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The removal characteristics of natural organic matter in the recycling of drinking water treatment sludge: Role of solubilized organics

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ABSTRACT

To clarify the role of solubilized organics derived from drinking water treatment sludge (DWTS) in the elimination of natural organic matter (NOM) in the DWTS recycling process, a probe sonoreactor at a frequency of 25 kHz was used to solubilize the organics at varied specific energies. The coagulation behavior related to NOM removal in recycling the sonicated DWTS with and without solubilized organics was evaluated, and the effect on organic fractionations in coagulated water was determined. The study results could provide useful implications in designing DWTS recycling processes that avoid the enrichment of organic matter. Our results indicate that DWTS was disrupted through a low release of soluble chemical oxygen demand (SCOD) and proteins, which could deteriorate the coagulated water quality under the specific energy of 37.87–1212.1 kW h/kg TS. The optimal coagulation behavior for NOM removal was achieved by recycling the sonicated DWTS without solubilized organics at 151.5 kW h/kg TS specific energy. Recycling the sonicated DWTS could increase the enrichment potential of weakly hydrophobic acid, hydrophilic matter, and <3 kDa fractions; the enrichment risks could be reduced by discharging the solubilized organics. Fluorescent characteristic analysis indicated that when recycling the sonicated DWTS without solubilized organics, the removal of humic-like substances was limited, whereas removal of protein-like substances was enhanced, lowering the enrichment potential of protein-like substances.

1. Introduction

Drinking water treatment sludge (DWTS), the byproducts generated at water treatment plants that is primarily composed of amorphous Fe/Al hydroxides, is generally produced during flocculation–sedimentation or flotation processes because Fe/Al-based coagulants are usually used. DWTS and solids within filter backwash water (FBWW) are the major components of water treatment residual. Okuda et al. [1] have reported that global production of solid residuals "might be totally 10,000 tons per day," of which European countries including Ireland, Germany, the Netherlands, United Kingdom, and Portugal make up 10.38%, and the United States and Chinese Taiwan account for 72.6% and 0.003%, respectively [2]. An increased generation of water treatment residues, coupled with environmental restrictions on disposal methods, has led to increased research into their reuse, which is an important avenue in realizing a reduction and reclamation of total waste residues. There is a general understanding of the role of magnetic seeding, micro-sand, diatomite, and recycled DWTS in enhancing coagulation. Magnetic seeding and micro-sand assisted flocculation are now widely accepted technical alternatives to strengthen separation during the treatment of drinking water, reclaimed water, or landscape water [3–8]. However, the application of recycled DWTS from drinking water treatment is restricted due to potential negative effects of complex components within the recycled DWTS. A major issue influencing the water quality in the recycling process is the release of extracellular or intracellular organics that originate from the DWTS [9]; therefore, an effective organic solubilization is key in the DWTS recycling process.

Several approaches to sewage sludge solubilization have been investigated, including thermal, chemical, acoustic-mechanical disintegration, and a combination of chemical and acoustic-mechanical methods [10–15]. Ultrasound can cause a series of compression and rarefaction cycles, which generate cavitation bubbles. Millions of these bubbles implode yielding localized temperatures as high as 5000 °C, pressures of 100 MPa and free radicals, e.g., $^{\cdot}OH$, $^{\cdot}HO_2$, and $^{\cdot}O$ [16]. The synergetic effects of high mechanical shear force, radical reactions, and thermal breakdown can cause the organic solubilization, the destruction of microbiological cells, and the oxidation of toxic chemical compounds [16].

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In previous studies [9,17], we have proposed ultrasound as a pretreatment in the coagulation process of recycling DWTS. We observed that ultrasound could effectively solubilize extracellular and intracellular organics, which was confirmed by the release of soluble chemical oxygen demand (SCOD), proteins, and polysaccharides, and the increase in fluorescent intensity of humic-like and protein-like substances. Meanwhile, we found that the coagulated water quality worsened after the addition of sonicated DWTS containing solubilizing organics, as compared to conditions without solubilized organics. We assumed that the solubilized organics adversely affected the coagulation efficacy of the DWTS recycling process. However, according to Liu et al. [18], the extracellular polymeric substances (EPS) of aerobic and anaerobic sludge, mainly composed of carbohydrates, proteins, and humic substances, had the same properties of bio-flocculant and could thus play a key role in sludge aggregation. The researchers pointed out that the loosely bound EPS always displayed a positive effect on the sludge aggregation as indicated by negative interaction energy. Furthermore, Yuan et al. [19] assumed polysaccharides in EPS excreted from *Bacillus megaterium* TF10 exhibited a high flocculation activity, whereas the proteins had no flocculation ability, conflicting with the findings of Takeda et al. [20], in which proteins in EPS extracted from *Nocardia amarae* did have flocculation ability. Additionally, Wen et al. [21] found that the addition of aluminum ions (Al^{3+}) could improve the flocculation and turbidity removal of activated sludge with an increasing single dosage, but deteriorate the sedimentation of activated sludge. To the best of our knowledge, the effects of extracellular or intracellular organics derived from DWTS on the flocculation behavior of DWTS have not been studied, and the effects on natural organic matter (NOM) removal in the DWTS recycling process remains unclear.

On the other hand, results of our previous investigation [9] have demonstrated that the residual dissolved organic carbon (DOC) and the residual UV at the wavelength of 245 nm (UV_{254}) in the coagulated water after recycling sonicated DWTS without solubilized organics, regardless of the ultrasound dose used, were lower than incidences with solubilized organics. However, these indicative parameters provided no insight into the composition and distribution of NOM. The common NOM properties are molecular weight (MW) distribution, hydrophobicity, and fluorescence [22]. Generally, lower-MW NOM tend to be more hydrophilic and biolabile, while higher-MW NOM tends to be more aromatic and hydrophobic, with a higher adherence to adsorption [23]. During the traditional coagulation step for drinking water treatment, the hydrophilic, low molecular weight (LMW), and protein-like compounds of NOM appear to be less efficiently removed than the hydrophobic, high molecular weight (HMW), and humic-like compounds; after the coagulation step, LMW, hydrophilic, and protein-like NOM are dominant in the residual organic matter [22–24]. In our previous study [25] of recycling FBWW to enhance coagulation of two low-turbidity source waters, we observed that high organic concentrations and increased low-MW fractions within the recycled FBWW did not enhance UV_{254} and DOC removal. Hydrophobic acid could be further eliminated when recycling particles produced mainly by sweep flocculation. Weakly hydrophobic acid and a hydrophilic fraction removal could be enhanced through recycling particles formed mainly by charge neutralization. Moreover, recycling FBWW could effectively improve the removal rate of humic-like substances, but protein-like substances were resistant to elimination.

Currently, little research exits on the NOM removal characteristics of the DWTS recycling process, and the effects on organic fractionations in coagulated water also need to be investigated further. In this study, to clarify the role of solubilized organics in the elimination of NOM in the DWTS recycling process, a probe sonoreactor at a frequency of 25 kHz was used to solubilize the extracellular or intracellular organics. The coagulation behavior during the recycling process with and without solubilized organics was then evaluated, particularly NOM removal, measured as UV_{254} and DOC removal. The XAD-4 and -8 resin adsorption and ultrafiltration technique, as well as three-dimensional excitation emission matrix (3D-EEM) fluorescence spectroscopy, were employed to fractionate the NOM.

2. Materials and methods

2.1. Raw water and DWTS used in the experiments

The raw DWTS sample used in the experiments was collected from a water treatment plant (Beijing, China) that handles 1,500,000 m^3/day using a coagulation, flocculation, sedimentation, and sand filtration process. Poly-aluminum chloride and ferric chloride ($FeCl_3$) were used as the coagulant. The alum and ferric sludge settled in the sedimentation basin and was discharged to the thickener. The DWTS samples were transferred immediately to the laboratory and stored at 4 °C. Prior to each experiment, sludge samples were warmed to room temperature. All of the experiments were performed within a week of sampling. As indicated in the Table 1, the characteristics of the raw DWTS were as follows: total solids (TS); volatile suspended solids (VSS); total chemical oxygen demand (TCOD) concentrations of raw DWTS, which were 1.10 ± 0.12 g L^{-1}, 0.26 ± 0.03 g L^{-1}, and 423.29 ± 15.91 mg L^{-1}, respectively; the SCOD; and the proteins in the supernatant of raw DWST, which were 15.36 ± 1.71 mg L^{-1} and 4.75 ± 0.84 mg L^{-1}, respectively. A low VSS to TS ratio in the raw DWTS, 23.64%, indicated a primarily inorganic makeup. The SCOD to TCOD ratio in the raw DWTS was 3.63%, indicating that a small proportion of the COD was associated with the soluble phase. The polysaccharide in the supernatant of raw and sonicated DWST was not considered, as it was detected at very low levels.

Raw water was collected from the same water treatment plant as the raw DWTS samples. The main physicochemical characteristics of the raw water were as follows (seen in Table 1): pH 8.21 ± 0.12; turbidity 0.774 ± 0.241 NTU; UV_{254} 0.039 ± 0.009 cm^{-1}; and DOC 6.003 ± 1.212 mg/L. The average specific UV absorbance, calculated as 100 times UV_{254} divided by DOC concentration, was 0.75 L/mg m, indicating the NOM was generally enriched in hydrophilic and LMW components and were difficult to eliminate through a traditional coagulation method [26].

Table 1
Main physicochemical characteristics of raw water and DWTS.

Analytes (Units)	Raw water Mean ± SD	Analytes (Units)	Raw DWTS Mean ± SD
Temperature (°C)	12 ± 0.5	Temperature (°C)	13 ± 0.5
pH	8.21 ± 0.12	TS (g L^{-1})	1.10 ± 0.12
Turbidity (NTU)	0.774 ± 0.241	VSS (g L^{-1})	0.26 ± 0.03
Ultraviolet absorbance at 254 nm (UV_{254}) (cm^{-1})	0.039 ± 0.009	VSS/TS (%)	23.64
Dissolved organic carbon (DOC) (mg/L)	6.003 ± 1.212	TCOD (mg L^{-1})	423.29 ± 15.91
Specific UV absorbance (SUVA) (L/mg m)	0.75 ± 0.15	SCOD in supernatant (mg L^{-1})	15.36 ± 1.71
Zeta potential (mV)	−14.9 ± 1.4	SCOD/TCOD (%)	3.63
		Proteins in supernatant (mg L^{-1})	4.75 ± 0.84

Note: SD means standard deviation. Number of measurements (n): for temperature and pH, $n = 5$; for total solids (TS), volatile suspended solids (VSS), total chemical oxygen demand (TCOD), sCOD and proteins, $n = 10$.