure.

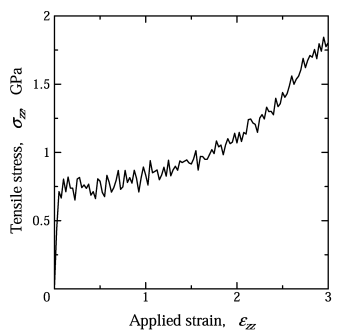

Fig.1 Stress-strain curve.

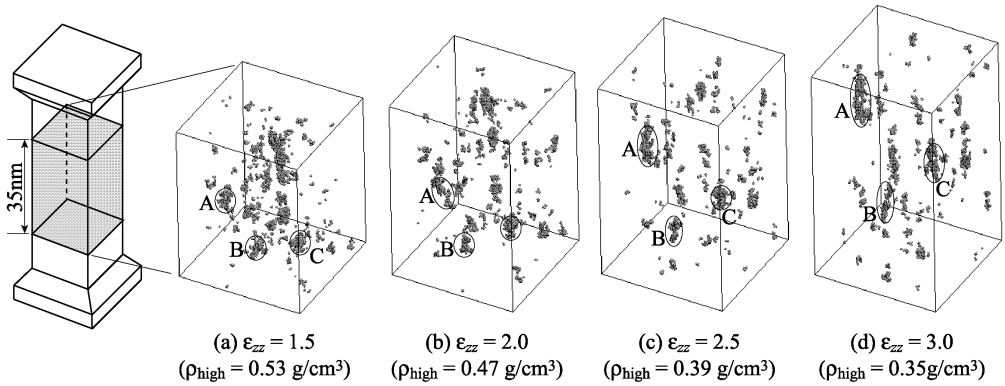

Fig.2 Chain entanglements evaluated by the high atomic density.