V-N Simulation of Molecular Clusters

Monte Carlo simulation is a powerful method for studying soft matter such as polymer, gel and biological molecules. We performed the structure analysis of chemical gel and characterized gel with cross-linkers. For vesicles, we studied self-assembly, fusion, adhesion and structural changes due to mechanical forces. In both systems, we obtained qualitatively good agreement with experimental results, and gave some insights to the mechanism of order formation of soft matter.

V-N-1 Characterization of Gel Using Modeled Radical Polymerization with Cross Linkers Performed by Monte Carlo Method

NOSAKA, Makoto[1]; TAKASU, Masako[1]; KATOH, Kouichi[2]
([1]IMS and Kanazawa Univ.; [2]Kanazawa Univ.)

[*J. Chem. Phys.* **115**, 11333 (2001)]

In this study, some physical quantities for characterization of gel are proposed. Polymer networks (gel) are investigated by Monte Carlo simulation using modeled free-radical cross-linked polymerization in continuous system. The distributions of degree of polymerization for clusters obtained in this simulation show qualitatively good agreement with the experimental results. Linkers are classified to two types according to the roles in networks, and their ratios are discussed. The normal and weighted ratios of gel are defined using percolation theory. These ratios are compared with the changes in distribution.

V-N-2 Analysis of Intra- and Inter-Linkers in Gels by Brownian Dynamics Simulation

NOSAKA, Makoto[1]; TAKASU, Masako[1]
([1]IMS and Kanazawa Univ.)

The process of gelation was analyzed by Brownian dynamics simulation using modeled radical polymerization with cross-linkers. Particle densities were set near the gelation threshold determined experimentally (monomer density d = 200 mM 400 mM and 600 mM). We performed simulations under two conditions that did (Rule D) and did not (Rule A) prohibit the formation of intra-linkers. With Rule D, we observed gelation at d = 600 mM, and clustering at d = 400 mM. On the other hand, with Rule A, we did not observe gelation with any of the densities tested. We only observed clustering at d = 600 mM. Some quantities were investigated by comparing the results under the two conditions.

V-N-3 Self-Assembly of Amphiphiles into Vesicles: a Brownian Dynamics Simulation

NOGUCHI, Hiroshi; TAKASU, Masako[1]
([1]IMS and Kanazawa Univ.)

[*Phys. Rev. E* **64**, 041913 (2001)]

We studied the vesicles of amphiphilic molecules using a Brownian dynamics simulation. An amphiphilic molecule is modeled as the rigid rod, and the hydrophobic interaction is mimicked by the local density potential of the hydrophobic particles. The amphiphilic molecules self-assemble into vesicles with bilayer structure. The vesicles are in fluid phase, and we calculated the lateral diffusion constant and the rate of the flip-flop motion of molecules in vesicles. The self-assembly kinetics into vesicles was also investigated.

V-N-4 Fusion Pathways of Vesicles, a Brownian Dynamics Simulation

NOGUCHI, Hiroshi; TAKASU, Masako[1]
([1]IMS and Kanazawa Univ.)

[*J. Chem. Phys.* **115**, 9547 (2001)]

We studied the fusion dynamics of vesicles using a Brownian dynamics simulation. Amphiphilic molecules spontaneously form vesicles with a bilayer structure. Two vesicles come into contact and form a stalk intermediate, in which a necklike structure only connects the outer monolayers, as predicted by the stalk hypothesis. We have found a new pathway of pore opening from stalks at high temperature: the elliptic stalk bends and contact between the ends of the arc-shaped stalk leads to pore opening. On the other hand, we have clarified that the pore-opening process at low temperature agrees with the modified stalk model: a pore is induced by contact between the inner monolayers inside the stalk.

V-N-5 Adhesion of Nanoparticles to Vesicles: a Brownian Dynamics Simulation

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[*Biophys. J.* **83**, 299 (2002)]

We studied the interaction of bilayer vesicles and adhesive nanoparticles using a Brownian dynamics simulation. The nanoparticles are simple models of proteins or colloids. The adhering nanoparticle induces the morphological change of the vesicle: budding, formation of two vesicles in which only outer monolayers are connected, and fission. We also show that the nanoparticle promotes the fusion process: fusion-pore opening from a stalk intermediate, a neck-like structure that only connects outer monolayers of two vesicles. The nanoparticle bends the stalk, and induces the pore opening.

V-N-6 Structural Changes of Pulled Vesicles: a Brownian Dynamics Simulation

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[*Phys. Rev. E* **65**, 051907 (2002)]

We studied the structural changes of bilayer vesicles induced by mechanical forces using a Brownian dynamics simulation. Two nanoparticles, which interact repulsively with amphiphilic molecules, are put inside a vesicle. The position of one nanoparticle is fixed, and the other is moved by a constant force as in optical-trapping experiments. First, the pulled vesicle stretches into a pear or tube shape. Then the inner monolayer in the tube-shaped region is deformed, and a cylindrical structure is formed between two vesicles. After stretching the cylindrical region, fission occurs near the moved vesicle. Soon after this the cylindrical region shrinks. The trapping force ~ 100 pN is needed to induce the formation of the cylindrical structure and fission.