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RAPID COMMUNICATION

ERRORS IN BOUND WATER MEASUREMENTS USING CENTRIFUGAL SETTLING METHOD

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Abstract—Centrifugal settling method (CSM) is commonly used to estimate the bound water content in sludges. The sludge sediment matrix should be purely plastic for accurate measurements to the bound water content. This communication, however, addressed for the first time that, owing to the inhibited elasticity of the sludge matrix, sediment height rebound was commonly observed for the flocculated sludge. Also, different sludges could have distinct elastic responses after centrifugation. There existed no simple correlation to incorporate the elastic rebound of sludge sediment in bound water measurement. The possible errors incorporated into CSM for measuring bound water contents were highlighted. © 2001 Published by Elsevier Science Ltd.

Key words—bound water, centrifugation, elasticity, rebound

INTRODUCTION

Owing to the presence of solid phase in the sludge, a certain part of water behaves very differently from the bulk water (Heukelekian and Weisberg, 1956). This portion of water is commonly termed as “bound water”, which resists freezing at a sub-zero temperature and is hard to be removed from the sludge body using mechanical means of dewatering (Smollen, 1990; Vesilind, 1994). Various methods have been proposed for differentiating the moisture in the sludge into bound and non-bound (free) water (Lee, 1994; Robinson and Knocke, 1992) and Lee and Hsu (1995) revealed that the bound water data measured via different techniques could be very different. Restated, the bound water is in fact an operationally defined value. Later, Lee (1996) hypothesized that the capability of bound water to resist freezing under low temperatures is attributed to the lowering of its thermodynamic activity by the water–solid bond strength. Based on this hypothesis we proposed a continuous classification scheme to differentiate the bound water in the sludge (Chen *et al.*, 1997; Wu *et al.*, 1997; Chu and Lee, 2001). The continuous classification scheme provides a physically relevant picture for describing the moisture in the sludge. However, owing to the simplicity in both experimental procedures and data interpretation, the discrete classification schemes, such as centrifugal

settling method (CSM), are still widely employed in applications.

Centrifugal settling method was originally proposed to measure the “packed cell volume” by extrapolating the sludge height against the rotational speed to $N \rightarrow \infty$ limit (Mitani *et al.*, 1983). In a lab test, a fixed amount of sludge is placed in a centrifugal tube rotated at a prescribed speed. After the sediment was sufficiently compacted, the rotational speed was declined to a complete stop with the final equilibrium sediment height being recorded. Normally, one would note a lowering sediment height at an increased rotational speed, a self-evident result. To extrapolate the sediment height versus rotational speed data to infinite rotational speed limit, if feasible, Kawasaki *et al.* (1990, 1991) and Matsuda *et al.* (1992) proposed that this *extrapolated* residual moisture in the cake represented the bound water content in the sludge. However, Lee and Lee (1995) demonstrated that the sediment height versus rotational speed plot revealed a highly nonlinear manner that put the extrapolation procedure proposed by Kawasaki *et al.* into question. To overcome this difficulty, some subsequent works simply regarded the residual moisture content existing in the sediment compacted after centrifugation as the bound water content equilibrated with the specific centrifugal field (Colin and Gazbar, 1995).

The basic assumption of using CSM for estimating bound water content is a purely plastic sludge matrix. Restated, once the cake was deformed under stress, the deformation would be retained and become unrecoverable after the stress was released. Hence,

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during the centrifuge de-acceleration phase the compacted cake would not “bound back”. However, the conventional filtration/consolidation theories commonly assumed a purely elastic filter cake (Lee and Wang (2000)). Chang and Lee (1998) demonstrated that the sludge filter cake collected from wastewater treatment plant exhibited an apparent visco-elastic characteristic under compaction. Chu and Lee (1999) demonstrated that the elasticity of the waste activated sludge matrix would be markedly deteriorated after polyelectrolyte flocculation. Also, Wu *et al.* (2001) noted that at the consolidation pressures lower than a threshold pressure the sludge matrix is mainly elastic and the deformation would be mostly recoverable. Exceeding such a threshold pressure, on the other hand, the sludge matrix would largely yield, while the deformation became less recoverable. Above, the literature demonstrates that the sludge matrix is not purely plastic in nature. Hence, to use conventional CSM for estimating bound water content in sludge may be questionable. This communication for the first time demonstrated that most centrifuged sediments (cakes) would rebound after centrifugation, thereby introducing measurement errors in bound water measurements.

EXPERIMENTAL

The samples

Two samples were examined in this work, including a sewage sludge taken from the Min-Shen Sewage Works, Taipei, Taiwan (sample #1), and one activated sludge from the wastewater treatment plant in Presidential Bread Plant, Chung-Li, Taiwan (sample #2). The Min-Shen Sewage Works handles 15,500 m³/day sewage using primary, secondary, and tertiary treatments. The sludge sample #1 was taken from the recycled stream in the secondary treatment stage, whose sediment was the testing sample with a solid weight percent of 0.91 w/w. The wastewater treatment plant in Presidential Bread Plant treated 250 tons of food processing wastewater per day using conventional activated sludge process. The sludge sample #2 was taken from the recycled sludge stream, whose sediment was the testing sample with a solid weight percent of 0.70% w/w.

For demonstrating the change in take characteristics subjected to flocculation, the original sludge samples were mixed with a cationic polyelectrolyte, indicated as polymer T-3052, from Kai-Guan Inc., Taiwan. The polymer T-3052 is a polyacrylamide with an average molecular weight of 10⁷ and a charge density of 20%. Sample sludge was first put in the mixing vessel. The polymer solution was then gradually poured into the mixing vessel with 200 rpm of stirring for 5 min followed by 50 rpm for another 20 min.

A capillary suction apparatus as described by Lee and Hsu (1992, 1993) was used to estimate the sludge filterability. The inner cylinder radius was 0.535 cm. The time required for the filtrate to migrate from 1.5 to 3.0 cm was defined as the capillary suction time (CST). Whatman No. 17 paper is the filter paper. Table 1 lists the measurements. The CST for original samples #1 and #2 were 128 and 45 s, respectively. After flocculation, the CST reached a minimum at a dose rate of 40 ppm for both samples, indicating the “optimal doses” in this work. The particle size distribution (PSD) was determined using a Coulter LS 230 particle size analyzer. The mean floc sizes for the original and flocculated sludges were listed in Table

1, which grew monotonically with the polymer dose. Zeta potentials of sludge-polymer aggregates were measured by the zeta meter (Zeter-Meter System 3.0, Zeter-Meter Inc., USA). Table 1 lists the measurements as well. Notably, the zeta potentials for sample #1 were all around -21 mV, while those for sample #2, all around -17 to -19 mV. No charge reversal was noticeable in the dosing range investigated.

Centrifugal setting measurement

The arm-suspended centrifuge proposed by Chu and Lee (2001) was refined and used in centrifugal settling tests. Transparent plastic made the settling cell for visual observation, whose inner diameter and length were 4 and 7 cm, respectively. The bottom of the setting cell was sealed for preventing the moisture leakage during centrifugation. The setting cell was connected to a rotating arm. The length from the center of rotation to the filter medium was 18.8 cm. The span angle from the center of rotation to the filter medium was measured approximately as 12°. The applied centrifugal field on the suspension mimicked a one-dimensional process.

Sixty-five millilitre of sludge was put into the settling cell. The initial height of sample was 6.0 cm (H_0). A variable-speed motor drove the belt, whence the cell, at a rotational speed ranging from 200 to 500 rpm, accelerating at the cell bottom of 8–50 *g*. The sediment layer on the cell bottom and the supernatant–sediment interface could be noticeable. A stroboscope emitting light synchronized with the rotating cell that “froze” the image of the centrifuged slurry. A video camera recorded the dynamics of the supernatant–sediment interface. Preliminary tests showed that the sediment could reach mechanical equilibrium within 800 s of centrifugation. Hence, the sludge sediment was rotated at a prescribed speed for 1400 s. The sediment height equilibrated with the centrifugal force was termed as H_c . Then the centrifuge was declined in the rotational speed in 10 s to a complete stop. Later, the sediment height was monitored continuously for another 1400 s for the occurrence of rebound. The final sediment height was named H_s .

RESULTS AND DISCUSSION

Centrifugal setting curve

The left panels for Figs 1 and 2 summarize the time evolutions of supernatant–sediment heights. The supernatant–sediment interface would move faster towards the cell bottom after sludge flocculation and/or at the higher rotational speed. For instance, the original sample #1 could reach its equilibrium sediment height for approximately 800 s at 200 rpm or for 400 s at 500 rpm.

Flocculation of sludge to a dose of 40 ppm represents the fastest sediment compaction rate for both samples, corresponding to the “optimal doses” identified using CST measurements (Table 1). For instance, to condition the sludge with 40 ppm of polymer, the corresponding centrifugation time to reach equilibrium reduces from 400–800 s to less than 30 s. Such a marked improvement is attributed to the very large flocs formed after flocculation, which lead to a high sedimentation rate and low cake resistance (Chu and Lee, 2001). Increase in rotational speed commonly produces a more compacted sludge cake. For example, sample #1 conditioned with 40 ppm polymer equilibrates with the centrifugal field at

Fig. 1. Sediment height versus time curves. Sample #1. Left panel: centrifugal tests; right panel: after the centrifugal force is released.

$H_C/H_0 = 0.26$ at 200 rpm. This ratio decreases to 0.17 when the rotational speed increases to 500 rpm. Hence, an increase in rotational speed expels more moisture from sludge, thereby leading to a dryer centrifuged cake.

Sediment rebound

The right panels of Figs 1 and 2 demonstrate the dynamics of sediment heights after the centrifugal

force is released. Some compacted sediments reveal rebound after the centrifugal field is released, especially for the tests to sample #2, which revealed significant rebound after centrifugation. For instance, centrifugation of sample #2 conditioned with 40-ppm dosage at 200 rpm (Fig. 2a) yields $H_C/H_0 = 0.51$. After the centrifugal field is released, the sediment “jumps” to around 0.64, corresponding to a rebound ratio $((H_S - H_C)/H_C)$ over 25%. At

Fig. 2. Sediment height versus time curves. Sample #2. Left panel: centrifugal tests; right panel: after the centrifugal force is released.

400 rpm, on the other hand, $H_C/H_0=0.35$ and $H_S/H_0=0.46$ (Fig. 2c), rebound at 31%. Figure 3 illustrates the rebound ratio thus calculated.

As Fig. 3a reveals that the original sample #1 exhibits no rebound for any rotational speed. That is, the original sludge sample #1 is purely plastic in nature. However, the rebound ratio increases with polymer dose, and reaches a maximum value at 40 ppm of dose, the optimal dose for sample #1.

Restated, the chemical conditioning to optimal dose has yielded the greatest rebound ratio at the prescribed rotational speed. In addition, although with certain data scattering, the rebound ratio first increases with rotational speed indicating that more elastic energy has been stored in the cake centrifugated at higher rotational speed. However, the rebound ratio markedly decreases at 500 rpm of rotation. The elasticity of the sediment network is

Table 1. Characteristics of sludge before and after flocculation^a

Polymer dose (g/kg DS)	CST (s)	Zeta potential (mV)	Floc diameter (μm)
Sample #1			
0 (original)	128	-20.1	35.5
20	40.0	-21.0	84.2
40*	25.3	-21.3	112
60	29.1	-21.0	135
Sample #2			
0 (original)	45.0	-17.1	75.6
20	29.1	-17.8	135
40*	23.0	-19.6	140
60	23.5	-18.6	186

^aOptimal dose based on CST measurements.

hence largely deteriorated at 500 rpm of rotation. There exists a threshold centrifugal force exceeding which the structure (whence the elasticity) of the sludge sediment is largely deteriorated. This observation correlates with the findings in Wu *et al.* (2001).

Sample #2 also resembles sample #1 by the dependence of rotational speed, that is, the rebound ratio would first increase with the rotational speed, after reaching a maximum at 300–400 rpm, and decrease when the speed goes up further. A threshold force also exists for sample #2 less than which the sediment matrix could be regarded as elastic. Meanwhile, sample #2 represents some distinct characteristics. When compared with the rather dramatic changes observed for sample #1, the rebound ratio decreases mildly with the polymer dose for sample #2. Also, the original sludge exhibits the greatest rebound. That is, the sediment of the original sample #2 has been (partially) elastic, while the presence of a polymer flocculant deteriorates the sediment's elasticity.

Errors in bound water measurement

Using the sediment height equilibrated with the centrifugal force (H_C) and that measured after the centrifuge is stopped (H_S), together with the solid density, the residual moisture retained in the centrifuged sediment could be calculated, which are referred as the bound water data measured using CSM, BW_C and BW_S , respectively. Their ratio in difference $(BW_S - BW_C)/BW_C$ closely corresponds to those depicted in Fig. 3. In all cases $BW_S > BW_C$. Restated, moisture would be "sucked back" into the dewatered cake after the release of centrifugal force. Under such a circumstance, the bound water measured using CSM would be overestimated with the magnitude depending on the polymer dosage and the centrifugal force applied. This dependence appears very complicated and sludge-dependent. That is, different sludges could exhibit very different sediment elasticities whose role is hard to predict a priori. Hence, the reliability of using CSM to measure bound water content in sludge is highly

Fig. 3. Rebound ratio versus polymer doses: (a) Sample #1 and (b) Sample #2.

questionable. The centrifugal dewatering test in which the free water is completely removed from the sediment through a septum (Chu and Lee, 2001) or the present centrifuge with which the sediment height could be easily observed, should be used.

CONCLUSIONS

This work demonstrated the possible errors incorporated into bound water measurement using conventional centrifugal settling method (CSM). We measured the responses of sludge sediment heights when compacted under a centrifugal field, with especial attention to the height rebound after the centrifugal force had been released. Two samples were examined: the recycled sludge from a Sewage Work and an activated sludge from the food industry. The sediment height equilibrated with the

centrifugal force decreased monotonically with increasing rotational speed, which correlated with the commonly accepted concept for bound water: higher shear could remove more bound water from the sludge matrix. However, after the centrifuge was stopped, most sediments revealed a rebound, whose magnitude depended on the polymer dose and the rotational speed. Also, different sludges could have distinct elastic responses under centrifugation. There existed a threshold rotational speed exceeding which the elasticity of sediment matrix was largely deteriorated. Neglecting the role of the sludge sediment's elasticity would introduce errors in bound water estimation. There existed no simple way to correlate such an error using conventional CSM. Instead, the use of centrifugal dewatering method or the refined centrifuge employed herein could provide more reliable measurements.

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REFERENCES

- Chang I. L. and Lee D. J. (1998) Ternary expression stage of biological sludge dewatering. *Water Res.* **32**, 905–914.
- Chen G. W., Chang I. L., Hung W. T., Lee S. F. and Lee D. J. (1997) Continuous classification of moisture content in waste activated sludge. *J. Environ. Eng. ASCE* **123**, 253–258.
- Chu C. P. and Lee D. J. (2001) Centrifugation dewatering of activated sludge after flocculation. *Water Res.* **35**, 2377–2384.
- Chu C. P. and Lee D. J. (1999) Three stages of consolidation dewatering of sludges. *J. Environ. Eng. ASCE* **125**, 959–965.
- Colin F. and Gazbar S. (1995) Distribution of water in sludges in relation to their mechanical dewatering. *Water Res.* **29**, 2000–2005.
- Heukelekian H. and Weisberg E. (1956) Bound water and activated sludge bulking. *Sewage Ind. Wastes* **28**, 558–574.
- Kawasaki K., Matsuda A., Ide T. and Murase T. (1990) Measurement of cell density and floc property of excess activated sludge conditioned by freezing and thawing process, using centrifugal settling. *Kagaku Kogaku Ronbunshu* **16**, 401–403.
- Kawasaki K., Matsuda A. and Mizukawa Y. (1991) Compression characteristics of excess activated sludges treated by freezing-and-thawing process. *J. Chem. Eng. Japan* **24**, 743–748.
- Lee D. J. (1994) Measurement of bound water in sludges: the use of centrifugal settling method. *J. Chem. Tech. Biotechnol.* **61**, 139–144.
- Lee D. J. (1996) Interpretation of bound water measured via dilatometric technique. *Water Res.* **30**, 2230–2232.
- Lee D. J. and Hsu Y. H. (1992) Fluid flow in capillary suction apparatus. *Ind. Eng. Chem. Res.* **31**, 2379–2384.
- Lee D. J. and Hsu Y. H. (1993) Cake formation in capillary suction apparatus. *Ind. Eng. Chem. Res.* **32**, 1180–1185.
- Lee D. J. and Hsu Y. H. (1995) Measurement of bound water in sludges: a comparative study. *Water Environ. Res.* **67**, 310–317.
- Lee D. J. and Lee S. F. (1995) Measurement of bound water in waste activated sludges: the use of differential scanning calorimetry (DSC). *J. Chem. Tech. Biotechnol.* **62**, 359–364.
- Lee D. J. and Wang C. H. (2000) Theories of cake filtration and consolidation and implications to sludge dewatering. *Water Res.* **34**, 1–20.
- Matsuda A., Kawasaki K. and Mizukawa Y. (1992) Measurement of bound water in excess activated sludges and effect of freezing and thawing process on it. *J. Chem. Eng. Japan* **25**, 100–103.
- Mitani T., Unno H. and Akehata T. (1983) A structure model of activated sludge flocs. *Suishitsu Odaku Kenkyu* **6**, 69–75.
- Robinson J. and Knoke W. R. (1992) Use of dilatometric and drying techniques for assessing dewatering characteristics. *Water Environ. Res.* **64**, 60–68.
- Smollen M. (1990) Evaluation of municipal sludge drying and dewatering with respect to sludge volume reduction. *Water Sci. Technol.* **22**, 153–160.
- Vesilind P. A. (1994) The role of water in sludge dewatering. *Water Environ. Res.* **66**, 4–11.
- Wu C., Huang C. and Lee D. J. (1997) Effects of polymer dose on alum sludge dewaterability characteristics and physical properties. *Colloids Surf.* **122**, 89–96.
- Wu R. M., Lee D. J., Wang C. H., Chen J. P. and Tan R. B. H. (2001) Novel cake characteristics of waste activated sludge. *Water Res.* **35**, 1358–1362.