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学 位 論 文 内 容 の 要 旨

博士の専攻分野の名称　博士（工学）　氏名　CHEN Yize

学 位 論 文 題 名

Hydrolysis of Polyaluminum Chloride and its Effect on Coagulation Performance: Role of Inorganic Ions

(ポリ塩化アルミニウムの加水分解とその凝集性能に及ぼす影響: 無機イオンの役割)

Application of polyaluminum chloride (PACl) coagulant is a popular mode of water treatment worldwide because of the high capacity of PACl to neutralize charge. PACls with various types of characteristics are manufactured and applied in different regions around the world. However, the selection of a proper PACl still requires empirical information and field testing. It is widely recognized that even PACls with the same property sometimes performs differently depending on the water to be treated. However, little attention has been paid to the effects of raw water quality, especially the concentrations of substances not targeted for removal, such as naturally-present inorganic ions on the PACl coagulation treatment. This study comprehensively investigated PACl characteristics and inorganic ion effect on coagulation PACl coagulation treatment.

First, PACls produced by $AlCl_3$-titration and $Al(OH)_3$-dissolution methods were compared on their performance in particle removal in CSF experiments. The removal targets were particles of superfine powdered activated carbon (SPAC), which are used for efficient adsorptive removal of micropollutants, but strict removal of SPAC is required because of the high risk of their leakage after CSF. Two high-basicity PACls (HB-PACl produced by $Al(OH)_3$-dissolution and HB-PACl-t produced by $AlCl_3$-titration) were the same in terms of the ferron assay and colloid charge, but their performance in CSF were completely different. HB-PACl formed large floc particles and yielded very few remaining SPAC particles in the filtrate, whereas HB-PACl-t not form large floc particles. Because the conventional indices of PACl were not able to explain this difference, this study newly focused the rate of PACl hydrolysis during coagulation and built the test method. The hydrolysis rate of PACl-t was much slower than HB-PACl, which was the reason for its poor performance.

Second, in addition to the above two PACls, a normal basicity PACl (NB-PACl) produced by the $Al(OH)_3$-dissolution method was added to the study and further compared. HB-PACl were superior to NB-PACl in lowering remaining SPAC particles because of their high charge neutralization capacity, although their floc formation ability was similar or slightly inferior. However, HB-PACl was inferior when the sulfate ion concentration in the raw water was low.

Sulfate ions were required in the raw water for HB-PACl to be effective in floc formation. In particular, very high sulfate concentrations were required for HB-PACl-t. The aluminum species in the HB-PACl, especially those in HB-PACl-t, was resistant to hydrolysis, but sulfate ions in raw water accelerated the rate of hydrolysis and thereby facilitated floc formation. NB-PACl was hydrolysis-prone, even without sulfate ions.

Next, I investigated the floc formation rate in coagulation-flocculation process and turbidity, which is usually the target of removal, using various raw waters under different conditions of pH and PACl dose. The rates of floc formation by PACls with different basicities and polymerization degrees (HB-PACl and NB-PACl) varied as a function of the characteristics of the raw water. Those variations were related to the hydrolyzability of the PACls. Rates of floc formation were strongly correlated with the rate of aluminum hydrolysis rate irrespective of raw waters and coagulation conditions. The rate of hydrolysis, which is related to the polymerization of aluminum species, is a key property, besides charge neutralization capacity, for proper coagulation, including formation of large floc particles.

Finally, the effects and their mechanism of all the inorganic ions commonly found in natural waters on PACl hydrolysis and coagulation performances were investigated. Among the common ions in natural water, the sulfate ion had the greatest ability to hydrolyze PACl because of its divalency and tetrahedral structure. This conclusion followed from experimental results using selenate, chromate, and thiosulfate ions. Bicarbonate ions and natural organic matter affected PACl hydrolysis, but chloride ions, nitrate ions, and cations had little effect on PACl hydrolysis. The abilities of sulfate ions to hydrolyze HB-PACl and NB-PACl were very similar. Bicarbonate ions were less effective in hydrolyzing HB-PACl than NB-PACl, and bicarbonate ions contributed little to the hydrolysis of HB-PACl in raw water with normal alkalinity. Sufficient coagulation with HB-PACl therefore usually requires a certain concentration of sulfate ions. The implication is that anions have a significant effect on the hydrolysis of PACl, and thus on its coagulation ability, but which anions are most influential depends on the constituents of the PACl. Moreover, sulfate ions, bicarbonate ions, and NOM affected the rate of hydrolysis of PACl in an additive manner. The rates of PACl hydrolysis were therefore described rather well by sulfate-ion-equivalent concentrations, which represent the additive effects of sulfate ions, bicarbonate ions and NOM. The rate of floc formation was low when the sulfate-ion-equivalent concentration in the raw water was low. Sulfate-ion-equivalent concentration could be applied as an index to estimate the coagulation performance when applying PACl.