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Research Article

Municipal Sewage Sludge Drying Treatment by an Composite Modifier

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A sludge composite modifier (SCM) which comprises a mixture of three cementitious components was proposed for sludge drying and stabilization. Effect of SCM components on sludge moisture content was analyzed using uniform design and the optimum composition of SCM was determined by computer-aided modeling and optimization. To compare the drying effect of SCM, quicklime, and Portland cement, the effects of material content and curing time on moisture content of sludge were also studied. The results showed that the optimum ratio of modifier component was slag/cement clinker/dihydrate gypsum = 0.64/0.292/0.068 and the moisture content of SCM-stabilized sludge decreased with the increasing material content and extending curing time. Besides, the experimental results showed that optimized SCM behaved better than quicklime and Portland cement in sludge semi-drying and XRD analysis revealed that the main hydrated product of stabilization was ettringite, which played an important role in the effective drying process. Sewage sludge stabilized using SCM could be used as an effective landfill cover.

1. Introduction

Sewage sludge is the waste that remains after the treatment of municipal wastewater by wastewater treatment plants. The disposal of huge quantities of sewage sludge is a major environmental problem. This problem is especially significant in cities [1, 2]. In China, the annual production of moist sewage sludge was about 11 million tons in 2010 [3], and it is expected that wastewater treatment percentage will reach 75% by 2015, thus production of sewage sludge in China will continuously increase [4]. It is expected that sewage sludge disposal will be one of the most complex environmental problems facing the engineering in this field in China. In general, sludge that is discharged from treatment plants in the form of dehydrated cakes has a high water content of 75–85% and one sludge stabilization option is to dry it, which yields a solid that is low in humidity. This solid can be easily stored, recycled, or transported to other facilities. Dried sewage sludge can be dumped in landfills, pyrolyzed, incinerated, or gasificated [5].

In recent years, conventional thermal drying and solar drying methods are usually adopted to dry sewage sludge. The typical thermal methods currently widely used to dry sludge include convection heat transfer of direct hot gas blasting and conduction heat transfer of steam inside screw and sludge [6]. Thermal drying is an efficient method that produces a biologically stable material with improved characteristics; however, the significant fuel consumption and potential emission of greenhouse gases (mainly CO_2) are important conditions [7]. Besides, Ulrich Luboschik [8] found that the operating costs of the solar drying system are competitive, but the characteristic feature of solar drying is the dependence on radiation intensity, which varies not only with daily cycle, but also with season and based on a year, approximately 750 kg water/m^2 ground space can be removed from the sludge. Mathioudakis et al. [9] reported the extended dewatering of sewage sludge in solar drying plants and found that during summer conditions, drying was completed within 7–12 days, but in autumn conditions the duration increased up to 32 days. Obviously, the conventional evaporative drying process may take long processing time, or involve high energy consumption.

On the other hand, many cementitious materials such as quicklime and cement have been used for sludge stabilization [10–13]. However, despite the fact that stabilization of sludge with cementitious materials has an extensive history of use, there exist few experimental studies for materials-based sludge drying as research is mainly oriented towards mechanical and sanitary properties of stabilized sludge. In addition, slag has received much attention as a cost-effective and efficient solidifying agent because of its cement-like characteristics and its abundance as a waste product from steel production plants. It is an economical material for sewage sludge stabilization and drying. Thus a sludge modifier (SCM) which comprises three kinds of cementitious materials (slag, cement clinker, and dihydrate gypsum) was proposed and optimized for more economical sludge drying and stabilization. Due to the complex components of SCM, it involves many parameters and complex interactions between them, the one-factor-at-a-time approach requires prohibitively large numbers of trials to systematically identify the drying effects of different components. In order to minimize the number of experimental trials in optimizing the sludge modifier formula, statistical and computational methods were proposed to be applied in this investigation. The uniform design is a new method established together by Wang et al. [14]. It is a fractional design which may be used when the underlying model between the response and factors is unknown or partially unknown. By uniform design method, the optimal proportion of SCM components was experimentally determined. Furthermore, the dewatering ability of SCM was investigated and compared with quicklime and Portland cement to examine the feasibility of its utilization as a drier in the context of economical drying process of sludge.

2. Experiment and Results

2.1. Materials

The sewage sludge used was obtained from the Wastewater Treatment Plant of Hanxi, Wuhan city. The main characteristics of sludge are listed in Table 1. The chemical compositions of quicklime, Portland cement, slag, cement clinker, and dihydrate gypsum are listed in Table 2.

Table 1: Main characteristics of the sewage sludge.

Moisture (%)	Density (g/cm^3)	PH	Organic matter (%)
80.87	1.04	7.07	60.20

Table 2: Chemical composition of quicklime, Portland cement, slag, cement clinker, and dihydrate gypsum (wt%).

	CaO	SiO_2	Al_2O_3	Fe_2O_3	SO_3	MgO
Quicklime	81.12	3.70	0.49	0.09	0.04	0.01
Portland cement	59.82	24.13	6.35	3.57	2.24	0.98
Slag	37.68	30.46	14.95	1.12	1.95	9.9
Cement clinker	63.83	21.48	4.72	3.63	0.67	3.70
Dihydrate gypsum	42.87	4.2	0.25	0.05	44.22	1.17

2.2. Sample Preparation and Measurement

Sludge samples in wet conditions were mixed with materials (quicklime, Portland cement, and SCM, resp.) and the mixtures were cured at room temperature for various periods. The mixing procedures are as follows: the wet sludge was placed in a mixer first, materials with a designed content were then added and the mixture stirred for 30 min. After homogenization, mixtures were placed into open containers and cured at room temperature. Representative samples were collected from each mixture at specific time intervals, that is, 3 h, 1, 3, 7, and 14 d, and were analyzed for the measurement of the moisture content on wet basis. The values of moisture content reported are the average of three specimens of each stabilized mixtures.

2.3. SCM Prescription Experiment

The influence of SCM components on stabilized sludge moisture content and the optimum proportion of SCM components were investigated with uniform recipe design method. In this study, samples were prepared by mixing wet sewage sludge and three SCM components in various ratios and all mixtures were stabilized for a period of 3 h. The property is the moisture content of stabilized mixtures, denoted by Y (%) and factors involved are proportions of sludge (x_1), slag (x_2), cement clinker (x_3), and dihydrate gypsum (x_4). The proper ranges of factors, obtained by previous experiments, were

$$0.5 \leq x_1 \leq 0.99, \qquad 0.004 \leq x_2 \leq 0.45, \qquad 0.0005 \leq x_3 \leq 0.275, \qquad 0.0005 \leq x_4 \leq 0.27. \quad (2.1)$$

The selections of these ranges are based on technical and/or economical reasons. Then the uniform design table and corresponding experimental scheme were obtained by the conditional probability distribution method [15]. The obtained uniform design table, together with the experimental scheme and results are listed in Table 3.

Furthermore, the optimum formula of SCM was determined by corresponding mathematical modeling and optimization. We used this optimized SCM in subsequent sludge drying experiments.

Table 3: Uniform design table and experimental scheme and results.

No	No. of columns			Factors				Y (%)
	1	2	3	x_1	x_2	x_3	x_4	
1	11	8	7	0.6070	0.0435	0.1624	0.1871	41.31
2	7	16	6	0.5144	0.1004	0.1458	0.2394	31.95
3	14	15	12	0.7027	0.0818	0.1727	0.0428	48.71
4	6	9	13	0.5749	0.0105	0.1145	0.3001	41.54
5	16	10	9	0.6817	0.0992	0.1201	0.0990	47.59
6	1	7	5	0.5012	0.0641	0.1385	0.2963	32.91
7	15	3	4	0.7729	0.0168	0.1309	0.0795	57.15
8	8	5	1	0.7022	0.0331	0.1453	0.1194	49.84
9	10	1	11	0.7599	0.1280	0.0512	0.0608	52.77
10	9	12	16	0.6472	0.0228	0.1667	0.1633	45.88
11	4	11	3	0.8052	0.0930	0.0931	0.0087	58.39
12	3	4	15	0.7352	0.0933	0.0307	0.1408	49.42
13	2	14	10	0.6752	0.1279	0.1715	0.0254	43.24
14	5	2	8	0.5280	0.0652	0.0720	0.3348	36.33
15	13	6	14	0.5070	0.0932	0.1478	0.2520	32.17
16	12	13	2	0.7165	0.0605	0.0105	0.2124	52.55

2.4. Sludge Drying Experiments

To evaluate the changes in sludge moisture content caused by SCM in relation to its content and duration of stabilization process, mixtures of sludge-SCM were prepared with different SCM contents and cured for different periods. Meanwhile, quicklime and Portland cement, as conventional binder, were used for comparison and sludge-quicklime and sludge-Portland cement samples were prepared similarly.

Here, materials content, which was defined as weight percentage of materials in sludge-material mixture, and curing time were the two considered factors. Various material content and curing time, respectively, ranging from 5% to 60% and 3 h to 14 d, were examined.

2.5. X-Ray Diffraction Analysis of Hydration Products of SCM

XRD was performed to identify and quantify the crystalline mineral phases present in the SCM hydration products. For this purpose, pure SCM pastes (without the addition of sludge) were prepared with a water/SCM ratio of 0.56 by weight. The pastes were analyzed with an X-ray diffractometer (Philip, PE Model 1729) using Cu Ka radiation.

2.6. SEM Analysis

SEM analysis was performed on raw sludge and the modifier-dried sludge sample with SCM. The samples were gilt with Au and SEM was operated at 20 kV of acceleration voltage.

Table 4: The verification experiments and results.

No	x_1	x_2	x_3	x_4	Y (%)	Y' (%)	$\lvert\Delta Y\rvert$ (%)
17	0.6	0.1	0.16	0.14	37.57	37.25	0.32
18	0.7	0.1	0.12	0.08	45.95	46.57	0.62
19	0.8	0.12	0.04	0.04	55.86	56.14	0.54
20	0.9	0.05	0.025	0.025	68.20	68.81	0.61

3. Discussions

3.1. Ratio of SCM Components Optimization

Results of uniform design scheme were analyzed using second-order polynomial stepwise regression analysis. The obtained equation, together with its significance level (P) and correlation coefficient after adjustment (Ra) are given in (3.1). Analyzed by F-test method, (3.1) is intensively significant. Furthermore, the validity of (3.1) was assessed experimentally and the results (Table 4) showed that the predicted value (Y) was close to the experimental result (Y'). Then, combing the results of uniform design and verification experiment, a similar regression analysis was made to improve veracity of model prediction. The results obtained are given in (3.2)

$$Y = -0.1656 + 0.9483x_1 - 2.2063x_2^2 + 0.3486x_4^2 \quad (P = 0.0001,\ Ra = 0.9952), \tag{3.1}$$

$$Y = 0.2669 - 0.5329x_2 + 0.5568x_1^2 - 0.3344x_3^2 - 0.8734x_3 \times x_4 \quad (P = 0.0001,\ Ra = 0.9977). \tag{3.2}$$

Thus, the final regression model between Y and x_i ($i = 1, 2, 3, 4$) can be expressed by (3.2). The model optimization step is to find x_1^*, x_2^*, x_3^*, x_4^* such that

$$\begin{gathered} Y^* = Y^*\left(x_1^*, x_2^*, x_3^*, x_4^*\right) = \min Y(x_1, x_2, x_3, x_4), \\ 0.5 \le x_1 \le 0.99, \qquad 0.004 \le x_2 \le 0.45, \qquad 0.0005 \le x_3 \le 0.275, \qquad 0.0005 \le x_4 \le 0.27, \end{gathered} \tag{3.3}$$

where $Y^*(x_1^*, x_2^*, x_3^*, x_4^*) = Y^*$ is given by (3.2). Here, Simulated Annealing method was used to optimize the model and determine the optimum ratio of SCM components. The result indicated that sludge moisture content exhibited its lowest value at the weight ratio of slag to cement clinker to dihydrate gypsum of 0.64 : 0.292 : 0.068.

3.2. Influence of Material Content on Stabilized Sludge Moisture Content

Figure 1 shows that SCM is more effective than Portland cement and compared with quicklime, SCM is more effective in sludge drying at a lower content (<50 wt%), whereas quicklime behaves better at higher content. This is probably caused by different physical-chemical interactions involved in different sludge-material systems.

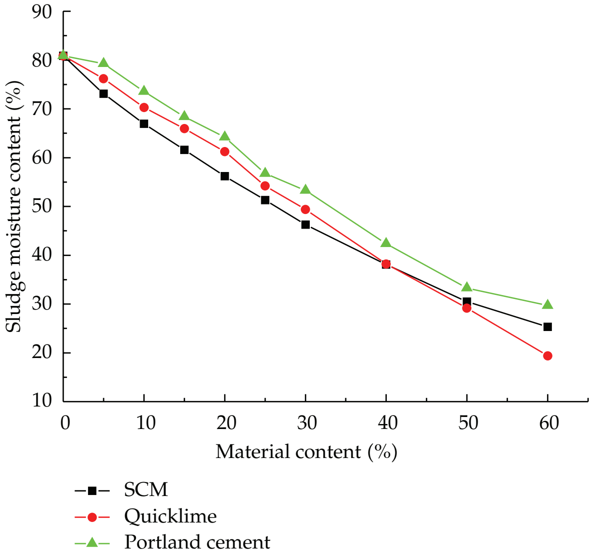


Figure 1: Moisture content of stabilized sludge cured for 3 h as a function of material content.

For Portland cement, the hydration reactions between SCM components and free water in sludge are responsible for the sludge drying and stabilization. The hydration product of Portland cement is C-S-H gel and a spot of ettringite ($3CaO{\cdot}Al_2O_3{\cdot}3CaSO_4{\cdot}32H_2O$, abbreviated as AFt), which can convert some free water in sludge into crystal water. However the amount of hydration products was few and the free water being converted was few due to the slow hydration rate of Portland cement. This is the reason why Portland cement has poor drying effect. Thus three kinds of cementitious components were chosen to speed the hydration rate and the formation of C-S-H gel especially Aft. Thus, the sludge moisture content decreases with the increase of SCM content; however, when the modifier content is relatively high, the tendency of decrease slows down because the amount of free water in sludge is limited.

For quicklime, sludge drying is attributed to both chemical reaction and physical evaporation. The reactions are expressed, respectively, in

$$\begin{gathered} CaO + H_2O \longrightarrow Ca(OH)_2 + Q \quad (Q = 64.9\,\text{kJ/mol}), \\ H_2O\ (l) + 2258\,\text{kJ/kg} \longrightarrow H_2O\ (g). \end{gathered} \tag{3.4}$$

From (3.4), 1 g CaO consumes 0.32 g water, while the amount of water evaporated by hydration heat is 0.51 g. Thus, physical evaporation is the main mechanism for quicklime-based sludge drying. This is the reason why the moisture content of quicklime-stabilized sludge decreases obviously with the quicklime content.

It can be seen that the SCM is more suitable for sludge semi-drying treatment, while the quicklime can be used for sludge-drying treatment. Obviously, the increase of materials content will increase the total quantity of sludge. Thus it will increase the treatment cost of the sludge disposal and it is not economical in industry to use such a large amount of quicklime.

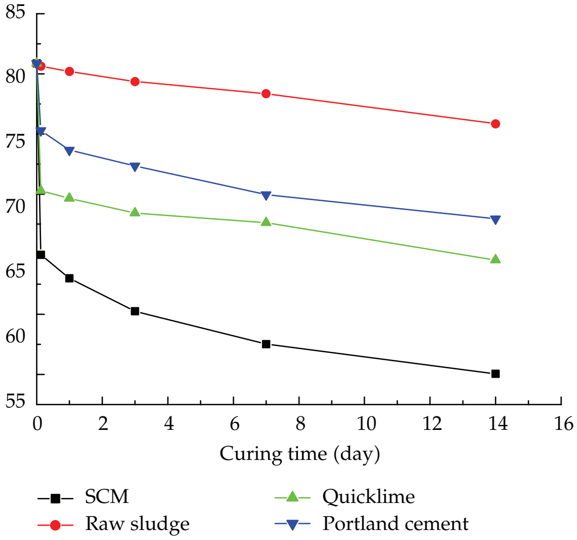


Figure 2: Moisture content of raw sludge and stabilized sludge (10 wt% material) versus curing time.

3.3. *Influence of Curing Time on Stabilized Sludge Moisture Content*

As shown in Figure 2, moisture content of raw sludge and both three stabilized samples decreased with the duration of stabling process. Here the three stabilized mixtures of sludge-SCM, sludge-quicklime, and sludge-Portland cement were prepared with a same content of 10%.

Obviously the drop of raw sludge moisture content, a relative slower one, is attributed to air-based water evaporation. For all three materials, the moisture content of stabilized mixtures drops fast at early ages (3 h), and then gradually slow at later ages. But compared with quicklime-added samples, the later reduction in moisture content of SCM-added sample is more significant, which may be due to further hydration of SCM and correspondingly the continuous chemical binding of free water in sludge.

Thus, for quicklime, sludge drying only depends on the physical evaporation, while for SCM, sludge drying depends on both physical evaporation and hydration reactions between modifier components and free water.

3.4. *Analysis of SCM Hydration Products*

From XRD patterns of SCM pastes shown in Figure 3, it can be seen that after 1 day of curing, ettringite ($3CaO{\cdot}Al_2O_3{\cdot}3CaSO_4{\cdot}32H_2O$, abbreviated as AFt) phase and calcium silicate hydrate ($CaO{\cdot}SiO_2{\cdot}nH_2O$, briefed as C-S-H) phase had been formed and AFt was the major hydration product. When it extended to 3 days of hydration, the major peaks of AFt and C-S-H became stronger.

Ettringite is a mineral which rarely occurs in nature but is widely present in the mineralogy of hydrated cements. Water is especially important to ettringite because of the high water content of the solid, which has the constitution, $3CaO{\cdot}Al_2O_3{\cdot}3CaSO_4{\cdot}32H_2O$. With a higher water content of about 46% in ettringite, it can convert more free water in sludge into crystal water. Therefore, from the XRD analysis, it can be inferred that the formation of AFt

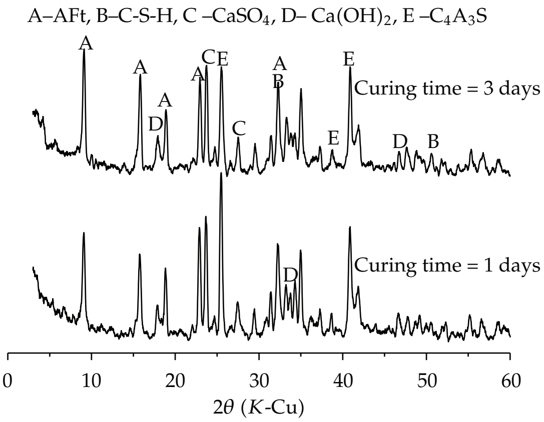


Figure 3: XRD pattern of SCM after hydration for 1 and 3 days.

plays an important part for the sludge drying. Besides, it can be concluded that the combined use of slag/cement clinker/dihydrate gypsum, together with their proportion optimization, promotes the appearance of AFt and in turn leads to the dramatic moisture content reduction.

3.5. SEM Analysis

SEM observations of raw sewage sludge and the dried sludge with SCM are shown in Figure 4. As shown in Figure 4(a), the platy construction of raw sewage sludge is disordered and incompact. What is more, the raw sewage sludge exhibited a poor structure with a rough and granular texture with a large amount of open porosity. Figure 4(b) shows that large amounts of hydration products (AFt and C-S-H) cover on the surface of sludge particles, and the particles aggregate together due to agglutination caused by the formation of crystal structure of AFt and C-S-H. It reveals that the addition of SCM and the mixing procedure resulted in a dense and homogeneous microstructure with less porosity. It may be the reason why the porosity ratio and water content of the dried sludge declines significantly.

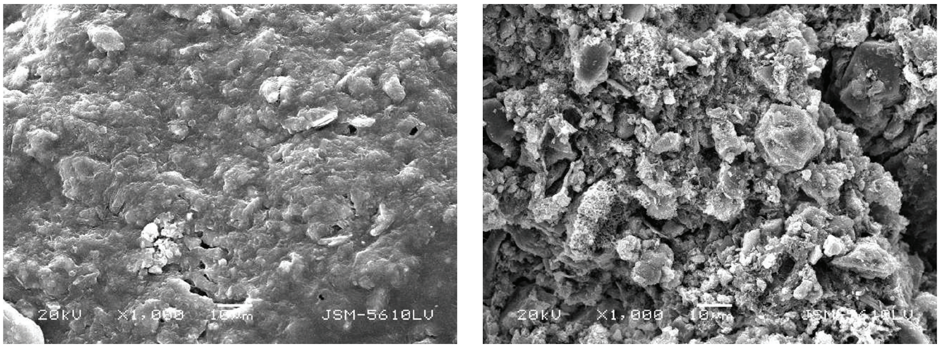
The SEM analysis of raw sludge and stabilized sludge also confirmed that the combined use of slag, cement clinker, and dihydrate gypsum, together with their proportion optimization, promoted the appearance of AFt and in turn leaded to the dramatic moisture content reduction. Thus, both sludge drying and stabilization process took place through the speedy formation of AFt and calcium silicate hydrate, converting most free water of sludge into crystal water.

4. Conclusions

The following conclusions can be drawn from this investigation.

(1) A composite modifier is proposed for sludge drying and stabilization. The experimentally optimized mix ratio of slag to cement clinker to dihydrate gypsum of 0.64 : 0.292 : 0.068.

(2) The moisture content of SCM-added sludge decreases with the increasing of SCM content and the extending curing time. However, with the further increase of SCM



(a) (b)

Figure 4: SEM micrographs of sludge: (a) raw sewage sludge and (b) dried sludge after 3 days of curing.

content and extending of curing time, the tendency of decrease slows down, due to the limited free water in sludge.

(3) Comparing with Portland cement, SCM shows better drying effect. Besides, the comparative experiment with SCM and quicklime reveals that SCM is more effective for semi-drying treatment of sludge, in which 40–60% water content was achieved.

(4) XRD analysis and SEM analysis of SCM hydration products demonstrated that combined use of slag, cement clinker, dihydrate gypsum, together with their mixing ratio optimization, result in a quick formation of AFt, which may play an important role in sludge drying and stabilization.

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