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# Influence of extracellular polymeric substances (EPS) treated by combined ultrasound pretreatment and chemical re-flocculation on water treatment sludge settling performance

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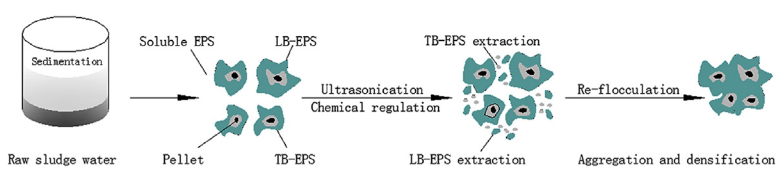
## HIGHLIGHTS

- A method is proposed to improve settleability of sludge water from waterworks.
- Combining ultrasound pretreatment and chemical re-flocculation is studied.
- Ultrasound treatment is capable of destroying extracellular polymeric substances.
- The operating parameters to achieve the optimal settling effect are obtained.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Extracellular polymeric substances (EPS) are high molecular weight polymers and play a significant role in floc stability, floc size, bioflocculation and sludge settleability. The destruction and reconstruction of EPS improve the performance of solid-water separation processes. In this study, the influence of combined ultrasound pretreatment and chemical re-flocculation on the spatial distribution and composition of EPS was examined. Settleability efficiency demonstrated that the optimal operating condition was an ultrasound pretreatment time of 15 min at pH 6. Sludge particles were greatly disintegrated and the protein-like substances were converted into smaller molecules after ultrasound treatment, and pH had important effects on solubilization and degradation of protein-like substances. The flocs of sludge water after addition of polyacrylamide were larger in size and denser in structure than those resulting from addition of polyaluminium chloride. However, polyaluminium chloride had a better capacity for degrading EPS, especially at a dosage of 1.2 g/g total suspended solids. The results of this research show that the combination of ultrasonication and chemical re-flocculation is effective in treating sludge water from a drinking water treatment plant.



## 1. Introduction

With water pollution becoming more serious, waterworks sludge treatment projects have received increased attention in an effort to achieve a zero discharge of wastewater. In a drinking water plant sludge mainly comes from the wastewater. Waterworks

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wastewater includes filter backwash water and sedimentation tank sludge water, and accounts for 4–7% of the total water inflow. There is a large quantity of suspended solids in waterworks wastewater including sediment, algae, bacteria and other material. When waterworks wastewater is discharged directly (i.e., without treatment), it wastes resources and causes environmental pollution. Therefore, the necessary direction of waterworks operation is the treatment and comprehensive utilization of wastewater.

In the waterworks sludge treatment process, dewatering is usually given more attention than the settling of sludge water, especially in conventional treatment units. In fact, the settling of sludge water is as vital a part of sludge treatment as dewatering. Poor sludge separation properties often increase the difficulty of subsequent sludge management operations and increase the disposal cost (Neyens et al., 2004). Poor sludge separation properties have a variety of origins, such as sludge characteristics including composition and structure (Jin et al., 2003), poor flocs formation and stability (Bache and Gregory, 2010), and high levels of extracellular polymeric substances (EPS) (Subramanian et al., 2010). Besides these aspects, sludge settling efficiency mainly depends on the selection of conditioning chemicals and settling methods.

Extracellular polymeric substances are high molecular weight polymers produced by microbial metabolism and may originate from bacterial secretion, hydrolysis products, ions adsorbed from wastewater and organics adsorbed on the flocs in the wastewater. The total mass of EPS reaches 80% of the total sludge mass (Chen et al., 2015). EPS content was found to greatly affect surface charge, floc stability and floc size (Mikkelsen and Keiding, 2002). Lin et al. (2016a) illustrated the effect of EPS constituents in the particle aggregation process using titanium oxide nanoparticles ($TiO_2$ NPs) and found that protein-like substances affected the stabilization of $TiO_2$ NPs through the combination of electrostatic and steric repulsion while the polysaccharide enhanced aggregation through intermolecular bridging and electrostatic forces (Lin et al., 2016a). Many previous studies also have demonstrated that sludge settling, bioflocculation and dewatering properties have a great relationship with the contents of EPS and the spatial distribution of EPS (Peng et al., 2014; Zhang et al., 2014). The ratio of protein (PN) to polysaccharide (PS) is well correlated with sludge settling (Murthy, 1999), and a high PN/PS ratio also has significant effects on sludge settling and dewatering (Higgins and Novak, 1997). More recently, some researchers have separated the slime EPS, loosely bound EPS, tightly bound (TB) EPS and the pellet fractions of EPS from the sludge floc (Yu et al., 2008). The study found that LB-EPS has a greater effect on sludge flocculation, sludge settleability and dewaterability than does the TB-EPS; the examination used a modified heat extraction method that is especially correlated with the concentration of TB-EPS (Li and Yang, 2007a). LB-EPS on the bacterial surface could enhance the cell-cell interactions and plays a more important role than TB-EPS in the aggregation processes (Zhao et al., 2015). Yu et al. (2008) reported that PN and PS have different distributions in sludge flocs resulting from combined ultrasound and centrifugation treatments (Yu et al., 2008). The distribution of PN and PS markedly impacts sludge water separation performance due to the content of PN and PS in different fractions.

Traditional chemicals including organic polymers and inorganic coagulants can help tiny sludge particles flocculate into larger flocs by neutralizing surface charges and bridging to accelerate the settlement process (Niu et al., 2013). However, various alternative methods have been developed to improve sludge settleability. These methods can disrupt flocs and cells and solubilize EPS components to accelerate solid-water separation, and include Fenton's reagent and Fenton-like process pretreatment (Liu et al., 2013; Xu et al., 2016), ultrasound treatment (Gong et al., 2015), the addition of acid or alkali (Chen et al., 2001) and enzymatic treatments (Ayol, 2005). Some researchers investigated ultrasonication was a useful method for sludge disintegration (Khanal et al., 2007; Xin et al., 2009). The ultrasonic treatment is performed through the cavitation effect including mechanical effect, thermal effect and sonochemical effect. Hydro-mechanical shear forces were shown to have a crucial role in disintegration (Wang et al., 2005). Ultrasonic energy could disintegrate sludge flocs and disrupt the walls of cells, which promoted the release of the substances internal or external to sludge flocs and in the cells. High energy input during ultrasonic treatment not only destroyed floc structure and cell membranes, but also minimized the floc size. Furthermore, ultrasonic pretreatment enhanced sludge settleability and the settling effects changed as ultrasonic density, treatment time and the amount of treated sludge varied (Wolski and Zawieja, 2012).

Many studies have concluded that ultrasonic treatment improves sewage sludge settling performance and the influence of EPS on activated sludge settleability. Few studies have focused on the settleability of sludge water in waterworks, especially on the influence of ultrasonication treatment and EPS distribution and composition. The use of ultrasonic treatment combined with chemical re-flocculation by different kinds of coagulants for sludge water in waterworks has not been well investigated. As a consequence, the aims of this study were to assess whether ultrasonic pretreatment improves sludge water settleability and to understand the relationship between EPS and sludge water settling performance. The specific objectives of this study were: (1) to examine the effect of ultrasonic pretreatment on the EPS properties of sludge water; (2) to understand the effects of pH on sludge water physicochemical properties after treatment with ultrasonication; and (3) to obtain further insight into the floc structure as well as soluble EPS characteristics under chemical flocculation.

## 2. Material and methods

### 2.1. Materials

Samples of sludge water were collected from the drinking water treatment plant in Wuxi, Jiangsu Province, China. The sludge water samples were retrieved from the horizontal flow sedimentation tank mud. The sludge water samples were stored at 4 °C prior to use. The basic properties of the sludge water are listed in Table 1, and the content and varieties of elements in the sludge are shown in Table 2. All the chemical reagents used were analytical grade.

### 2.2. Sludge water treatment with ultrasonication

A 500-ml suspension of sludge water was prepared in 1000-ml beakers and mixed fully before sonication. Then, the suspension was placed into the ultrasonic cell disruptor (JY98-IIIN, Ning Bo Scientz Biotechnology Co., LTD.). Meanwhile, the probe was extended 1 cm below the liquid level and placed in the center of the beaker. Some batches of sludge water were treated with a fixed ultrasonic energy intensity at 1.2 W/ml. Both ultrasonic time and interval time were 5 s and the work times were controlled to adjust the ultrasonication energy by the exposure time (from 0 min to 30 min). Then, the suspension was prepared for the distribution and composition of different EPS fractions. During the ultrasonic process, the temperature of the solution increased as the ultrasonic treatment time increased. To address this effect, the treated beaker was immersed in an ice bath to ensure that the temperature was not more than room temperature after ultrasonication, thereby minimizing or avoiding the impacts of increasing temperature