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# 論 文 要 約

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| 系・コース：<br>Department of, Graduate major in | 応用化学 系<br>応用化学 コース | 申請学位（専攻分野）：<br>Academic Degree Requested | 博士 (Science)<br>Doctor of |
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要 約（500 字程度）


Polymer crystallization is a process where the chains evolve from the random coils to form regular lamellae structures. Several crystallization mechanisms including Lauritzen-Hoffman theory, aggregation model, bundle model and the multistage model are proposed to explain the crystallization process at the chain level. Many techniques such as neutron scattering (NS), X-ray diffraction (XRD), nuclear magnetic resonance (NMR), infrared spectroscopy (IR), Raman spectroscopy and microscopy are applied to extract the chain trajectory in the crystal formed from dilute solution and melting state. However, some of the above techniques cannot access molecular scales and others require specific morphology and samples (e.g., mono-disperse oligomer at the chain end labeled by 2H). The questions of how, when and where the polymer chains fold during the crystallization are still under debate. Recent results from Zhang group by advanced SMFS-AFM have made it possible to investigate molecular structure of individual polymer chain such as polyethylene oxide (PEO) and polylactide (PLA). This advanced method has been successfully utilized to investigate the molecular structures in crystals formed from dilute solutions affected by crystallization temperature, concentration and chain conformation. However, Temperature effect (Tc) on chain-level structures were not realized in past studies.

In this dissertation, I have focused on polyethylene oxide (PEO) which can be synthesized with double thiol ends and applied the AFM-based SMFS to investigate crystallization mechanism and chain folding structure of PEO.

The AFM-SMFS results showed the different chain-level structures along crystallization time. It suggested that i) the chains will self-folded as small nuclei in solution by primary nucleation as the first stage and ii) the subsequent aggregation step resulted in morphological differences dominated by kinetics as the second stage. Next, the temperature effect was systematically investigated. The SMFS results indicated that i) all PEO adopted similar chain-folding structure but different chain-folding structures; ii) a larger supercooling $\Delta T$ generated intermediate model with smaller Rg in crystal. The difference of the chain-folding structure on $\Delta T$ demonstrated that chain-folding was a local process in the first stage of crystallization but finally form different chain structures. Moreover, cluster unit and inter-chain structures will be discussed.

To further study chain sliding during crystal melting, we conducted in situ-AFM to investigate transition of chain-folding structure of PEO. However, the chain behavior during melting has been debatable in the past decades. Thus, we investigated morphology, chain structure, and self-assembled single chain structure of PEO. It is found that the intermediate chain structure in multi-layer crystal will transits to thick adjacent re-entry chain-folding structure first during melting. It indicated the crystal will exhibit re-crystallization and re-melting during melting process, which results from the chain sliding at high temperature.


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