博士論文の内容の要旨

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論文題目	Study on hydrogel microspheres adsorbed at the air/water interface (気水界面における高分子ハイドロゲル微粒子に関する研究)

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Soft hydrogel microspheres (microgels), which are colloidal particles with three-dimensional polymer networks that swell in water, have received attention as a soft material that may be able to stabilize fluid interface such as liquid/liquid or gas/liquid interface. Soft microgels can be better stabilizers mainly due to their high colloidal stability and stimuli responsivity, which add further fascinating properties to their interfacial behavior. Changes in the physicochemical properties of the microgels in response to external stimuli can be expected to lead to many applications in e.g. the food industry, cosmetics, and smart delivery systems. Moreover, the size and structures of microgels can be changed upon adsorption at the interface due to their softness, which might change the interfacial properties such as the interfacial distortion, interfacial tensions, and the rheological properties. In order to better understand the mechanisms operative at such interfaces and to develop applications, intimate knowledge of the behavior of soft deformable microgels at fluid interfaces is of paramount importance. However, yet theoretically and computationally describing the behavior of microgels at the fluid interface is challenging, given that the structure of the microgels depends on their environment. Although the interfacial behavior of microgels at fluid interfaces has been investigated using various observation techniques and theoretical analysis methods, the details of the adsorption behavior, the deformation conformation, and the organization of the structure of microgels at the fluid interface remain unclear. Therefore, the author investigated the interfacial behavior, i.e., the adsorption and deformation of microgels at the air/water interface using microgels whose size allowed direct in situ observation. The behavior of the microgels at the interface is discussed by systematically combining macroscopic/microscopic evaluation methods with previously established knowledge.

In **Chapter 1**, the deformed structure of microgels adsorbed at the air/water interface was investigated, which proved to be difficult due to the limitations associated with the resolution of the microscopy technique and the size of the microgels. Consequently, larger, micron-sized microgels were prepared by a modified aqueous precipitation polymerization method, although the synthesis of such large particles proved to be non-trivial due to the fact that gravity effects strongly influence particle motion and colloidal stability. Using the developed microgels that can deform to a size of 20  $\mu$ m upon adsorption at the air/water interface, the deformation dynamics of soft microgels could be visualized clearly and analyzed quantitatively.

In **Chapter 2**, the focus was placed on the effect of charge incorporated in microgels, which is one of the most important parameters in the particles, and the adsorption, deformation, and self-organization behavior of such charged microgels were investigated. A series of experiments afforded information that clarified the adsorption, interpenetration, and deformation behavior of charged microgels at the air/water interface.

These findings on the interfacial behavior of microgels at the air/water interface can be expected to develop further applications, e.g., foams and emulsions stabilized by microgels, where it is of paramount importance to understand the interfacial behavior of microgels at the air/water interface.