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The role of coagulation in water treatment

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Water industries globally consider coagulation/flocculation is one of the major treatment units used to improve overall treatment efficiency and cost effectiveness for water and wastewater treatment. And then fundamental and applied studies have never been ceased although the modern coagulation has been applied for water treatment since the early 1900s. This review paper then outlines recent development of novel composite coagulants and hybrid processes combining the coagulation with other treatment units, explores the properties of flocculation flocs, and introduces practical schemes for the coagulant dose control. Possible future work in the area is suggested.

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Introduction

Coagulation is a process for combining small particles into larger aggregates (flocs) and for adsorbing dissolved organic matter on to particulate aggregates so that these impurities can be removed in subsequent solid/liquid separation processes. The modern use of coagulants for water treatment started more than 100 years ago, when ferric chloride and aluminum sulfate were used as coagulants in full-scale water treatment works. The coagulation mechanism was firstly explained by the Schultz–Hardy rule and the Smoluchowski's particle collision function, which form the theoretic basis of coagulant demand and changes in particle number in flocculation process. Mattson [1] firstly derived that the hydrolysis products of Al and Fe salts were more important than the trivalent ions themselves, although this approach was widely accepted and accorded its proper position in coagulation chemistry 30 years later. Black and co-workers [2] conducted a series of studies on the effect of pH and various anions on the time of floc formation. After these early studies, the coagulation research focused on the study to produce better flocs and search for better coagulant aids including bentonite, silicates, and limestone.

Starting from the late of the 1940s, a new coagulation theory was developed by Langelier and Ludwig [3], who distinguished two mechanisms for the removal of colloidal impurities: (a) the double layer compression, a process to allow the particles to overcome the repulsive forces and thus agglomerate and precipitate; and (b) precipitate enmeshment, a process in which small particles are physically enmeshed by metal precipitates when they are forming and settling. These two mechanisms have been elaborated upon and substantiated on a theoretic basis by LaeMar and Healey [4], who proposed the terms 'coagulation' based on (a) and 'flocculation' based on (b).

A significant step in the development of a comprehensive theory of coagulation during 1960s was the introduction of micro-electrophoresis [5] to the study of colloidal destabilization which allowed the quantification of electrical charge on colloidal particles. Study of the stoichiometric relationship between the coagulant dose required to neutralize the colloids and the concentration of colloidal impurities in water also started since that decade. These studies pointed out the effect of pH, ionic strength and the properties of pollutants on the removal efficiency of colloidal particle. These studies also re-emphasized the importance of hydrolysis products of the coagulants, as originally proposed by Mattson, and established an adsorption model to detail the coagulation mechanism of hydrolysed metal coagulants.

Study of coagulation mechanism and the approaches to improve the coagulation performance are always in conjunction with the needs to match the water quality standards. This was highlighted since 1970s, when haloforms and other halogenerated organic compounds (e.g., trihalomethanes, THMs) were firstly found in the treated waters [6], which are resulted from the disinfection process due to the reactions of natural organic matter (NOM) with chlorine. Optimization of coagulation performance for the removal of NOM has been systematically studied since 1980s [7]; using various real surface waters and by the evaluation of a range of variables such as the type and dose of coagulants, coagulation pH, restabilization zones and water quality characteristics including water temperature. Another tool added to the coagulation technology was the streaming current detector (SCD) [8], which is used widely together with jar test in controlling the coagulant dose and hence, coagulation process.

The outbreak of cryptosporidiosis [9] led to the re-concern of microrganism impurities, including *Cryptosporidium*, *Giardia*, *Legionella*, and the viruses. Microbiological particulates can be controlled through a multiple barrier or integrated water treatment process design that uses coagulation, filtration and disinfection. To achieve the expected targets, coagulation is essential to ensure attachment of particles to the filter media; therefore, the microbial particulates can be effectively removed.

Coagulation/flocculation is affected by upstream treatment, e.g., pre-oxidation process, and has effects on the downstream processes — settling/flotation, filtration, activated carbon adsorption, oxidation and disinfection. Since 1990s, researches have been carried out to investigate the effect of pre-ozonation on the coagulation performance, the influence of different fractions of NOM (i.e., NOM fractions with different molecular weight, surface charge and hydrophilic/hydrophobic affinity) on their treatability by coagulation, and the impact of coagulation performance on the effectiveness of activated carbon adsorption. In addition, *enhanced coagulation*, either by addition of excess coagulant dose and lowering the coagulation pH [10,11], or through developments in coagulant chemistry to prepare the more effective coagulants [12•], have been evaluated for improving the removal of NOM and general coagulation performance.

Subsequent sections will introduce recent researches in coagulation areas, including (1) the development of new type of coagulants, especially the composite polymeric coagulants; (2) investigation on the characteristics of flocs developed in the coagulation/flocculation; (3) studies on the hybrid processes combining coagulation with other technologies for water and wastewater treatment, and (4) practical approaches on the control of coagulation.

Development of polymeric and composite coagulant

Coagulants used for water and wastewater treatment are predominantly inorganic salts of iron and aluminum. When dosed into water the iron or aluminum ions hydrolyse rapidly and in an uncontrolled manner, to form a range of metal hydrolysis species. Considerable interest and attention have been paid to preparing pre-hydrolysed metal-ion coagulants, based on either aluminum (e.g., poly-aluminum chloride), or ferric iron (e.g., polyferric sulfate), and or, a mixed polymeric coagulant (e.g., poly-aluminum-iron-sulfate). These have been shown to perform better in some cases, in comparison with conventional coagulants such as aluminum sulfate (AS) or ferric sulfate (FS) [12•].

The traditional preparation methods of pre-polymerised Al/Fe(III) coagulants consist of steps and conditions of mixing under high temperature and/or high pressure, and subsequently aging with heating and/or base addition. By controlling various preparation conditions, polymeric species can not only be produced but are also stable for a reasonable period [13].

Against the above stated method, a membrane reactor was developed in a study to prepare poly aluminum chloride (PACl) [14]. The hollow-fiber ultrafiltration membrane (UFM) was made from PPESK, a novel membrane material with properties of excellent thermal, chemical and mechanical stability. In this reactor, a sodium hydroxide solution was permeated slowly through the microspores of UFM into $AlCl_3$ containing solutions. Because the alkali drop size was reduced to extremely fine-scale, it is beneficial to dramatically reduce localized supersaturation and enhance the formation of polymeric Al species, Al_{13}.

Composite coagulants present a new type of coagulant chemicals for water and wastewater treatment. One is clay based; montmorillonites (K10 and KSF) modified by the polymeric Al or Fe and/or Al/Fe mixing polymeric species. The comparative performance results demonstrated that after being modified with mixing polymeric Al/Fe species, two montmorillonite coagulants possess greater properties to remove particles (as suspended solids) and organic pollutants (as COD and UV_{254}-abs) from the sewage and to enhance the particle settling rate significantly [15]. In another study polymeric Al modified montmorillonites were used to treat arsenic. The authors investigated the effect of purity, positive charge and special Keggin structure of Al polymer, (Al_{13}), on the arsenic removal. With increased Al_{13} content, arsenic removal was improved significantly [16].

Recent studies on the composite coagulants also include magnetically integrated inorganic polymeric coagulants [17,18]. The Fe_3O_4–SiO_2 core–shell particle and superfine iron were compounded with poly aluminum chloride (PACl). The assessment results showed that ferromagnetic nanoparticle poly aluminum performed better than original PACl in the removal of turbidity and DOC when dosed less than 0.06 mmol/L as Al [17]. Large and loose flocs were produced which were preferred for separating and recycling the magnetic powder from coagulation sludge. The magnetic composite coagulant was also prepared by magnetic nanoparticles combined with polyferric chloride, characterized in terms of structure and morphological analysis by the transmission electron microscopy (TEM), X-ray diffraction (XRD) and infra-red spectra (FT-IR) and used for the removal of *Microcystis aeruginosa* [18]. The coagulation performance was compared with polyferric chloride for the given pH and coagulant dosages. The results show that the composite magnetic coagulant exhibits improved coagulation efficiency with higher removal percentages and slighter pH dependence. The better performance could be attributed to the co-effect of magnetic nanoparticles, which sorbs *M. aeruginosa* and favors the formation of settleable flocs.