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Using sewage sludge as a denitration agent and secondary fuel in a cement plant: A case study

Ping Fang, Zi-Jun Tang, Jian-Hang Huang, Chao-Ping Cen *, Zhi-Xiong Tang, Xiong-Bo Chen

South China Institute of Environmental Sciences, Ministry of Environmental Protection, Guangzhou 510655, Guangdong, China

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ABSTRACT

The influences of sludge feed rate, feed point, feed method, and air-staged combustion were systematically investigated in a Chinese cement plant with emphasis on NO_X removal. Results indicate that the use of sludge as a secondary fuel is conducive to NO_X reduction, which depends primarily on the feed rate and feed point. Moreover, both feed method and air-staged combustion influence NO_X removal to some extent. Nonetheless, these factors have little effects on CO removal. When the sludge feed rate is 9 $t \cdot h^{-1}$ in a precalciner reburning zone in the combustion temperature range from 850 to 1000 °C, NO_X removal reached 75.82% in the air-staged combustion condition. The NO_X concentration easily met the national emission standards. Finally, the use of sludge as alternative fuel does not affect clinker quality negatively. This method utilizes sludge effectively and reduces NO_X emissions; hence, the application of sewage sludge as a denitration agent and secondary fuel in cement production is a worthy option.

1. Introduction

As a result of the rapid progress of urbanization and the continuous improvement of sewage treatment capacity in China, an increasing amount of municipal sewage sludge is produced annually [1]. The composition of sludge is complex and contains toxic and hazardous substances. Therefore, its improper disposal pollutes the environment severely [2]. Technologies have been developed to handle sewage sludge, and the most common treatments are agricultural utilization, landfilling, and incineration [3–7]. Approximately 45% of the sludge produced in China is utilized in agriculture; 34.5% is landfilled; 3.5% is incinerated; 3.5% is used for greening, and 13.5% is untreated [1]. These treatment technologies are important in practice; however, sludge agriculture and landfilling application are limited by the reduced availability of land, increased public concern with environmental risks, potential contamination of the food chain by heavy metals from sludge. The use of sludge incineration technology is controversial in this country because of the resultant environmental pollution and the cost [1,8]. Thus, the disposition of sludge safely and with economic efficiency is a major environmental problem of urban development in China.

According to data obtained from the national bureau of statistics of China [9], a total of 2.42 billion t of cement were manufactured in this country in 2013. This amount accounted for approximately 50% of the global annual output. Over 90% of this cement was produced by the new suspension preheater (NSP) cement production lines. The cement industry has contributed significantly to China's economic development; however, Portland cement clinker production consumes large amounts of raw materials (limestone and clay) and fuel (coal, oil, and gas) while generating large amounts of air pollutants. For instance, particle, SO_2, and NO_X emissions in this industry accounted for approximately 15%–20%, 3%–4%, and 10%–12% of total emissions, respectively, in 2011 [10]. Therefore, the cement industry has become the third-largest industrial source of NO_X emission in China [11]. As a result, this industry has garnered increasing attention as a major emitter of air pollution [12]. Most cement plants in China are equipped with flue gas cleaning systems [bag filter + wet flue gas desulfurization (wet limestone–gypsum process) units] that effectively control particle and SO_2 pollution. Low NO_X burners and staged combustion technologies have also been widely used to control NO_X; however, their removal is only roughly 30%. Thus, they fail to meet emissions standards. The selective non-catalytic reduction (NH_3-SNCR) method was introduced in response [13], and the combination of the techniques described above can control NO_X effectively. Nonetheless, the NH_3-SNCR technology remains problematic (high cost, ammonia supply and storage, ammonia slip, and production safety issues) and must be improved further. Therefore, the identification of other economical raw materials and the development of cost-efficient $DeNO_X$ technology are immediate and long-term concerns in this regard.

Sewage sludge is a high-potential alternative as a secondary fuel and raw material for the cement industry because of its calorific power and inorganic chemical composition [14,15]. It has multiple advantages, such as the capability to reduce fuel and raw material use, to dispose safely of sludge, and to lower production costs [16]. Hence, the utilization of sludge in the cement industry is considered a sustainable sludge-disposal method in many countries, including Japan and the

* Corresponding author. Tel./fax: +86 2085557692.
E-mail address: cenchaoping@scies.orig (C.-P. Cen).

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United States [17]. In China, sewage sludge has gradually been applied in practice as a secondary fuel in cement plants. However, researchers have expressed concern that the co-processing of sludge in cement kilns may emit toxic substances [heavy metals and dioxins (PCDD/Fs)] [18,19]. Furthermore, the environmental impact and potential risks to human health are uncertain. Nonetheless, recent works have revealed that the co-processing of sludge in cement kilns is feasible and does not risk the health of nearby residents. Moreover, it does not change the overall environment [20–23]. However, knowledge on the use of sewage sludge as a denitration agent to control the NO_X emissions from cement kiln flue gas is limited.

In the current study, a sequence of experiments is conducted to evaluate the potential of dried sewage sludge as a denitration agent in cement clinker production with NSP cement production lines. The optimal experimental conditions are established, and the denitration reaction mechanism in the process is hypothesized through theoretical analysis and experimental simulation.

2. Material and methods

2.1. Drying of dewatered sludge

The dewatered sludge was obtained from the municipal wastewater treatment plants around the Yue Bao cement plant. The moisture content of this sludge was approximately 78%–83%. Given the high moisture content and low calorific value of dewatered sludge, the management of the cement plant built a sludge drying plant as well. High-temperature (*ca*. 300 °C) flue gas from a cement kiln was used to heat the dewatered sludge, and the moisture content in the sludge was reduced from 78% to less than 30% after drying. The dewatered sludge treatment capacity of the plant was 600 $t \cdot d^{-1}$, and the annual treatment capacity reached 186,000 t.

The sludge was pretreated, stirred, and crushed. Meanwhile, the flue gas from the cement kiln was dedusted in a cyclone. The sludge swirled around a spouted dryer, and the dedusted flue gas was utilized to dry the sludge to a moisture content of below 30%. The fine-grain dried sludge was collected by a baghouse and then conveyed to a precalciner through a measuring pocket and a screw pump. Finally, the grains were burned in the precalciner.

2.2. Sewage sludge characterization

The dried sludge sample was obtained from the sludge drying plant constructed within the Yue Bao cement plant, which is located in the suburbs of Guangzhou City (South China). The dried sludge was used for co-disposal in the cement plant. The sample was dried further at 105 ± 2 °C for 24 h and then analyzed. As Table 1 lists, the sludge has lower fixed carbon content than anthracite, as well as higher concentrations of volatiles, inorganics, nitrogen, and sulfur elements. The calorific value easily exceeds the minimum requirement of the cement industry for alternative fuels (6250 $J \cdot g^{-1}$) [24]. As listed in Table 2, the main chemical compounds of sludge and its ashes are Al_2O_3, SiO_2, Fe_2O_3, and CaO. These compounds are also the primary compounds of some raw materials in cement production. Thereby, sludge can theoretically be used as an alternative fuel and as a raw material in this regard.

Elemental analysis was performed by oxidizing the sample at 980 °C. Combustion products (CO_2, H_2O, NO, and SO_2) were detected subsequently using an elemental analyzer equipped with a TCD detector (EuroVector-EA3000, Italy). Net calorific value was determined using a calorimeter (Bo Hai-ZDHW8, China). The chemical composition of the sludge and its ashes was confirmed by a wavelength dispersion X-ray fluorescence spectrometer (ZSX Primus II, Japan).

2.3. Description of the cement production process

The Yue Bao cement factory is a modern cement production enterprise and employs two NSP cement production lines with a clinker capacity of approximately 6000 $t \cdot d^{-1}$ per line. The experiment was conducted on one of these lines. As illustrated in Fig. 1, the kiln system had three main parts: two groups of five-stage cyclone preheater systems, a large volume precalciner with a volume of roughly 3260 m^3, and a rotary kiln (Φ5.2 m × 70 m). Cement production is a multistage process that involves raw material preparation, preheating, and calcination, as well as clinker burning and cement grinding. First, the raw material is dried by the hot exit gases from the precalciner and the rotary kiln. Part of the carbonate is decomposed in this stage. Then, the dry raw material is calcined and decomposed in the precalciner, which is located between the rotary kiln and the preheater section. The calcination process can almost be completed (>90%) before the raw material enters the rotary kiln when 60% of the total fuel (coal) of the kiln system is added to a secondary combustion chamber that is installed in the precalciner. The process begins by decomposing $CaCO_3$ at roughly 900 °C to retain CaO and CO_2. In the subsequent clinker burning process, oxidizing conditions and a maximum material temperature of 1450 °C must be maintained to facilitate the required sintering reactions. Finally, the clinker is obtained, cooled, and then ground together with gypsum (approximately 5%) and other materials to produce cement. The flow type between the gas and solid phases is countercurrent in the production process.

In the production process, the dried sludge provided 3%–10% of alternative energy and was fed to the precalciner. As shown in Fig. 1, there are two sludge feed points in the precalciner. Point A was in the reburning zone, in which the temperature was roughly 850 °C–1000 °C. Point B was in the primary combustion zone, where the temperature was approximately 1000 °C–1100 °C. In this study, a sequence of experiments was conducted to evaluate the influences of different operational parameters on NO_X removal. Specifically, the influences of feed rate, feed point, and air-staged combustion were investigated systematically. The system stability must be maintained for at least 1 h when the operating condition changes. Concentrations of pollutants (NO, CO, and O_2) were monitored continuously using a flue gas analyzer (testo350XL, Germany) for 2 h. As Fig. 1 displays, the detection point was located on the flue gas outlet pipe of the five-stage cyclone preheater system.

NO_X removal (R_{NOx}) is calculated according to the equation:

$$R_{NOx} = (A - B)/(A) \tag{1}$$

where *A* and *B* denote the NO_X concentrations in the original flue gas and the flue gas when sludge is used as secondary fuel, respectively.

Table 1
Sewage sludge and anthracite characterization.

Sample	Proximate analysis (wt.%)				Ultimate analysis (wt.%)				$S_{t,d}$ (wt.%)	Net calorific value ($J \cdot g^{-1}$, ar)
	M_{ar}	A_d	V_d	FC_d^{diff}	C_{daf}	H_{daf}	N_{daf}	O_{daf}^{diff}		
Sludge	3.60	58.96	39.94	1.10	56.31	8.33	10.09	>24.14	1.13	11,147.32
Anthracite	6.48	21.48	19.44	59.08	78.28	3.50	1.87	>15.60	0.75	23,039.50

diff: calculated by difference.