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●特集　膜濾過と固液分離の接点

Membrane Filtration of Soft Colloids

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The solid–liquid separation of soft particles or colloids, such as water gels, proteins, emulsions, microbial cells and flocs, has become increasingly important in biochemical, food, biomedical, pharmaceutical, wastewater treatment, and chemical industrial processes in recent years for the production or recovery of high–quality or high–valuable products. Comparing with the filtration of rigid particles, the filtration of soft colloids exhibits more complex behaviors which causes much difficult in filtration analysis and device selection. The typical filtration phenomena of soft colloids, including particle deformation, creeping cake compression and liquid transfer across the interface between colloids and surroundings solvent, always result in a filter cake with extremely high specific cake filtration resistance and extremely low cake porosity. A thin compact cake layer is formed next to the membrane surface which exhibits most filtration resistances. Soft colloids are therefore very hard–to–filtered. In this article, the features of the filtration of soft colloids are described. The current development in filtration mechanisms and future prospects are reviewed and presented.

Key words : soft colloids ／ deformable particles ／ solid–liquid separation ／ membrane filtration ／ cake property

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1.　Introduction

Because different kinds of soft particles and colloids, such as microbial cells, hydrogels, proteins, emulsions, biopolymers, are desired or valuable products in various pharmaceutical, biochemical and chemical engineering processes, the demand for solid–liquid separation of soft particles has been markedly increased in recent years. The solid–liquid separation of soft particles is always much difficult due to the high deformability and complex compression behavior. Membrane filtration is an efficient method for this purpose compared to the other conventional operations, e.g., sedimentation, centrifugation, etc. However, the filtration characteristics of soft colloids are more complex compared to those of rigid particles. The high deformability, time–dependent compression behavior, and extremely compact structure of cakes formed by soft colloids cause the difficulty not only in filtration operation but also in the data analysis.

The development of membrane filtration technique for soft colloids has become increasingly active in recent years. Microfiltration and ultrafiltration were widely used for the separation of biological products consisting of various soft colloids. In case of a filter cake formation, Tiller and Green [1)] observed that a “skin layer” was constructed next to the membrane surface due to the highly compressible behavior. Such compact layer has only 20% cake thickness but depletes most solid compressive pressures and exhibits more than 90% of the total filtration resistance. Therefore, to analyze the mechanism of the skin layer formation to reduce filtration resistance and mitigate the flux decline is an important issue for this kind of operation in the future.

This article presents a brief overview of the filtration characteristics of soft colloids. The current development in the filtration mechanisms and future prospects are reviewed and presented.

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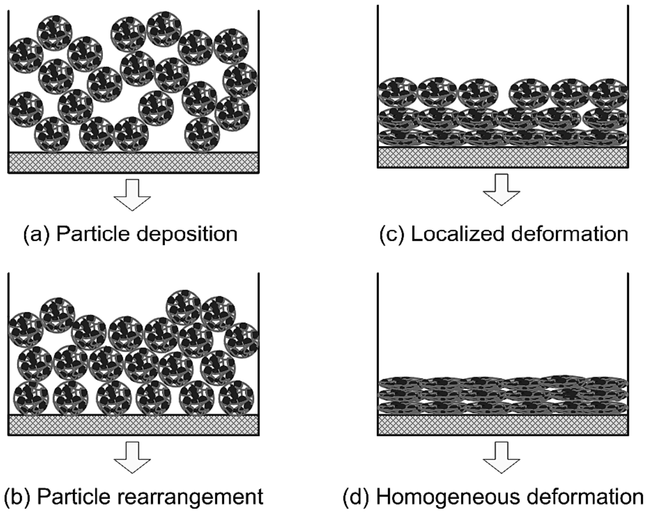
(a) Particle deposition
(c) Localized deformation
(b) Particle rearrangement
(d) Homogeneous deformation

Fig. 1 The four stages of cake compression during a filtration[35].

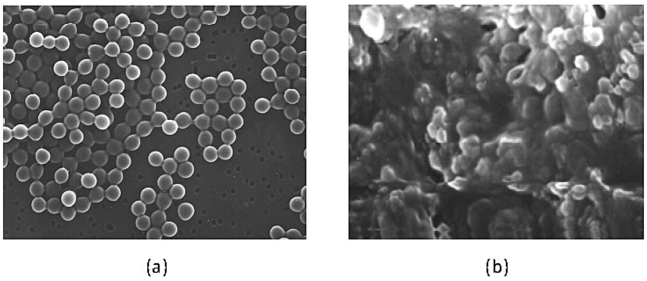
(a) (b)

Fig. 2 A comparison of cake structures formed by rigid and soft particles (a) PMMA rigid particles, (b) pseudomonas cells.

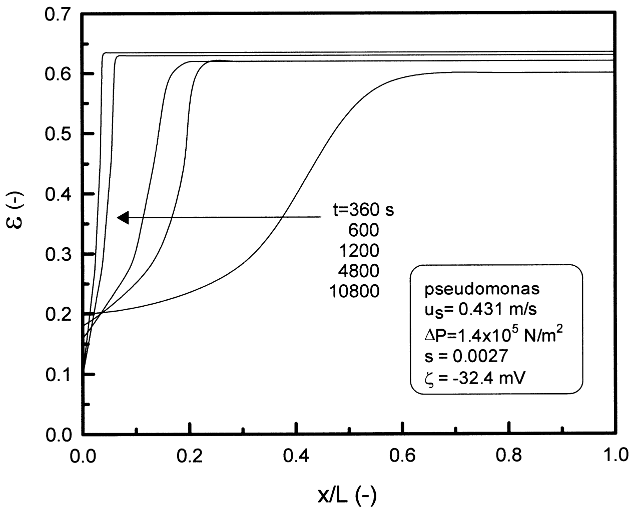

Fig. 3 Distributions of local cake porosity in cake at different filtration times[3].

2. Studies on the filtration of soft colloids

Soft colloids have several filtration characteristics, e.g., particle deformability, creeping effect during cake compression, liquid flowing through particle surface, etc., which play important roles in determining the filtration performance and should be taken into consideration in the selections of devices and operating conditions. Many researchers and engineers therefore not only paid their attentions but also devoted their efforts on those subjects. Some recent researches are reviewed as below.

2.1 Effect of particle deformation

Most soft colloids are deformable. They may deform their shape under compression, and, in some cases, a part of liquids contained in colloids are squeezed out continuously during a filtration. The entire filtration and compression course can be ideally divided into four stages, as shown in Fig. 1. In the early periods of a filtration, particles deposit on the membrane surface to form a filter cake, as shown in Fig. 1(a). In Fig. 1(b), particles rearrange to construct a more compact cake but remain their original shape in the second stage. A localized cake deformation comes after a retardation time, and the particles near the membrane surface come into being out of shape irreversibly, as shown in Fig. 1(c). Surface contacts between particles occur, and a compact skin layer forms in the third stage. If the compressive pressure is high enough, the cake will be finally homogeneous deformation. The cake densification is possible to result in particle ruptures or a solid–like structure, as shown in Fig. 1(d).

Surface contacts always occur between soft colloids packed in a filter cake. This factor may cause soft colloids to form a filter cake with extremely low porosity and high filtration resistance. Fig. 2 shows a typical comparison of cake structures formed by rigid and soft particles. The rigid PMMA particles are still spherical after deposition on the membrane surface, as shown in Fig. 2(a). However, the soft particles, pseudomonas cells, have deformed their shape even under a transmembrane pressure of 1 bar. The original spheroidal shape can no longer been observed in Fig. 2(b)

Hwang et al. [2, 3] studied the cake properties formed by pseudomonas cells in microfiltration. Fig. 3 shows the distributions of local cake porosity during a crossflow microfiltration, which was simulated using a dynamic analysis method. The local cake porosity decreased, and a compact skin layer with a porosity as low as 0.1 formed gradually next to the membrane surface as filtration time increased. The thickness of the skin layer was smaller than 10% as filtration time exceeded 5000 s.

Nakanishi et al. 4) and Tanaka et al. 5) found that the specific filtration resistance of cakes formed by microorganisms was strongly dependent upon filtration pressure. Lanoisellé et al.6) developed a mechanical expression model for cellular materials. The variations of extraparticle, extracelluar and intracellular volumes under constant pressure compression were calculated by solving a set of partial differential equations. Meeten7) carried out a constant pressure filtration of Sephadex, a kind of deformable particle, using a planar septum. His results indicated that the compressibility of Sephadex cake varied in accordance with the pressure magnitude. Hwang and coworkers 2, 8, 9) indicated that the local tangents of the filtration curve might increase the slopes due to particle compression and deformation. The cake compressibility therefore increased continuously until it reached the maximum value during a filtration. Lu et al. 10 ~ 12) derived a linear viscoelastic model to characterize the particle deformation under a given hydraulic drag and mechanical loading based on the Hertz theory. The parameters for viscous and elastic behaviors were determined through mechanical compression experiments and could be used to explain the drastic increase in specific cake filtration resistance during filtration 2, 13, 14).

Kersey et al. 15) used filamentous hydrogel nanoparticles to study the effects of aspect ratio and deformability on nanoparticle extravasation through 0.2 μm pores. They indicated that longer nanoparticles may be able to enter a pore at any point along their axis, bend under the pressures exerted during filtration, and deform to allow the particles to translate through the pores under the same pressures. Tung et al. 16) studied the membrane filtration of soft particles. They concluded that softer particles resulted in more severe pore blocking problems and greater flux decline. Hwang and Wang 17) studied the microfiltration characteristics of *Bacillus subtilis* under different culture conditions. They found that longer culture time and lower filtration pressure were beneficial for protein/polysaccharide separation due to the cell deformable characteristics. Qu et al. 18) conducted a dead–end filtration of milk casein micelles, a kind of soft and sponge–like natural colloids. They indicated that the deposit of casein micelles was packed closely due to the compression in a filtration, and the micelles relaxed to their original dimensions once pressure was released.

2.2 Effect of cake compression

Several efforts have been devoted to analyze the viscoelastic compression behaviors of cake formed by soft particles in recent years. Christensen et al. 19) measured the hydraulic pressure at the sample-piston interface to correct the overestimation of the transition time of cake consolidation using conventional theory. Recently their studies on the dynamic cake compressibility demonstrated that the specific cake filtration resistance increased with time due to the nonlinear filtration behavior 20). The time functions of cake porosity and filtration resistance are considerable for organic materials' dewatering. In the conditions of cake consisting dual soft materials, the contribution of each component to cake compressibility was studied by Hwang and Yang 21). They proposed a compression model for microbial cells/polysaccharide dual component cake. The variations of cake compressibility under different pressures were reasonably explained by the soft materials' deformations. In later study on the same materials' microfiltration, Hwang et al. 22) concluded that the filter cake exhibited a more compact structure and much higher filtration resistance when more dextran molecules packed into the yeast cake structure.

Iritani et al. 23) revealed that the dynamic deposition behaviors of protein molecules in a dead–end ultrafiltration could be accurately described using a compressible cake filtration model. The cake tended to have a more compact structure at the membrane surface in comparison with a relatively looser condition at the cake surface. Kapur et al. 24) proposed an approximate model for the cake consolidation stage for taking cognizance of the facts that the porosity and thickness of the compact layer decreased and the fluid stress dropped continuously with filtration time. Andersen et al. 25) proposed a method based on Terzaghi's consolidation model to obtain cake permeability and compressibility from experimental data. The coefficients in the model were calculated accordingly. Meireles et al. 26) used a mechanical combined osmotic model to describe the variations in volume fraction of yeast cells in cake. The specific cake filtration resistance was calculated by integrating the Lattice Boltzmann model into the basic filtration equation.

Some researchers paid their attentions on the creeping effects of cake compression. Iritani et al.27) studied the deformation behavior of hydrogel beds under pressure. They used a modified Terzaghi model to examine the creeping effect in bed consolidation and expansion. The roles of osmotic and mechanical pressures on the particle deformation behaviors were discussed. Cao et al.28) found that the cake formed by O/W emulsion was highly compressible due to the deformability of oil droplets, and the average volume fraction of oil droplets and the average specific resistance were kept constant throughout the course of a dead–end filtration. Iritani et al.29) examined the cake properties in unstirred dead-end microfiltration of emulsion-slurry using the same model in their previous study27). The results demonstrated that the filtration resistance could be evaluated in terms of a resistance-in-series model for the bi–layer filter cake composed of O/W emulsion and fine particles. Christensen and Keiding30) concluded that the local solid volume fraction of sludge cakes changes not only with effective pressure but also with time. They developed a new filtration model by adopting the concept of cake creeping to satisfactorily describe the experimental data of waste-activated sludge. Christensen and Hinge31) used a Terzaghi–Voigt combined model to describe the cake consolidation. Their results showed that the solid volume fraction in cake increased by 17～35% after the transition point of creeping consolidation. Iritani et al.32) studied the cake consolidation formed by tofu and okara using a compression-permeability cell. They developed a multi–stage creep model consisted of a number of Voigt elements connected in series to describe the complicated consolidation behaviors.

Christensen et al.33) synthesized different latex particles and used for filtration experiments. They indicated that higher acrylic acid content caused the latex particle cake to have more obvious creeping effect during a filtration stage. In their later study, Christensen et al.34) claimed that the cake consolidation can be divided into a primary consolidation stage controlled by hydrodynamic effects and a secondary consolidation stage controlled by creeping effects. The creeping behavior of activated sludge was more evident, but no clear transition between primary and secondary consolidation was found. Hwang et al.35, 36) prepared three kinds of porous gel particles with different compression properties used in dead–end and cross–flow microfiltration experiments. They used a Voigt–in–series model to describe the cake compression behavior and indicated that the particle softness was the most important factor affecting the cake properties and filtration flux. The cake compressibility and two additional parameters for particle compression, the retardation time and softness index, were introduced to well interpret the viscoelastic compression behaviors of porous gel particles. The variations of local cake properties can be explained using their proposed cake compression model35). Fig. 4 shows typical relationships between the average specific cake filtration resistance, α_{av}, and transmembrane pressure, ΔP, at different filtration times. The data shows that α_{av} increases with time for a given pressure and increases with increasing filtration pressure at a fixed time. The cake compression during filtration results in more compact cake and higher filtration resistance. Although power relationships exists between α_{av} and ΔP, the relation varies with time for porous gel particles.

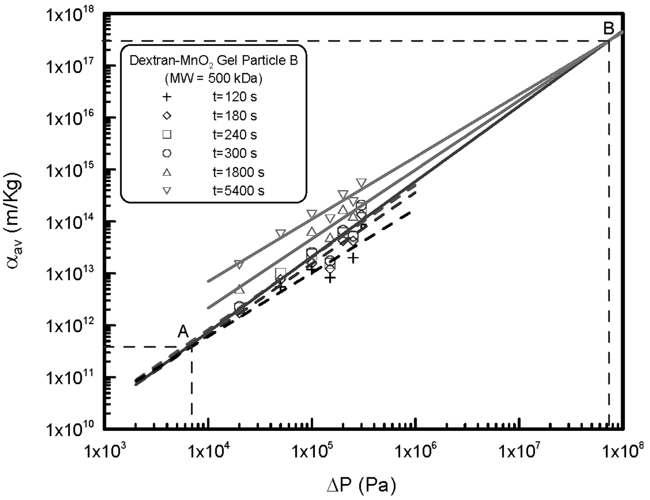

Fig. 4 The relationships between α_{av} and ΔP at different times35).

2.3 Effect of particle interactions

Soft particles often exhibit strong interactions with neighbor particles, such as the van der Waals, electrostatic, capillary, affinity, and hydrophobic interactions. These interactions significantly influence the cake structure and filtration resistance. For instance, the electric double layer caused by surface charge affects the particle packing as well as the cake structure. Tim et al.[37] fractionated casein micelles from whey proteins using microfiltration. They claimed that the specific cake filtration resistance depended on pressure and increased as the hydrophilic repulsion between casein micelles was reduced at a pH beyond the isoelectric point. Jimenez–Lopez et al.[38] indicated that the cross-flow microfiltration performance of skimmed milks was ionic strength dependence. The deposit on the membrane surface became more cohesive and harder to remove with the increase in ionic strength. The relations between filtration performance, deposit layer characteristics and colloidal properties were well discussed.

3. Conclusion

Filtration technologies of soft particles and colloids have become increasingly important in the manufacture processes of highly valuable products in recent years. Compared with the filtration of rigid particles, soft colloids have many filtration characteristics which affect the filtration performance significantly. Recent studies on the filtration characteristics of soft colloids were reviewed in this article. The current development in filtration mechanisms were presented.

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