



HOKKAIDO UNIVERSITY

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博士の専攻分野の名称 博士（工学） 氏名 中沢 禎文

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Fate and Effective Control of Superfine Powdered Activated Carbon Particles in Drinking Water
Treatment Process Consisting of Coagulation-Flocculation, Sedimentation, and Sand Filtration
(凝集沈澱砂ろ過浄水処理における微粉化活性炭の処理性と高効率制御)

Superfine powdered activated carbon (SPAC; particle diameter 1 μm) has greater adsorptivity for organic molecules than conventionally sized powdered activated carbon (PAC). SPAC is, however, not used in conventional water treatment involving coagulation–flocculation, sedimentation, and rapid sand filtration (CSF), because it is unclear whether CSF can adequately remove SPAC from the water. In this study, I therefore investigated the residual SPAC particles in water after CSF treatment.

First, I developed a method to detect and quantify trace concentration of carbon particles in the sand filtrate. This method consisted of 1) sampling particles with a membrane filter and then 2) using image analysis software to manipulate a photomicrograph of the filter so that black spots with a diameter $>0.2 \mu\text{m}$ (considered to be carbon particles) could be visualized. This method revealed that CSF removed a very high percentage of SPAC: approximately 5-log in terms of particle number concentrations. When waters containing 7.5-mg/L SPAC and 30-mg/L PAC, concentrations that achieved the same adsorption performance, were treated, the removal rate of SPAC was somewhat superior to that of PAC, and the residual particle number concentrations for SPAC and PAC were at the same low level (100–200 particles/mL). However, the activated carbon particles after sand filtration were smaller in terms of particle size and were charge-neutralized to a lesser extent than the activated carbon particles before sand filtration. Therefore, the tendency of small particles to escape in the filtrate would appear to be related to the fact that their small size leads to a low destabilization rate during the coagulation process and a low collision rate during the flocculation and filtration processes.

I further focused on the two key components of coagulation (mixing intensity and coagulants) and investigated how to effectively reduce the residual SPAC after CSF.

Astonishingly, the flash mixing (the first process of CSF), especially its G (velocity gradient) value, played the most important role in determining the residual SPAC in the filtrate of sand filter (the fourth process). Even if the slow mixing time was short, a sufficiently large G value but short T (mixing time) value in flash mixing effectively reduced the residual SPAC. When the total GT value of flash and slow mixing was fixed at a constant, priority should be given to flash mixing to reduce the residual SPAC.

Among 23 PACl (poly-aluminum chloride) coagulants, PACl with a high-basicity (basicity 70%) and with sulfate ion (0.14 of sulfate/aluminum in molar ratio), produced by $\text{Al}(\text{OH})_3$ -dissolution, were the most effective to reduce the residual SPAC after CSF. PACls produced by base-titration, which have

been intensively investigated in previous researches, were not effective due to lack of floc-formation ability. However, their Al species composition determined by the ferron method were almost the same as those of PACl by $\text{Al}(\text{OH})_3$ -dissolution, and their charge-neutralization capacities were higher. PACls produced by $\text{Al}(\text{OH})_3$ -dissolution possessed both charge-neutralization and floc-formation abilities, but the former ability was more important to minimize the residual of SPAC.

Additionally, I examined the production and fate of SPAC particles with lacking in charge neutralization as a source of the residual in sand filtrate after CSF treatment by using a flow-through mode CSF plant. Almost all of the carbon particles in the water were charge-neutralized by coagulation treatment with rapid mixing, but a very small amount ($\leq 0.4\%$ of the initial concentration) of very fine carbon particles with a lesser degree of charge neutralization were left behind in coagulation process. Such stray carbon particles were hardly removed by subsequent flocculation and sedimentation processes, and some of them remained in the sand filtrate. The concentration of residual carbon particles in the sand filtrate varied similarly with that of the stray carbon particles. The stray and residual carbon particles were similarly smaller than the particles before coagulation, but the residual carbon particles had less charge neutralization than the stray carbon particles. The turbidity of water samples collected after sedimentation was not correlated with the residual carbon concentration in the sand filtrate, even though it is often used as an indicator of treatment performance with respect to the removal of suspended matter. Based on these findings, I suggest that reduction of the amount of stray particles should be a performance goal of the CSF treatment. Examining this concept further, I confirmed that the residence time distributions in the coagulation and flocculation reactors influenced the concentration of stray carbon particles and then the residual carbon particle concentration in sand filtrate, but found that the effect was dependent on coagulant type. A multi-chambered-reactor configuration lowered both the stray carbon particle concentration after coagulation and the residual carbon particle concentration in sand filtrate compared with a single-chambered reactor configuration. When a normal basicity PACl that consisted mainly of monomeric Al species was used, the stray carbon particle concentration was decreased during coagulation and then gradually decreased during subsequent flocculation because the monomeric Al species were transformed to colloidal Al species via polymeric Al species. In contrast, when a high-basicity PACl that consisted mostly of colloidal Al species was used, coagulation largely decreased the stray carbon particle concentration, which did not decrease further during subsequent flocculation. These findings will be valuable for controlling residual carbon particles after the CSF treatment.