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# Effects of Acidification on Dewaterability and Aluminum Concentration of Alum Sludge

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**This study utilizes the Terzaghi-Voigt model to characterize the effects of acidification on the dewaterability of alum sludge. Alum sludge, which was obtained from the sedimentation basins of a water treatment plant using poly aluminum chloride as coagulant, was acidified to different pH levels with sulfuric acid. The dewaterability of the acidified sludge was characterized by expression tests. The results show dewaterability enhancement was insignificant until pH was below 4. Further improvement in dewaterability can be achieved by polymer conditioning. The Terzaghi-Voigt model can be applied to explain the difference in dewaterability between acidified and original sludges. Results also show dissolved aluminum concentrations were controlled by minerals in the influent rather than amorphous aluminum hydroxide.**

**Keywords** acidification; alum sludge; creeping factor; dewaterability; expression; secondary consolidation

## INTRODUCTION

The cost of waste sludge including dewatering and final disposal is a major operation cost for water and wastewater treatment plants (1). Due to its amount and cost for proper disposal, alum sludge produced during the purification process has caused severe problems for water supply facilities in Taiwan. Reuse and recycle (or recovery) are the most effective treatment alternatives for the large amounts of sludge. The Taiwan government has a sludge management strategy that aims to reduce landfill disposal and increase beneficial sludge reuse. To implement and meet the goal of zero sludge, the Taiwan government has established many strategies for sludge recovery and recycling. Among these strategies, non-hazardous inorganic sludge can serve as raw materials for manufacturing bricks, aggregates, and other reusable products using thermal treatment or chemical transformation technologies. On the other hand, sludge acidification with aluminum recycle is one of the alternatives being considered for effectively reducing the sludge volume and treatment cost in Taiwan. This technology has been reported and applied in many countries (2–6). The most significant advantages of the process are the reuse of coagulant resulting from the recovery of alum and the treatment cost reduction from the decrease of sludge volume. In addition, acidification of sludge also improves the dewaterability of sludges, thus reducing the volume of sludge cakes (3,6–7). Previous research focusing on the quality (purity) and coagulation efficiency of the recycled alum coagulants has made great contributions toward large-scale application of this technique.

Although the improvement of settling ability and dewaterability of acidified sludge has been confirmed, the mechanisms involved are still not well understood for wastewater/water treatment sludge. The methods for improving the dewaterability of sludge have been reported frequently. Li et al. (7) employed both acidification and alkalization treatments on textile chemical sludge to recover the coagulant and found only the acidification could improve the dewaterability of sludge. They attributed the improvement of dewaterability to charge neutralization of sludge particles through acidification. Treating activate sludge with sulfuric acid at pH 2.5 was an effective method to remove the exocellular polymer (ECP) from the sludge surface, which resulted in a more compact sludge aggregate and the improvement of mechanical dewatering (8). However, based on the conventional perspective of sludge condition/dewatering, charge neutralization is not the dominant mechanism for dewaterability improvement and the content of ECP on the surface of alum sludge is far less than that of activated sludge. Therefore, the reasons (mechanisms) for improvement in dewaterability of activated sludge by acidification cannot be used to explain the improvement of dewaterability of alum sludge by acidification.

During the sludge dewatering process, the removal of moisture can be divided into two stages: filtration followed by consolidation. The expression test and Terzaghi-Voigt model have been used to clarify the change of moisture

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removal stages and to determine the value of consolidation ratio and the relative ease of mobility of constituting particles during sludge dewatering (9–11). The Terzaghi-Voigt rheological model describing the expression process can be written as Eq. (1) (10).

$$U_c = \frac{L_1 - L}{L_1 - L_f} = 1 - \beta \exp(-\eta\theta_c) \quad (1)$$

where, $U_c$ is the consolidation ratio, $L$ is the cake thickness, $L_1$ and $L_f$ are the initial and final cake thickness, respectively, $\beta$ is the fraction of moisture removed by the secondary consolidation to total consolidation, $\eta$ is the creeping factor, and $\theta_c$ is the expression time. The primary consolidation mechanism was proposed to be the removal of pore liquid and the collapse of cake structure, whereas the secondary consolidation mechanism was proposed to be the disturbance of the particle bonding. The creeping of particles enables the packing between particles to a more stable state (12).

Several indicators are used to evaluate the improvement of sludge dewaterability according to the Terzaghi-Voigt model, i.e., a decrease of the secondary consolidation, $\beta$, and an increase of creeping factor, $\eta$. A large $\beta$ value indicates that a large portion of water is still within the sludge after primary consolidation, and therefore is undesired. A large $\eta$ value indicates more ease for the creeping of aggregates, which is favored during the dewatering of the sludge. The previously described improvement of sludge dewaterability is usually achieved by polymer conditioning or freeze/thaw techniques (10). However, it is unclear whether acidification achieves improvement of sludge dewaterability by the same mechanisms as polymer and freeze/thaw conditioning. Thus, the objective of this study is to utilize the Terzaghi-Voigt model to characterize the effects of acidification on the dewaterability of alum sludge.

## MATERALS AND METHODS

Alum sludge samples were taken from sludge thickening tanks in the Fang-Yuan water supply facility of the Taiwan Water Supply Company in central Taiwan. The average turbidity of raw water was $30 \sim 70$ NTU and the coagulant used was poly aluminum chloride (PACl) with a dosage between 1.5 and 4.0 ppm. Prior to acidification, samples were thickened again by sedimentation till the total solid content of sludge reached about 25%.

The settled sludge was acidified with 98% $H_2SO_4$ to the desired pH values and stirred for 2 hrs in a 1-L beaker placed in a water bath, of which temperature was set at 25°C. In our preliminary study, the aluminum concentration in the supernatant of acidified sludge should reach a stable level after 2 hrs of acid extraction.

The dewaterability of settled sludge was evaluated by an express unit using a pressure of 150 psi. During the expression test, the weight of the filtrates was automatically recorded by an electronic balance connected to a computer. After the expression test, the sludge was subjected to 800 psi of pressure to further extract water. The residual moisture of the expressed cake was determined by placing the sample in an oven maintained at a constant temperature of 105°C for 24 hrs. This residual moisture is defined as bound water content of the sludge in this study. Specific resistance of filtration (SRF), cake moisture content, $U_c$, $\beta$, and $\eta$ values were determined from the corresponding filtrate volume and time during the expression tests.

The values of sludge SRF were determined by applying Eqs. (2) and (3) to the data of filtering the sludge in a Buchner funnel under a constant vacuum differential of 49 kPa (13).

$$t/V = bV \quad (2)$$

where $t$ is the filtration time, $V$ is the volume of the filtrate, and $b$ is a constant as described in Eq. (3).

$$b = \frac{\mu r c}{2PA^2} \quad (3)$$

where $\mu$ is the filtrate viscosity, $r$ is the SRF of the sludge, $c$ is the solid concentration of the sludge, $P$ is the applied pressure differential, and $A$ is the area of the filter. According to Eq. (2), plotting $t/V$ versus $V$ yields a straight line with a slope of $b$, which can be substituted into Eq. (3) to estimate the value of SRF, $r$. Typically, SRF has a unit of m/kg.

A cationic polymer (PC-320) with a $1.5 \times 10^7$ molecular weight and 20% charge density was used to condition the sludge. The optimal polymer dosage was selected using the lowest obtained capillary suction time (CST). The aluminum concentration in the supernatant was analyzed with a flame atomic absorbance spectrometry (Varian AA-240).

## RESULTS AND DISCUSSION

### Effects of pH on Moisture Removal

The effect of acidification on moisture removal is insignificant at pH = 5 and becomes significant when pH values are below 4 (Fig. 1). In fact, the expression curves of the original (pH = 7.0) and pH = 5 sludges are almost identical. When the sludge is acidified to pH values below 4, most of the free water is released after 200 seconds (s) of expression, whereas the original and pH = 5.0 sludges still retained a large portion of water, as shown in Fig. 1. It took about 1,500 sec of expression time for the original and pH = 5 sludges to reach the same degree of moisture removal as those with lower pH values (Fig. 1). This suggests that

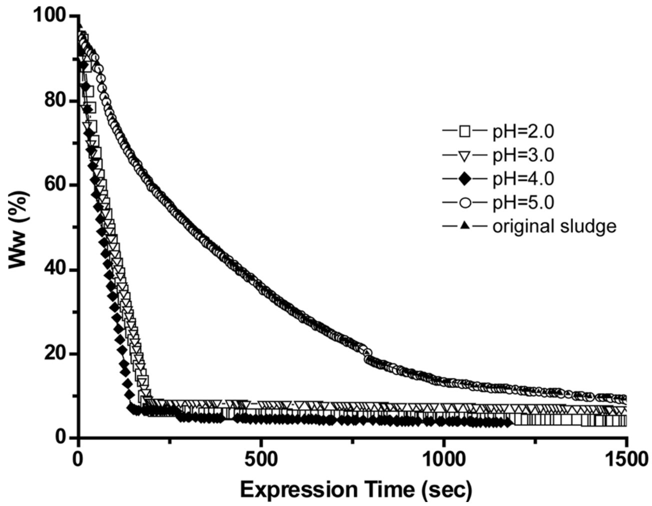


FIG. 1. Sludge cake moisture content with expression time at various pH values.

acidification of the sludge to pH values below 4 significantly enhances the dewaterability of the sludge.

### Effects of pH on $\beta$ and $\eta$ Values

The Terzaghi-Voigt model curves for expression of the Fang-Yuan sludge at various pH values are shown in Fig. 2. In general, each curve consisted of two regions of different gradients with the first region having a steeper gradient than the second region. The first region corresponds to the filtration stage, whereas the second region corresponds to the consolidation stage. In practice, a steep gradient in the first stage indicates good dewaterability.

The corresponding $\beta$ and $\eta$ values of the curves in Fig. 2 are shown in Table 1. The original and pH = 5 sludge have comparable $\beta$ (0.92) and $\eta$ (0.002) values (Table 1). This indicates acidification of sludge to pH = 5 causes a minor effect on the dewaterability of sludge. The effect of acidification on $\beta$ values becomes more significant at pH values lower than 4 with values decreasing from 0.92 of the original sludge to less than 0.05 of the acidified sludges (Table 1). This indicates that when the sludge pH is lowered to less than 4, more than 95% of the water can be easily separated from the solids. Compared to less than 10% of the water removed at the same stage of expression for the original and pH = 5 sludges, this is a dramatic improvement of dewaterability. On the other hand, acidification has little effect on the $\eta$ values, decreasing from 0.002 of the original sludge to approximately 0.001 of the acidified sludges (Table 2). This suggests that under high expression pressure, the original and acidified sludges have similar particle structures.

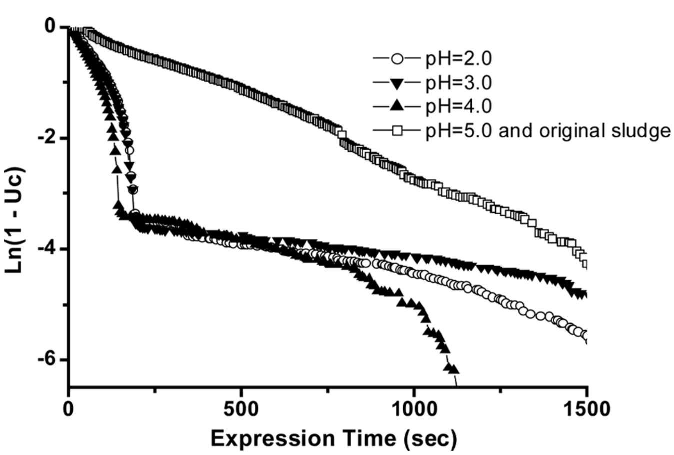


FIG. 2. $Ln\,(1 - U_c)$ with expression time of sludges at various pH values.

TABLE 1
$\beta$ and $\eta$ values of sludges at different pH values

| pH | $\beta$ | $\eta$ |
|---|---|---|
| Original | 0.92 | 0.0020 |
| 2.0 | 0.0322 | 0.0009 |
| 3.0 | 0.0308 | 0.0006 |
| 4.0 | 0.0484 | 0.0016 |
| 5.0 | 0.9171 | 0.0020 |

$\eta$: (1/sec).

### Effects of pH on SRF

Similar to moisture content, $\beta$ and $\eta$ values, the effect of acidification on SRF is not significant until the sludge pH is lowered to below 4 (Table 2). The SRF decreases by approximately one order of magnitude indicating significant improvement of dewaterability. To test the reversibility of acidification, the pH of the acidified sludge was

TABLE 2
SRF of sludges at different pH values

| Samples | SRF (m/kg) |
|---|---|
| With $H_2SO_4$ | |
| Raw (pH = 7.0) | $4.89 \times 10^{11}$ |
| pH = 2.0 | $6.41 \times 10^{9}$ |
| pH = 3.0 | $1.69 \times 10^{10}$ |
| pH = 4.0 | $5.09 \times 10^{9}$ |
| pH = 5.0 | $4.34 \times 10^{11}$ |
| pH = 6.0 | $4.54 \times 10^{11}$ |
| NaOH addition after acidification | |
| pH = 2.0 → 7.0 | $1.20 \times 10^{10}$ |
| pH = 3.0 → 7.0 | $1.06 \times 10^{10}$ |
| pH = 4.0 → 7.0 | $7.69 \times 10^{9}$ |
| pH = 5.0 → 7.0 | $4.23 \times 10^{10}$ |

brought back to 7.0, and the SRF was measured again. The SRF values of the acidified and base-treated sludges are of the same magnitude (Table 2). This means that once the sludge is acidified, the dewaterability of the sludge is unaffected by the increase of pH. The irreversible effect of acidification on the dewaterability of alum sludge is desired during dewatering and subsequent treatment and disposal of the sludge.

## Effects of Polymer Conditioning

Polymer (5 mg/L) was added to the acidified sludge to further improve the dewaterability of the sludge. The results of the expression curves indicate that the polymer can further improve the dewaterability of the acidified sludge (Fig. 3). However, the improvement is not as significant as that of acidification on the original sludge as shown in Fig. 1. Polymer conditioning affects the dewaterability of the acidified sludge in two directions. First, it reduces the final (residual) water content from about 5% to 2% (Fig. 3b). Second, it reduces the time to reach the final water content of the acidified sludge from approximately 200 s to 50 s (Fig. 3b). This suggests that polymer conditioning of the acidified sludge could further enhance the sludge dewaterability and reduce the final volume of sludge cake as well as the disposal cost for it.

The $\beta$ value for the polymer-conditioned sludge is 0.02, a slight improvement from the acidified sludge ($\beta$ between 0.03 and 0.05) and the $\eta$ value is 0.0008. This indicates that the majority of water is easily removed during the initial filtration stage and little water is trapped within the cake. The increase on the creeping factor, $\eta$, also illustrates that the flocs are aggregated due to charge neutralization and bridging and the creeping behavior among flocs in the acidified sludge was further enhanced by conditioning. Flocculation enhances moisture transport during cake expression.

## Effects of pH on Aluminum Concentration

The significant improvement of sludge dewaterability when the sludge pH was lowered to less than 4 could be due to the dissolution of aluminum hydroxide and other aluminum oxide. PACl was used in the water treatment plant as a coagulant and contributed to the sludge solid content in the form of aluminum oxide/hydroxide. As aluminum oxide/hydroxide has poor dewaterability, it is generally regarded as the main cause for the poor expression behavior of sludge as shown in Fig. 2 for the original and pH = 5. This can be verified with the aluminum concentration (in log scale) in the supernatant (Fig. 4). The aluminum concentration in the supernatant of the original and pH = 5 sludges were almost the same and were approximately 0.2 mg/L. The aluminum concentration increases significantly to 39 mg/L at pH = 3.5 and approximately 2000 mg/L at pH = 2. The behavior of

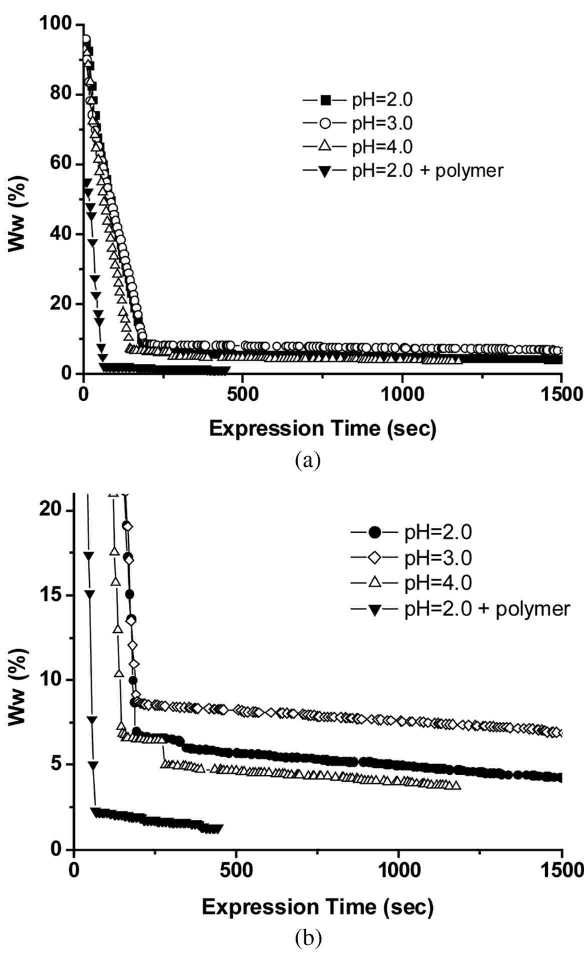


FIG. 3. Sludge cake moisture content with expression time for acidified and polymer-conditioned acidified sludge. (a) entire moisture range; (b) moisture ranges between 0 and 20%.

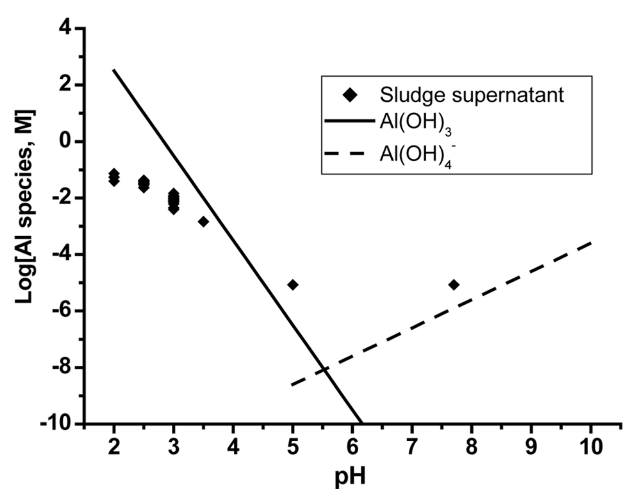


FIG. 4. Aluminum concentration in the supernatant as a function of pH.

aluminum concentration in the supernatant with pH is similar to that of sludge dewaterability, i.e., the pH of the sludge needs to be adjusted to below 4 for significant effects to take place. Therefore, the dissolution of aluminum hydroxide/oxide indeed contributes to the enhancement of sludge dewaterability.

The log aluminum concentration (mg/L) is almost linear with pH with a slope of $-1$ when the pH is below 4. This suggests that the log(Al)-pH relationship is not governed by amorphous aluminum hydroxide in this pH range, in which case it should yield a slope of $-3$ (Fig. 4). The aluminum concentration is also well below that of the amorphous aluminum hydroxide solubility when the pH is below 4. This further confirms that aluminum concentration is controlled by minerals rather than amorphous aluminum hydroxide in this study. These minerals are probably from the suspended solids in the influent of the treatment plant and are easily dewatered. When the pH is above 5, the aluminum concentration is above the solubility of aluminum hydroxide. This could also be due to the contribution of aluminum as a result of the dissolution of soil minerals.

The characteristics of minerals that control the aluminum concentration in the pH ranges shown in Fig. 4 are unclear and are not the subject of this study. However, the log(Al)-pH relationship shown in Fig. 4 is comparable to the results of other research (14,15). In both studies, the results showed that the aluminum concentration is undersaturated with respect to gibbsite ($pKa = 8$) at pH less than 4.5. In their studies, the log(Al)-pH curves of different soil compositions at pH less than 4.5 shows a similar pattern (i.e., slopes between $-2$ and $-1$) as shown in Fig. 4. Thus, further study is needed to clarify the mechanisms controlling the dissolution of aluminum during acidification of alum sludge.



## CONCLUSIONS

Lowering the pH of alum sludge from pH 7.0 (original sludge) to less than 4.0 by sulfuric acid results in significant improvement of sludge dewaterability. The enhancement of sludge dewaterability by acidification is irreversible. The enhancement of dewaterability of sludge can be characterized with the Terzaghi-Voigt model using the model parameter. It indicates that more than 95% of water can be easily dewatered after sludge acidification. The SRF of the acidified sludge decreases more than one order of magnitude than that of the original sludge, further confirming the improvement in sludge dewaterability. Further improvement in dewaterability of the acidified sludge could be achieved by polymer conditioning. Improvement of sludge dewaterability by acidification is accompanied by dissolution of aluminum-containing minerals. The aluminum equilibrium by acidification is not controlled by the dissolution of aluminum hydroxide, which is the main product formed during coagulation. It is proposed that soil minerals in the influent of the water treatment plants controlled the equilibrium of aluminum. More studies are needed to confirm this mechanism.

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