

REVIEW

Applications of Inorganic Coagulant for Typical Organic Contaminants Removal from Water

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Organic pollution has become one of the most serious environmental issues and it possesses a potential threat to human health. The coagulation-flocculation as a water treatment technology has been used in the removal of contaminant from the water. In this review, it represents the concentration of typical organic contaminants in the water as well as their treatment by traditional and novel inorganic coagulants. The coagulation mechanism is also discussed.

Keywords: Inorganic coagulant, Coagulation mechanism, Organic pollution, Water treatment.

INTRODUCTION

There are numerous organic contaminants in the water. Especially, the water is derived from the industrial sewage. Currently, some organic contaminants, such as persistent organic pollutants (POPs), endocrine disrupting compounds (EDCs), pharmaceutical and personal care disrupting compounds (PPCPs), disinfection by-products (DBPs), humic acid (HA), have attracted more attention¹. Although they are usually trace in the natural water, their potential threat to the eco-environmental and human health are not able to be ignored. And some organic pollutants are persistent in the environment, which would be more harm to human beings².

Therefore some physicochemical methods and biological methods are applied in organic pollutant treatment. For the water/wastewater treatment, there are some available technologies, such as coagulation, activated sludge, membrane bioreactor, nanofiltration/reverse osmosis, advanced oxidation process, activated carbon adsorption³⁻⁵. The latter four have a better performance in water/wastewater treatment. However, its operating cost was always higher than the conventional water treatment technologies⁶. And the removal rate of trace organic contaminants by activated sludge is very low in most of the cases⁷. So the coagulation is often used as one of the critical processes to treat these strong biodegraded contaminations due to its lower operating cost, more mature operation process and wider application conditions^{8.9}.

In the coagulation processes, the coagulant is a very important factor. Its characteristic almost determines the treatment efficiency^{10,11}. In this review, the inorganic coagulant is mainly discussed. Before that, the type, concentration and characteristic of contaminants in the water should be investigated to provide enough information to the choice of inorganic coagulant. The removal efficiency by inorganic coagulants and the coagulation mechanism are described. Some novel coagulants are mentioned due to the excellent performance in the treatment. Moreover, it is also represented the interaction among the contaminants.

Characteristic of organic contaminants: At present, more and more organic contaminants have been detected in the water, soil and air. Their physico-chemical characteristic should be studied and the international standard of maximum contamination level should be also enacted. To assess the pollution level of organic contaminants, the evaluation criterions, such as the COD, TOC and DOC, have been used. These indexes only describe the total quantity of organic contaminants. However, they do not represent the concentration of hazardous organic pollutants and persistent organic pollutants respectively. Sometimes it is necessary to monitor the concentration of every organic contaminants¹². Furthermore, the constitution of organic contaminants is complicated in the water. Therefore, it is significant to choose some emerging and poisonous organic contaminants as the research objective. The previously mentioned organic contaminants, such as persistent organic pollutants,

ADVERSE EFFECTS AND CONCENTRATIONS OF ORGANIC CONTAMINANTS IN WATER							
Name	Adverse effects	Туре	Maximum Contamination Level (From WHO)	Actual concentration	Ref.		
Toxicity POPs: disruptic produc		Aldrin+ Dieldrin	0.03 µg/L	0.015 µg/L (Nsavam)	16		
		Chlordane	0.2 µg/L	0.06 µg/L (Weija)			
	I oxicity; Carcinogen; Genotoxicity; Endocrine	DDT	1 μg/L	0.013 µg/L (Weija)			
	production: Reproductive system disease ¹⁵	Endrin	0.6 µg/L	0.015 µg/L (Nsavam)			
	production, reproductive system discuse	HCB	-	0.111 ng/L (Seomjin River)	17		
		PFOS	-	<0.017-27 (Valencia area)	18		
PPCPs Endocrin		Naproxen	-	1.17 µg/L (River Ely)	21		
		Paracetamol	-	178 μg/L (River Ely)	21		
		Ibuprofen	-	2.30 µg/L (River Ely)	21		
	Endocrine disruption; Antimicrobial resistance ^{19, 20}	Diclofenac	-	12.7 µg/L (River Ely)	21		
			-	0.26 µg/L (River Ely)	21		
		Antibiotics	-	3.2-1718ng/L (Hong Kong)	22		
		Hormones	-	0.03-18.9 ng/L (U.S.)	23		
EDCs: Alt		Phthalates	-	100 µg/L (Drinking water)	26		
	Alter femal reproductive development;	Alkylphenols	-	1.52 µg/L (Jialu river)	27		
	Reduce sperm counts ^{24, 25}	Natural	-	6.8 ng/L (Brazil)	28		
		hormones					
DBPs:		Trihalomethanes	0.3 mg/L	0.26 mg/L (Rawalpindi)	31		
	Carcinogen; Toxicity ^{29,30}	Haloacetic acids	-	26.94 µg/L (WTP)	32		
		Bromoform	0.1 mg/L	5.5 pmol/L	33		
Humic		Humic acid	-	NOM: 14.5 mg C/L			
Substan Precur ces:	Precursors of DBPs; Kashin-Beck disease ^{34,35}	Fulvic acid	-	(River Oulujoki)	36		

TABLE-1

Note: '-' Represented no guideline value from WHO. Natural organic matter (NOM) is usually used to describe the concentration of humic acid (HA) and fulvic acid (FA) in the natural water roughly

endocrine disrupting compounds, pharmaceutical and personal care disrupting compounds, disinfection by-products, humic acid, has aroused worldwide concern over the recent years^{13,14}.

The classification of these organic contaminants is not

unique. For example, some organic contaminants not only belong to EDCs, but also belong to POPs. In the Table-1, the characteristic of organic contaminants is described. Most of them are trace in the natural water and they are on the order of ng/L or µg/L. It will be a challenge to treat the contaminant of trace concentration level by coagulation. And the choice of coagulant, the aluminum speciation in hydrolysis and the coagulation condition influence the coagulation performance. Though these organic contaminants are always trace in the environment, they seriously threaten the human health for toxicity, carcinogen, genotoxicity, endocrine disruption. Or they would be toxicity after oxidation during disinfection process.

Removal of organic contaminant: Aluminium-based and ferric-based coagulants have been widely used in the treatment of the organic contaminants (Table-2)³⁷. And the removal of typical organic contaminants by coagulation is always coremoved with natural organic matter³⁸. But when only one type of contaminant exists in the water, it is not always efficiency due to the weaker co-precipitation³⁹. To improve the removal rate, the enhanced coagulation is used to treat the trace organic contaminants. Enhanced coagulation can increase the amount of aluminum hydroxide flocs and it provides more available surface area for adsorption or enmeshment of the trace organic contaminants⁴⁰. The result showed that the surface area of aluminum hydroxide flocs freshly formed at a hydrolysis ratio

of 2.5 is 597 m²/g⁴¹. However, the increasing sludge significantly enhances the operating costs.

Another method to improve the removal efficiency is combination of multiple water treatment technologies. For instance, inorganic coagulants combine with high molecular organic polymer to improve the flocculation, since flocculant can make up for the deficiency of inorganic coagulant in the adsorption, bridging and enmeshment. The better performance is obtained to treat the trihalomethane formation potential⁴², fulvic acid⁴³, humic acids⁴⁴ (Table-2). The dosage of flocculant is often low and it also decreases the dosage of coagulant⁴⁵. Moreover, due to the effect of combination, the residual monomer and the residual aluminum are usually lower than they are used separately⁴⁶. Besides, carbon nanomaterials and powdered activated carbon as the adsorbents which improve the removal rate of typical organic contaminants is often used with coagulation. The results showed that the initial addition of the coagulants without the carbon nanomaterials did not result in the removal of the EDCs. However, after adding the carbon nanomaterials, the dosage of aluminum sulfate decreased to 20 mg/L and the removal efficiency increased to 90.4 % 47 . In addition, it cannot neglect the role of membrane filtration in the water treatment for its special purifying capacity. However, the problem with membrane fouling should be solved urgently. Coagulation combines with membrane filtration that not only increases the removal efficiency of organic contaminants, but also forms unconsolidated cake layer so as to relieve the membrane fouling. However, there is a critical dose of aluminum that could exacerbate membrane fouling when inorganic coagulants over dose48.

There are some novel coagulants, such as Fe(VI), titaniumbased coagulants and zirconium oxychloride based coagulants, have been studied as potential water treatment chemical for further application in water treatment plant. Fe(VI) possesses the high-efficiency removal of organic contaminants, especially to trace contaminants⁴⁹. Most of the interest focuses on its oxidation to organic matter for no generation of DBPs⁵⁰. However, the study about its coagulation after oxidation is less investigated. These novel coagulants have obvious advantages, like low dosage, higher removal efficiency, robust flocs structure and nontoxic, whereas the deficiencies are mainly about the cost. Their stability in different conditions should be further discussed.

It represents that the distribution of aluminum and ferric species in water after hydrolysis always determines the removal efficiency of organic contaminants. Al₁₃ has been claimed to be the critical species with higher charges, higher molecular weight and contributes to the higher performance than traditional monomeric Al coagulants^{54,55}. And at the optimum pH range and aluminum concentration, aluminum hydroxide precipitates would be the predominant species, which corresponds with high removal efficiency⁵¹. In most of the cases, the

		REMOVAL	OF ORG	ANIC CON	TABLE-2 JTAMINANT	S BY INORGANIC COAGULANTS	5			
	Combinat			Remo	val Efficie	ency	Ref.			
Туре	Sources	Coagulant	Dosage	pН	ion	Flocs characteristics	IC	RC	RR (%)	
					DODe				(70)	
Perfluoroalkyl substances (PFASs)	Synthetic surface water	Alum	110 mg/L	4.5-6.5	-	Fractal and highly porous aggregates made up of many primary particles; high surface area; Larger and settleable flocs (longer flocculation time).	0.4 μM	-	28	40
					PPCPs					
Diclofenac	Selected pollutants add to wastewater	Fe(VI)	10 mg Fe/L	7.0	Oxidation process of Fe(VI)	-	100 μg/L	-	>99.3	25
					EDCs					
Nas-oxidized	Oil sands process affected water	Alum	250 mg/L	6.5-7.4	Flocculati on (PolyDA DMAC)	Easy breakage of the flocs at the mixing rapid of 220rpm.	31.6 mg/L	4.3 mg/L	86.4	51
					DBPs					
CHCl ₃ CHCl ₂ Br	Natural water from Myponga Reservoir	Polymeric aluminium	2.16Al mg/L	-	(Chitosan) [Chi]:[Al] = 0.8	The size of flocs grew 155 μm; Increasing the dosage of Al did not enhance the flocs obviously. Scattering exponent: 1.43.	172 μg/L 186	49 (88) μg/L 36 (9)	71 (48) 80	42
CHClBr ₂	(Adelaide, Australia)					The size of flocs formed by PACI-Chitosan coagulant was larger (about 180 μm); Scattering	μg/L 55 μg/L	μg/L 14 (5) μg/L	(95) 74 (90)	
CHBr ₃						exponent. 1.00.	3 11g/I	9 (8) цо/L	-	
Humic Substances				μ <u></u>	P-8-					
DOC (FA)	Simulated water	Titanium tetrachloride	50 mg/L	4.37	(CPAM: 3 mg/L)	TiCl ₄ and CPAM yielded higher floc growth rate (550 μm/min), larger floc size (800-850 μm), the flocs with more open structure, resisted cycles of shear, reformed larger flocs. The relative S _f , R _f , D _f was stated in this manuscript.	4.82 ± 0.3 mg/L	-	61.7 (>80)	43
2,6-DCBQ	Otonabee River (Peterborough, Canada)	Aluminum sulfate	2.2 mg/L	8.0	-	-	3.21 mg/L	-	69	52
НА	Landfill leachate (Changsha, China)	Ferric chloride	10 g/L	8.0	-	The structures of precipitates using ferric chloride had larger aggregates in comparison with the one using ferric sulfate.	598 ± 2.94 % mg/L	130 mg/L	78.37	44
НА	Simulated water	Alum	0.1 mM	7.93 ± 0.12	Ultra filtration	Flocs in sub-CUF were significantly larger than it in CUF, and with less small flocs; dramatically higher growth ability of flocs; D _f (sub-CUF): 2.53, D _f (CUF): 2.65.	4.4 ± 0.3 mg/L	2.4 ± 0.3 mg/L	52 ± 4.5	53

Note: IC = Initial Concentration; RC = Residual Concentration; RR = Removal Rate; S_f = strength factor; R_f = recovery factor; D_f = fractal dimension; 2,6-DCBQ = 2,6-dichloro-(1,4)benzoquinone. The bracket stated the processes combined with coagulation, and the relative removal efficiency of coagulation-combination processes. Without the bracket, it was only coagulation process, and its removal efficiency.

surface of the precipitates is positively charged. It contributes to the destabilization of the colloidal particles through charge neutralization⁵⁶. Moreover, some studies show that adsorption is the main coagulation mechanism for the removal of the organic contaminants, especially trace organic contaminants⁴⁰. The organic contaminants could adsorb on the surface of the colloidal particles. And it is discussed that there is a complexation between the aluminum and trace organic contaminants. For these reasons, the organic contaminants would be removed through co-precipitation with hydroxide precipitates⁴⁰. In the precipitation process, the enmeshment could also contribute the organic contaminants removal. The characterization of floc, such as growth rate, size, strength, compactness, recoverability and the fractal dimension, formed by coagulation and flocculation is another important factor to influence the organic contaminants removal⁵⁷. Floc aggregation not only depends on coagulants (type, dose range, forms of hydrolyzates, etc), but also the hydraulic parameters (solution pH, shearing force, etc)⁵⁸. The floc size and strength are sufficiently relevant with coagulant type and dosage. The floc recoverability decreases with the increasing dose. Some novel coagulants have larger floc size and they are less affected by solution pH than traditional coagulants. However, titanium tetrachloride yields the flcos with the weakest recoverability comparing with ferric chloride and aluminum sulfate⁵⁷. The floc growth rate of zirconium oxychloride bases coagulant is lower than the aluminum sulfate. It takes longer time to reach the steady state size⁵⁹.

Simultaneous removal of inorganic and organic contaminants: As the previous statement has represented, simple pollutant is hardly removed by coagulation for the weaker coprecipitation. It would contribute to the organic contaminants removal when colloidal particles are formed in the water. The mutual effect among the coagulants, inorganic contaminants and organic contaminants is complicated. In case of the colloidal particles are generated via the reaction among them, the removal efficiency is higher than the contaminants are treated separately⁶⁰. Additionally, at low coagulant dosages, organic matter would compete with inorganic contaminants for the sorption sites, which leads to lower removal rate of organic contaminants. However, the influence is hardly discovered at high coagulant dosages. The removal rate of organic matter is consistent with that of inorganic matter⁶¹. Nevertheless, the organic matter, such as iminodiacetic acid and EDTA, bonds with inorganic matter through chelation and the compounds show high solubility. It decreases the contaminant removal significantly^{62,63}. To cope with this problem, the coagulation is combined with oxidation to improve the removal rate of pollutants, or the other water treatment technologies are applied as the post-processing.

Conclusion and future scope of research: Coagulation as the conventional water treatment technology has been widely applied for its obvious superiority. The current studies focus on the coagulation mechanism, emerging contaminant removal by it and green coagulant synthesis (including the novel coagulant).

It is not always efficient to remove the trace organic contaminants by coagulation from the water. The methods of enhance coagulation and combination with other technologies are used to improve the removal efficiency. The main coagualtion mechanism of typical organic contaminants is adsorption, charge neutralization and enmeshment. Moreover, the performance of the coagulation-flocculation should be further discussed at different environmental condition. And it is not cost-effective to adjust pH in the water treatment plant due to the buffer capacity of water even though the optimal pH is easily achieved in the jar test. There must be a transition from the small-scale coagulation test to the field application. The new research achievements of coagulation-flocculation and the novel coagulants should be applied in the practical application.

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