Feasibility and Carbon Footprint Analysis of Lime-Dried Sludge for Cement Production

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Abstract: Cement manufacturing and the treatment of sludge are considered both energy-intensive industries and major greenhouse gas (GHG) emitters. However, there are still few studies on comprehensive carbon footprint analysis for adding municipal sludge in the cement production. In this study, the lime-dried sludge blended with calcium oxide at the mass mixing ratio of 10% was utilized as raw material for the preparation of Portland cement. The chemical and physical properties of sludge were analyzed. A set of carbon footprint calculation methods of lime-drying treatment of sludge and reuse in cement kilns was then established to explore the feasibility of coprocessing lime-dried sludge in cement kilns. The results showed lime-dried sludge containing CaO, SiO₂, Al₂O₃, and Fe₂O₃ was ideal for cement production as raw material. However, the water content of lime-dried sludge should be strictly limited. The lime-drying process presented the biggest carbon emission (962.1 kg CO₂-eq/t sludge), accounting for 89.0% of total emissions. In the clinker-production phase, the lime-dried sludge as raw material substitute and energy source gained carbon credit of 578.8 and 214.2 kg CO₂-eq/t sludge, respectively. The sludge used for producing cement clinker could reduce carbon emissions by 38.5% to 51.7%. The addition ratio of lime and stacking time in the sludge lime-drying process could greatly affect the carbon footprint of coprocessing lime-dried sludge in cement kiln.

Keywords: wastewater treatment plant; lime-dried sludge; cement production; energy; carbon footprint

1. Introduction

Due to rising urbanization and the increase of wastewater treatment ratio in China, a large amount of activated sludge has been generated as byproduct during the wastewater treatment process [1]. Currently, the treatment of sludge has become one of the significant challenges for wastewater treatment plants (WWTPs) [1]. Although near 80% of WWTPs in China are equipped with sludge-handling facilities, there are still over 80% of municipal not treated well [2]. Particularly, the sludge has not been well stabilized due to the lack of anaerobic digestion facilities. Moreover, in order to meet the more stringent landfilling standard, the moisture content of the sludge needs to be further reduced. In recent years, many sanitation agencies have invested sludge-drying facilities to achieve lower water content and volume. Typically, thermal and lime drying are two commonly used sludge-drying methods [3,4]. The lime-drying technique is widely and frequently adopted in WWTPs due to its advantages such as...
simple operation, low cost, and almost odor-free [4,5]. Some applications in Xiaohongmen WWTP and Fangzhuang WWTP in Beijing, and Jiading WWTP in Shanghai, are using a large amount of lime for further sludge dewatering.

Many studies on adding lime-dried sludge in cement production have been conducted in recent years [6–9]. Since the basic chemical compositions of lime-dried sludge (mainly including CaO, Al₂O₃, Fe₂O₃ and SiO₂) in cement production are very close to the traditional raw materials, lime-dried sludge can partially replace raw materials in the production of cement. Cocombustion of lime-dried sludge in cement-producing kilns has the advantages of feasibility and environmental protection, which renders it a proper alternative technology [10].

As climate change is occurring, the reduction of energy and greenhouse gas (GHG) emissions has become a significant global concern [11]. Cement production consumes a large amount of energy and is one of the major sources of GHG emissions, accounting for 26% of the emissions from all industries [12,13]. Literature reported that the GHG emissions from Portland cement-manufacturing clinker were around 850 kg CO₂/ t clinker, with the consuming of energy of 850 kcal/kg clinker [14]. Due to its high dependence on natural resources, the industry of cement production may have the challenge of resource shortage in the future. Thus, it’s urgent for the industry to reduce natural resource consumption, explore alternative solutions and conduct carbon footprint assessment [15,16].

Carbon footprint assessment can help identify and recognize the energy consumption and carbon emission of different parts and stages of human activities. Many researchers have conducted carbon footprint assessment on the cement production industry [13,17] and tried to explore GHG reduction measures [18]. Although different approaches have been used to address the carbon footprint of cement production in recent years, there are still few studies on comprehensive carbon footprint analysis for adding municipal sludge in cement production, which requires the consideration of carbon debit and carbon credit emissions from the sludge for its whole life cycle. Therefore, this study aims to investigate the feasibility of utilizing lime-dried sludge for cement production and analyze the energy balance as well as carbon footprint. The results can provide technical support for the sludge resource reuse in China.

2. Materials and Methods

2.1. Materials

In this study, raw sewage sludge (RS) sample was derived from a WWTP in Beijing, China, where an Anaerobic/Anoxic/Oxic (A²/O) process was adopted. Lime-dried sludge (LS) was prepared by blending RS with calcium oxide (CaO) at the mass mixing ratio of 10%. The sludge samples were dried at 105 °C for 24 h and then crushed into particles with the size of 2 mm in order to analyze the water content and to remove the moisture in the sludge without destroying the chemical composition, preparing for the subsequent analysis. The pH value of the sludge samples was analyzed by a pH meter (HACH Company, USA). According to the sludge test standard in China (CJ/T 221-2005), organic matter was measured by weight method and total nitrogen (TN) was analyzed by alkaline potassium persulfate digestion followed by UV-vis spectrophotometry. Total carbon (TC) were analyzed by a TOC analyzer (Shimadzu, Japan). The characteristics of raw sewage sludge and lime-dried sludge are shown in Table 1.

| Table 1. Characteristics of raw sewage sludge (RS) and lime-dried sludge (LS). |
|-----------------|----------------|---|---|---|----------------|
| **Sample**     | **Water Content (%)** | **Organic Matter (%)** | **pH** | **TN (mg/g)** | **TC (mg/g)** | **Lower Heating Value (LHV) (MJ/kg)** |
| RS             | 86.6            | 60.4            | 7.16   | 50.7          | 370.6         | 15.9                             |
| LS             | 74.4            | 25.5            | 12.04  | 23.7          | 172.5         | 7.2                              |
2.2. Analytical Methods

Sludge samples of around 0.6 g were taken to measure their heat value by adopting oxygen bomb type-calorimeter. Loss on ignition (LOI) was determined by using the ASTM C114 method (section 18), which are standard test methods for chemical analysis of hydraulic cement published by ASTM International. X-Ray Fluorescence (XRF) was utilized to analyze the chemical composition of sludge samples and cement raw material. The Differential Scanning Calorimetry (DSC) characteristic in the thermal process was investigated by using a STA 449C Jupiter Synchronous thermal analyzer (Netzsch, Germany) at a rate of 10 °C/min.

2.3. Carbon Footprint

The steps to establish carbon footprint calculation methods of lime-drying treatment of sludge and reuse in cement kilns mainly include determining the accounting boundary and inventory, developing the accounting model and conducting the calculation. Based on the guidelines of the Intergovernmental Panel on Climate Change (IPCC) and the United Nations Framework Convention on Climate Change (UNFCCC), this study determined the accounting boundary and inventories, analyzed the materials and energy consumed, and finally established the calculation methods, combined with the case study of the WWTP and cement kiln in Beijing, China.

2.4. Sensitivity Analysis

As for sludge lime-drying, the amount of lime addition has a significant influence on carbon emission. Moreover, the water content of sludge varies with the duration of lime treatment. It has been reported that stacking sludge for a week could significantly reduce water content and improve thermal efficiency [5]. Thus, the lime dosage and treatment duration were identified as two key parameters for sensitivity analysis in this study.

3. Applicability of Lime-Dried Sludge to Cement Production

3.1. Raw Material Substitution Analysis

The chemical compositions of raw materials for cement production, RS and LS, were measured and compared, as shown in Table 2 and Figure 1. Limestone had the highest CaO content (more than 96%), which was the main source for cement clinker. As for the clay raw materials such as clay shale and sandstone, Al₂O₃ was the principal ingredient, accounting for 72% and 90% of the total chemicals, respectively. Iron ore with a high percentage of Fe₂O₃ was also used as a type of raw material. Both RS and LS presented the similar chemical compositions, while the chemical composition of LS was closer to that of the raw materials in terms of LOI and CaO. Some MgO and P₂O₅ were also found in RS and LS. Appropriate amount of P₂O₅ in sludge could help the formation of cement clinker [5], but the excessive P₂O₅ could also significantly reduce tricalcium silicate (C₃S) [19,20], which is a significant component that could influence the quality of cement clinker.

Figure 2 shows that the water contents of RS and LS were 80.3% and 46.3%, respectively. As a result, the ash content of LS was approximately 5 times more than that of RS. The addition of lime dramatically decreased the water content of the sludge, consequently increasing inorganic LOI. This result also suggested LS could provide more cement raw materials.

| Table 2. Chemical compositions of CRM, RS, and LS (average, wt%). |
|----------------|-----|------|-----|------|-----|-----|-----|
| Sample | LOI   | SiO₂  | CaO | Al₂O₃ | Fe₂O₃ | MgO | P₂O₅ |
| CRM    | 33.85 | 14.15 | 42.64| 4.29  | 3.48  | 0.94 | 0.06 |
| RS     | 65.26 | 6.09  | 3.22 | 5.74  | 6.09  | 0.98 | 6.02 |
| LS     | 38.47 | 3.32  | 42.49| 1.81  | 3.81  | 1.02 | 3.29 |
3.2. Themostability of Sludge

Thermogravimetric (TG) Analysis and Differential Scanning Calorimetry (DSC) were adopted to investigate the possible influence of the thermal treatment on RS and lime-dried sludge. As shown in Figure 3a, the RS basically went through three stages of weight loss. The weight of RS decreased by 10% in the first stage from 150 to 240 °C due to the volatile organic emissions. In the second stage from 240 to 640 °C, the combustion of massive organic matter contributed to around 55% weight loss [21]. After 640 °C in the third stage, only 1.4% of weight loss was observed due to the thermal decomposition of the minerals [22,23]. Similarly, the TG curve in Figure 2 represents three weight loss stages. From 0 to 400 °C, the lime-dried sludge lost much less weight than RS did, due to the dilution of organic chemical in the sludge. Moreover, a certain portion of organic matter reacted with metal ions forming thermally stable chelate compounds during this period. Some relatively thermally stable chelate started decomposing from 400 to 700 °C, leading to 5.96% weight loss [24–28]. Over 700 °C it showed 19.7% weight loss and an apparent endothermic peak at approximately 800 °C attributed to the breakdown of CaCO$_3$ from the sludge [29]. Therefore, RS as well as LS could provide calorific value during the temperature-rise period. Furthermore, CaO in the sludge could be important composition for partial replacement of raw material in the production of cement.
3.3. Energy Balance Analysis

The energy balance of the sludge sample in the cement kiln is one of the most important concerns for its applicability to cement production. The sludge sample as a part of raw materials intended for cement production not only provides thermal energy but also consumes energy for evaporation. When the sludge sample is calcined in cement kiln, the energy balance can be calculated by:

\[ H_N = H_S - [W \cdot C \cdot (100 - T) + W \cdot Q_W], \tag{1} \]

where \( H_N \) is the net energy value when the sludge sample is burned in cement kiln (MJ/t sludge); \( H_S \) is the lower heat value of the sludge (MJ/t sludge); \( W \) is the water content of LS; \( C \) is the specific heat of water which is 4.2 kJ/kg °C; \( T \) is the temperature of the sludge (°C), which is assumed 25 °C; and \( Q_W \) is heat of evaporation of 1 kg water, 2.26 MJ/kg.

As shown in Table 1, the lower heating value of RS was slightly higher than the average value of municipal sludge in China \[30\]. Raw sludge with 86.6% water content possessed −98.2 J/g of residual calorific value. The addition of lime would cause the dilution of organic chemical concentration in sludge, resulting in the decreasing of the heat value of LS. It can be calculated that LS with 74.4% water content exhibited −496.5 MJ/t lime-dried sludge of residual calorific value. The negative heat value

![Figure 3. Thermogravimetric (TG) Analysis and Differential Scanning Calorimetry (DSC) curves for raw sewage sludge (a) and lime-dried sludge (b).](image-url)
of LS suggested that additional fuel was needed to compensate the thermal energy. Therefore, the water content of sludge was a significant constraint in the combustion of sludge, which should be strictly controlled.

4. Carbon Footprint Analysis

4.1. Carbon Footprint Calculation Model

In order to study the characteristics of GHG emissions from this particular sludge treatment and reuse route, a set of carbon footprint calculation methods was established. Figure 4 shows the process of sludge handling in WWTP and the utilization of lime-dried sludge with cement raw materials for clinker production. The system boundary for carbon footprint accounting in this study included indirect fossil CO\textsubscript{2} emissions associated with electricity use for sludge mechanical equipment and polymers use for sludge thickening and dewatering, direct CO\textsubscript{2} emission from transportation, as well as the N\textsubscript{2}O emissions from the rotary kiln. In addition, carbon credit, also referred to as avoided emission, may be acquired in the phase of cement production due to the provision of cement raw materials and calorific value. The function unit for carbon footprint analysis was 1 ton of dry matter raw sewage sludge. All material flows were normalized to this functional unit.

![Diagram process of utilization of lime-dried sludge for cement production.](image)

4.1.1. Calculation of fossil CO\textsubscript{2} emissions

The electricity and chemical consumption during the sludge-handling process results in CO\textsubscript{2} emission. The CO\textsubscript{2} emissions associated with electricity and chemical use are calculated as follows:

\[ E_{Ele} = \sum_i E_{Ele,i} \cdot EF_{Grid}, \] (2)

\[ E_{Chm} = \sum_i E_{Chm,i} \cdot EF_{Chm}, \] (3)

where \( E_{Ele} \) and \( E_{Chm} \) are the CO\textsubscript{2} equivalent emissions due to electricity generation and chemical use, respectively (kg CO\textsubscript{2}-eq/t sludge); \( E_{Ele,i} \) is the electricity consumption for each sludge-handling unit (kWh/t sludge); \( E_{Chm,i} \) is the chemical consumption for each sludge-handling unit (kg/t sludge); \( EF_{Grid} \) is the regional grid emission factor which varies with energy source portfolio of the power plant (kg CO\textsubscript{2}-eq/kWh); \( EF_{Chm} \) is the CO\textsubscript{2} emission factor of the chemical (kg CO\textsubscript{2}-eq/kg); and \( i \) is the unit type.
It was assumed that the transportation tool of lime-dried sludge is a truck with hauling capacity of 20-ton load. The fossil CO$_2$ emission attributed to the transportation can be determined by the following equation:

$$E_{Tr} = (E_{Tr,CO_2} \cdot D) + 25(E_{Tr,CH_4} \cdot D) + 298(E_{Tr,N_2O} \cdot D),$$

where $E_{Tr}$ is the CO$_2$ equivalent emissions due to the transport of lime-dried sludge to cement kiln site by truck (kg CO$_2$-eq/t sludge); $E_{Tr,CO_2}, E_{Tr,CH_4}$ and $E_{Tr,N_2O}$ are the emission factors for CO$_2$, CH$_4$ and N$_2$O by truck per ton sludge per kilometer distance, 0.179, 0.018x10$^{-4}$, and 0.049x10$^{-4}$ kg/t/km, respectively (US EPA); $D$ is the transportation distance (km), assumed about 50 km from the WWTP to the cement plant; and 25 and 298 are the global warming potential of CH$_4$ and N$_2$O, respectively.

4.1.2. Calculation of N$_2$O emissions from Kiln

Nitrogen in sludge could be converted into a minor amount of N$_2$O during incineration in a rotary kiln. The N$_2$O emission can be estimated by the following equation:

$$E_{Kil} = 298 \cdot EF_{Kil} \cdot C_N,$$

where $E_{Kil}$ is the CO$_2$ equivalent emissions from cement rotary kiln (kg CO$_2$-eq/t sludge); $EF_{Kil}$ is the N$_2$O emission factor (kg N$_2$O/kg N), which is 0.007 accordingly [31]; $C_N$ is the nitrogen content of sludge (kg N/t sludge); and 298 is the global warming potential of N$_2$O.

4.1.3. Calculation of Carbon Credit

The carbon credit can be obtained if the heat value of the sludge is more than the energy required for the evaporation of water. Therefore, the sludge can be seen as alternative fuel for cement production process. If not, then additional fuel is required to achieve neutral energy balance. The energy balance value $H_N$ is obtained in the section of 3.3. Assuming that coal was used as the additional fuel, the CO$_2$ emission from heat consumption can be calculated by the following equation:

$$E_{Fs} = H_N \cdot EF_{Coal},$$

where $E_{Fs}$ represents the CO$_2$ emission from fuel consumption (kg CO$_2$-eq/t sludge) and $EF_{Coal}$ represents the CO$_2$ emission factor of coal (kg CO$_2$-eq/MJ), the value of which is 0.23 (IPCC, 2006).

The substitution of lime-dried sludge for a certain amount of cement raw materials was also regarded as the carbon credit in this study. According to the LOI of lime-dried sludge, it was calculated that 1.08 tons of ash from 1.75 ton of lime-dried sludge was incorporated into the cement products, replacing the same amount of raw materials. The raw material substitution could help reduce the amount of carbon emission mainly generated from carbonate decomposition during the production of cement. The process emission factor in China is 536 kg CO$_2$/t cement (Cao et al., 2016). Therefore, the avoided CO$_2$ equivalents emission was 578.8 kg CO$_2$-eq/t MSS.

4.2. Carbon Footprint Results

Carbon emissions of each LS treatment unit for cement production are shown in Figure 5. The lime-drying process presented the biggest carbon emission (962.1 kg CO$_2$-eq/t sludge), accounting for 89.0% of total emissions as a result of large amounts of lime consumption. Sludge thickening and dewatering together showed carbon emissions of 114.3 kg CO$_2$-eq/t sludge, and attributed to electricity consumption. The transport of LS to cement plant provided negligible contribution (0.7%) to the overall emissions, and it was in accordance with the results for other sludge management alternatives [32,33].

For the clinker-production phase, it gained a carbon credit of -214.2 kg CO$_2$-eq/t sludge because of positive energy balance of the LS in the kiln. In addition, the LS as raw material substitute in the rotary kiln contributed negative carbon emissions of -578.8 kg CO$_2$-eq/t sludge, and the N$_2$O emission
from the sludge was at a relatively low level. The clinker-production phase itself achieved a net carbon emission of \(-993.0 \, \text{kg CO}_2/-\text{eq/t sludge}\) on account of carbon credit by using LS as energy and cement raw material resource. The overall carbon emission for the whole process was \(362 \, \text{kg CO}_2/-\text{eq/t sludge}\). Compared with the conventional landfilling of sludge with emission in the range of 1564 to 1992 kg CO\(_2\)/t sludge in China \([34,35]\), the sludge in this study used for producing cement clinker could reduce carbon emissions by 38.5% to 51.7%. Therefore, it is recommended the LS being landfilled can be applied to manufacturing cement clinker in terms of carbon reduction effect.

**Figure 5.** Carbon footprint of lime-dried sludge for cement production.

4.3. Sensitivity Analysis

The effect of lime dosage and duration of stacking on water content was investigated and the results are presented in Figure 6. The water content obviously declined as the stacking time increased. When the stacking time was 7, 14, and 21 days, the water content of sludge with 10% lime reduced from 86.6% to 46.3%, 16.3% and 8.4%, respectively. Besides, the increase in lime addition also led to the decrease of water content of sludge.

A sensitivity analysis with four different lime dosages and stacking durations was conducted to determine their effects on the carbon footprint. Figure 7a depicts the effect of 5%, 10%, 15%, and 20% lime dosage stacking for 1 day on the carbon footprint. The results showed that both the carbon debit and carbon credit increased with the lime dosage. As a result, the net emission presented a rising trend. That is to say, increasing the lime dosage would lead to more carbon footprint. Although the addition of lime in sludge can generate avoided emission through raw material substitution, the appropriate amount of lime addition should retain a relative low level. Figure 7b shows the effect of LS (10% lime) stacking time on carbon footprint. The carbon credit to offset raw materials increased as the stacking time rose, while the carbon debit did not change. This was because increasing stacking time could significantly reduce the moisture in the sludge and therefore saving more thermal energy for evaporation. Finally, the net carbon footprint decreased by approximately 66.2% from 1 to 7 days of stacking. This clearly indicated that the stacking time played an important role in reducing carbon footprint of LS for cement production.
Figure 6. Water content of LS with different lime addition.

Figure 7. Effect of lime dosage (a) and stacking time (b) on carbon footprint.
5. Conclusions

The lime-dried sludge and cement raw material had similar chemical compositions such as CaO, SiO$_2$, Al$_2$O$_3$, and Fe$_3$O$_4$. The energy balance of lime-dried sludge for cement production was dependent on the lime addition and water content. Carbon footprint results indicated that the lime-drying process presented the biggest carbon emission (962.1 kg CO$_2$-eq/t sludge), accounting for 89.0% of total emissions, with 10% of lime addition and water content of 46.3%. In the clinker-production phase, the lime-dried sludge as raw material substitute and energy source gained carbon credit of 578.8 and 214.2 kg CO$_2$-eq/t sludge, respectively. The sludge used for producing cement clinker could reduce carbon emissions by 38.5% to 51.7%. The addition ratio of lime and stacking time in sludge lime drying process could greatly affect the carbon footprint of coprocessing lime-dried sludge in cement kilns.

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