Extension of Reverse Monte Carlo Method for 2D-USAXS Experimental Data

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Structures of fillers (nano-particles) in an elastomer has been attracted much interest.[1, 2] Some of the motivations of this interest are as follows. First, the reinforcement effects[3] of elastomers by addition of fillers such as carbon black and silica are important for industrial applications such as tiers and rubbers. Secondly, how bulk structures of nano size particles in an elastomer can be observed is an interesting problem of synchrotron radiation physics. Recently, the Two-dimensional Ultra-Small-Angle X-ray Scattering (2D-USAXS) method is established by using the Beam line BL20XU of Spring-8 (Hyogo, Japan) in order to overcome the limitations of 3D-TEM: its observable area is only around surfaces of a sample and observations of samples stretched outside is difficult. Thirdly, the motions of fillers and polymer in an elastomer play an important role of the mechanical properties. Simulational study of this system is one of challenging problems of computational physics.

Shinohara *et. al.* observed 2D-USAXS patterns of filled rubber under elongation in order to investigate structural changes of the fillers.[1] The X-ray CCD detector coupled with X-ray Image Intensifier [4] was used as the detector. Used samples are Styrene-Butadiene Rubber filled with silica particles, of which diameter is about 100nm. The 2D-USAXS images show hysteresis corresponding to that of the stress-strain curve. The 2D-USAXS clarified the structure changes of the fillers in stretched rubber for the first time, which had been only speculated based on viscoelastic experiments.

The Reverse Monte Carlo analysis[5] are widely used as a method for generating three-dimensional structural models from the data of the structure function obtained by X-ray scattering experiments for disordered systems. In the Reverse Monte Carlo (RMC) method[5], the three-diomensional structure is expected as the difference between observed and calculated structure functions is minimized. The RMC process starts with the initial configuration of particles. In loops of RMC process, the structure functions are recalculated for the trial move of the particle chosen randomly and the change $\Delta(\chi^2)$ in the goodness-of-fit parameter χ^2, which is given as $\chi^2 = \sum_q (S^{\rm exp}(q) - S^{\rm comp}(q))^2/\sigma^2$ for a simple example, where $S^{\rm exp}$ means the structure function obtained from experiments, $S^{\rm comp}$ one calculated, and σ is the parameter such as a standard deviation. For $\Delta(\chi^2) \leq 0$, every trial move is accepted. Trial moves which worsen the fit ($\Delta(\chi^2) > 0$) are accepted with a probability of $P = \exp(-\Delta(\chi^2)/2)$. Repeat above loops until χ^2 converges good enough. The original RMC are applied only for the one-dimensional data such as $S(q)$ and the pair distribution function $g(r)$, which are described as a function of a wave number q and a distance r, respectively.

In this study, We extended the Reverse Monte Carlo method for 2D-USAXS Experimental Data by using two-dimensional structure functions $S(q_h, q_k)$ instead of one-dimensional one $S(q)$. Here, we call this extended version as 2D-RMC. We introduce the implements using Super Computers, show the results of 2D-RMC analysis for two-dimensional structure functions calculated from artificial configurations of particles and discuss the validity of 2D-RMC analysis. Preliminary results of 2D-RMC analysis for 2D-USAXS images will be also discussed.

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