論　文　の　要　旨

題　目　　Preparation of Macroporous Particles Decorated with Cellulose Nanofiber and Their Protein Adsorption Performance

（セルロースナノファイバが担持したマクロポーラス微粒子の合成とタンパク質吸着特性）

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Effective protein adsorption has attracted attention for broad application in the biomedical field. To date, continuous improvement on adsorbent particles was developed by the utilization of various materials or modifying their physicochemical properties to increase the ability of adsorption, which is adjustable according to the required application. This dissertation introduced a combination of TEMPO-oxidized cellulose nanofiber (TOCN) and macroporous particles through a spray process as the new material for advanced protein adsorbent. It aims to gain a better understanding of the effect of surface functional groups and nanostructures on the adsorption behavior of protein as one of the macromolecules. Several parameters in the process of synthesis and adsorption performance have been carried out to investigate the effectiveness of this combination and to understand whether the driving factors in the adsorption process. A brief description of each chapter in this dissertation is shown below.

Chapter 1 provides the background and motivation for current research on development powder technology in protein adsorption. The theoretical explanation and review of the previous studies were presented in the development of physicochemical properties modification of adsorbent.

At first, in **Chapter 2**, this dissertation focused on the scheme selection of an effective way to prepare the macroporous particle by the spray-templating method. Macroporous carbon particles were prepared through poly (methyl methacrylate) (PMMA) templating in the variation of PMMA concentration and particle size to evaluate the formation of macroporous structures. The characteristics of resulted microporous carbon particles are then compared with those produced through polystyrene latex (PSL) templating. It was found that with similar morphological results, the PMMA template reduced the energy consumption for decomposition and produced a higher specific surface area of macroporous carbon particles compared to that using the PSL template.

PMMA templating method then used for synthesis macroporous SiO_2 particles in **Chapter 3** as the host of TEMPO-oxidized cellulose nanofiber (TOCN) deposition. The performance of TOCN decorated macroporous SiO_2 (TOCN@macroporous SiO_2) particles then evaluated for lysozyme adsorption. The mass ratio of TOCN and SiO_2 particles was varied to understand the contribution of each component as well as to obtain the optimum conditions. The results show that TOCN@macroporous SiO_2 particles have a unique cellulose nanofiber network structure on the macroporous with highly-negative zeta potential(-62 ± 2 mV) close to the zeta potential of TOCN but possess higher specific surface area. The presence of macroporous SiO_2 particles leads to the improvement of lysozyme adsorption 17% higher than TOCN particles. Furthermore, TOCN@macroporous SiO_2 particles have very high reusability (> 90% adsorption capacity) and good

adsorbate release (> 80%) after 10 times of use.

In **Chapter 4**, the macropore size of silica particles was adjusted to understand how TOCN depositions decorate the macropore structure. In addition, the adsorption kinetics, thermodynamics, and isothermal parameters were studied to analyze the driving factors of protein adsorption. The result shows that the adsorption process occurred spontaneously at any temperature with adsorption equilibrium achieved before 10 minutes due to the contribution of electrostatic interaction and hydrogen bonding. Furthermore, the protein adsorption capacity depends on protein interaction with carboxylate group and the accessible pore space of adsorbent. The pore size corresponding to the length of TOCN can affect the TOCN deposition both to the external surface or penetrate the macropore structure. This deposition behavior was resulting in a broad pore size distribution with abundant accessible active sites that can adsorb large molecules such as lysozyme through a multi-layer adsorption process *via* a pore-filling mechanism.

General conclusions of all topics are listed in **Chapter 5**.