



Rapid removal of bound water from dredged sediments using novel hybrid coagulants

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ABSTRACT

Dredging operations produce sediment masses that require chemical conditioning and dewatering before dewatering of dredged sediments. Conventional chemical conditioners have difficulty removing the bound water parceled in extracellular polymeric substances (EPS), and the removal of bound water determines the dewatering ratio and the rate of sedimentation. In this study, the conventional conditioners FeCl_3 , $\text{Al}_2(\text{SO}_4)_3$, and cationic polyacrylamide (CPAM) and a new covalently-bound hybrid coagulant (CBHyC) were investigated for dewatering sediments in laboratory conditions that simulated real-world conditions. The water content of dewatered cakes was 61.4–68.3% for sediment conditioned with FeCl_3 , $\text{Al}_2(\text{SO}_4)_3$, and CPAM, while the water content was 52.6% with CBHyC. CBHyC achieved a setting rate that was 66–359% faster than conventional conditioners, decreased specific resistance to filtration (SRF) by 72–86% compared with conventional conditioners. CBHyC achieved the best sediment dewaterability due to its structure, which consists of hydrophobic group of a long carbon chain and hydrophilic groups of Fe-O-Si complexes and quaternary ammonium. This structure increases electrical neutralization ability and bridging effects, allowing formation of large dense flocs in sediments. Moreover, CBHyC also functioned as a surfactant, dissolving EPS (approximately 59.7%), especially proteins (approximately 59.9%) and humic substances (approximately 25.4%), from the sediment into water. The result was approximately 37.1% bound water parceled in EPS was transformed into free water. Lastly, the sediment conditioned with CBHyC exhibited a discontinuous and porous structure; thus, the water was more likely to flow quickly outward from interior locations. These results demonstrate the potential application of CBHyC for high efficiency and rapid dewatering of dredged sediments.

1. Introduction

Sediment dredging helps to maintain hydrological functions and improve the water quality of rivers, lakes, and other aquatic ecosystems. The process produces dredged sediment masses with high contents of water and pollutants, which could cause secondary pollution to the surrounding environment if not properly disposed. Dewatering is the first step in any sediment disposal technology. Present dewatering technologies include mechanical dewatering [1,2], biological dewatering [3], and chemical conditioning dewatering [4]. As riverway pollution increases, sediment compositions become more complex, and the increase of organic matter makes dewatering more difficult and prolongs the dewatering time. In this case, mechanical and biological dewatering processes are not effective, and chemical conditioning

becomes the primary dewatering technology. As such, is necessary to study the conditioners to determine those most effective for rapid dewatering of sediments.

The chemical conditioners used for sediment dewatering are mainly inorganic compounds such as iron salts and aluminum salts and organic polymers such as polyacrylamide (PAM) and poly dimethyldiallylammonium chloride (PDMDAAC). Sediment particles aggregate with inorganic compounds by charge neutralization between their positive charges and the negative charges of sediments surface [5], or organic polymers through adsorption and bridging effects [6]. This process forms larger flocs, which accelerate the separation of water from solids. However, the dewatering time is long, and the water content of sediment cakes is still high after conditioned with these conventional chemicals. This is because conventional conditioners can only reduce the

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free water and are not able to release bound water from the sediments. Bound water mainly exists in complex colloids formed by extracellular polymeric substances (EPS). High EPS presence has a negative effect on sediment dewatering [7,8]. A critical factor to improve the sediment dewaterability, especially to release more bound water from sediment surfaces, is to decrease EPS [9,10]. Prior studies show some surfactants dissolve EPS from the sediment surfaces into water, thereby releasing bound water, reducing viscosity of the sediments, and strengthening the sediment mechanical dewatering performance [11,12]. At present, there is little research on the use of conditioners in dredged sediment dewatering, especially those conditioners that can remove EPS and rapidly decrease bound water.

Covalently-bound hybrid coagulant (CBHyC) is a new type of coagulant that can effectively remove more low molecular weight organic contaminants from water. It is prepared by combining aluminum or iron salt with a kind of quaternary ammonium salt through covalent bonding. The resulting coagulant has the advantages of inorganic coagulants (high positive charge density) and organic coagulants (high molecular weight) [13]. CBHyC also functions similar to surfactants due to the hydrophobic components of the long carbon chain and the hydrophilic components of aluminum/iron-silicon compounds and quaternary ammonium [14]. Therefore, CBHyC behaves like conventional coagulants to remove free water by charge neutralization and bridged effect, and it also behaves like surfactants to remove bound water. As such, it is worthy to explore CBHyC with respect to dewatering performance and dewatering mechanism.

In this study, dredged sediments from Maozhou river, a highly-polluted river in Shenzhen, China, were used to test the dewatering properties of conventional conditioners (FeCl_3 , $\text{Al}_2(\text{SO}_4)_3$, and CPAM) and a new chemical conditioner (CBHyC) under simulated real-world conditions. The study focused on the dewatering mechanism of CBHyC by measuring and analyzing the changes in EPS, EPS fractions, and bound water in sediments during the dewatering process.

2. Materials and methods

2.1. Dredged sediments

Raw sediments (RS) used in this study were sampled from Maozhou river, Shenzhen, China, and had the characteristics listed in Table 1. Samples were transported to the laboratory in polypropylene containers. The branches and stones were removed from the samples before storing at 4 °C.

2.2. Chemical conditioners

CBHyC, the chemical conditioner, was synthesized according to a modification of the method described in previous study [14]. The major modification was the replacement of aluminum salt with iron salt during the synthesis. Briefly, trimethyl 3-(triethoxysilyl) propyl ammonium chloride (Bailingwei, Beijing, China), 1.0 M FeCl_3 solution (calibrated), and deionized water were mixed in a 500-mL flask at the Si/Fe molar ratio of 0.4, in which trimethyl 3-(triethoxysilyl) propyl ammonium chloride was used as Si source. Under rapid stirring, the

Table 1
Characteristics of raw sediments.

Density (g/mL)	Water content (%)	pH	Organic matter (% DS [*])	D ₅₀ (μm)	Zeta potential (mV)	TSS [†] (g/L)	VSS/VSS [‡] (%)
1.1	85.6	7.9	14.3	14.2	−38.1	143.5	14.6

^{*}DS dry solids.

[†]TSS total suspended solids.

[‡]VSS/TSS the ratio of volatile suspended solids to total suspended solids.

mixture was titrated by NaOH (2.5 M) to the target basicity (OH/Fe molar ratio) of 1.5. Then deionized water was added to achieve a total iron concentration of 0.2 M. CBHyC was obtained after aging for 24 h.

Conventional conditioners $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$ were purchased as analytical-grade reagents (Sigma-Aldrich, St. Louis, MO). Cationic polyacrylamide (CPAM) was purchased as analytical-grade reagents (Shanghai Chemical Reagent Co., China). Deionized water was used in all solutions.

2.3. Sediment dewatering

For dewatering experiments, 300 mL of sediments were transferred into 500-mL beakers, and different doses of chemical conditioners were added to the beakers. Dosages were determined using the solid contents of the chemical conditioners. Sediment samples were mixed using an agitator as follows: a slow mix for 5 min at 150 rpm followed by a rapid mix for 10 min at 250 rpm.

After conditioning, the conditioned sediments were sampled to measure the bound water and EPS contents, and 100 mL of the conditioned sediment samples were transferred to graduated cylinders. The change in sediment volume with precipitation time was observed. After 2 h sedimentation, specific resistance to filtration (SRF), zeta potential, and particle sizes were determined. After 6 h sedimentation, the sediment samples were vacuum filtered for 2 min under a pressure of 0.1 MPa, and the water content, composition, and structure of dewatered sediment cakes were determined.

2.4. Bound water content measurement

The bound water content was measured using a differential scanning calorimeter (DSC) (TA, USA). It is reported that the bound water does not freeze at −20 °C [15]. In this study, sediment samples (approximately 5–15 mg) were subjected to a temperature of −20 °C to freeze all free water. The samples were then heated at 2 °C/min until reaching 20 °C. The amount of free water was determined by the area of the heat absorption curve and calculated by Eq. (1). Then, the bound water content was determined by calculating the difference between the total water (measured by gravimetric method) and the free water.

$$\text{FW} = K \times A \quad (1)$$

where FW is the mass of the free water in sediments (mg), A is the area of heat absorption curve (mJ), and K is the conversion coefficient (mg/mJ) calculated by the thermal analysis curves of pure water with known quality.

2.5. EPS characterization

To measure EPS in this study, the sediments were first centrifuged in a 50-mL tube at 3000g for 5 min. The centrifugal liquor (CL) was used for analysis. The sediment pellets in the tube were used in a heat extraction method to extract soluble EPS (SEPS), the loosely-bound EPS (LB-EPS), and the tightly-bound EPS (TB-EPS).

The CL, SEPS extract, LB-EPS extract, and TB-EPS extract were filtered with polytetrafluoroethylene membranes with a pore size of 0.45 μm prior to organic analysis. TOC was measured by a TOC analyzer (Multi N/C 2100, Analytik Jena). The proteins (PN) and polysaccharide (PS) contents in the released EPS were determined by the bicinchoninic acid (BCA) assay [16] and the anthrone-sulfuric acid method with glucose as the standard [17,18], respectively. The humic substances (HS) content was measured by a UV/VIS spectrophotometer following the modified Lowry method with humic acid as the standard [19].

2.6. Other analytical methods

The dewatered sediment cakes were dried at 105 °C to determine water content. SRF was measured by a multi-coupled measuring device

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