



Mechanisms of hydrogen sulfide removal by ground granulated blast furnace slag amended soil



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HIGHLIGHTS

- GGBS amended soil removes H₂S through oxidation and acid-base reaction.
- Major products of H₂S-GGBS amended soil reaction were AVS, S(0) and thiosulfate.
- AVS plays a significant role in H₂S removal during regeneration and reuse cycles.
- Higher soil water content results in increased removal capacity.

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ABSTRACT

Ground granulated blast furnace slag (GGBS) amended soil has been found able to remove gaseous hydrogen sulfide (H₂S). However, how H₂S is removed by GGBS amended soil and why GGBS amended soil can be regenerated to remove H₂S are not fully understood. In this study, laboratory column tests together with chemical analysis were conducted to investigate and reveal the mechanisms of H₂S removal process in GGBS amended soil. Sulfur products formed on the surface of soil particle and in pore water were quantified. The test results reveal that the reaction between H₂S and GGBS amended soil was a combined process of oxidation and acid-base reaction. The principal mechanism to remove H₂S in GGBS amended soil was through the formation of acid volatile sulfide (AVS), elemental sulfur and thiosulfate. Soil pH value decreased gradually during regeneration and reuse cycles. It is found that the AVS plays a significant role in H₂S removal during regeneration and reuse cycles. Adding GGBS increased the production of AVS and at the same time suppressed the formation of elemental sulfur. This mechanism is found to be more prominent when the soil water content is higher, leading to increased removal capacity.

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1. Introduction

Ground granulated blast furnace slag (GGBS) is a by-product of iron and steel industry. Large amounts of GGBS are generated each year (e.g., production capacity of 10 million tons, “K. Wah Construction Material,” 2016). GGBS is rich in minerals, finely granulated and highly alkaline. Its most popular use is to replace cement in concrete to improve strength, durability, decrease permeability and retard setting (Oner and Akyuz, 2007). GGBS has also been sometimes used for soil solidification and stabilization

(Kogbara and Al-Tabbaa, 2011).

Landfill is a source of odorous gas, mainly in the form of H₂S. The odorous gas would migrate through landfill cover soil and cause serious environmental problems. GGBS has been shown to be an effective soil conditioner to reduce H₂S concentration (Ng et al., 2017). The laboratory study shows that GGBS amended soil could reduce H₂S to a level lower than the olfactory threshold of 0.02 ppm (i.e., the lowest H₂S concentration that human nose could sense), and it can be regenerated multiple times to maintain its removal capacity. The mechanisms involved in H₂S removal and its regeneration/reuse are, however, unclear. Factors that control the capacity of GGBS for H₂S removal are not known.

Linz-Donawitz Steel Slag (LD) and Steel Making Slag (SMS) have been also found effective in removal of H₂S (Kim et al., 2012; Montes-Morán et al., 2012). Steel slags have comparable Ca and

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Mg contents with GGBS. Kim et al. (2012) reported that in steel making slag, Ca and Mg content is about 45% and 5%, both of which are fairly similar to that found in GGBS. The existing studies show that elemental sulfur S(0) can be found as a product of the LD-H₂S reaction. Kim et al. (2012) estimated sulfur transformation during the removal of aqueous H₂S by SMS and found that the major products were S(0) and manganese sulfide (MnS). Sulfur transformation in unsaturated soil could be affected by the amount of soil water content, which would affect the physical state of reactant (e.g., gaseous or aqueous), hence the reaction kinetics.

The objective of this paper is to quantify the sulfur transformation and phase transfer upon H₂S removal by GGBS amended unsaturated soil. Sulfur products in soil samples before/after reaction and during each regeneration/reuse cycles were measured. Influences of soil water content on the removal mechanisms are then investigated.

2. Materials and methods

2.1. Material properties

Loess soil (silty clay; ASTM D2487-11) was collected from Xi'an, China. GGBS was provided by K. Wah Construction Company, Hong Kong. Loess soil samples were amended with 0% and 30% (by mass) GGBS. pH values of loess and GGBS are 8.4 and 11.7, respectively. pH value of loess amended with 30% GGBS is 11.7. Measurements show that after adding 30% GGBS (mean particle size of GGBS is 9 μm), the mean particle size of amended soil shifts from 35 μm to 28 μm. Metal contents of loess soil and GGBS were obtained using X-ray fluorescence (XRF), and they are summarized in Table S1 in the supplementary information (SI). Chemico-physical properties of loess, GGBS and their mixtures were measured and are listed in Table S2 in the SI. Water used in all the tests in this study was ultrapure water. Chemicals were provided by Sigma-Aldrich.

2.2. Sample preparation and analysis methods

Dynamic H₂S removal tests and regeneration tests were carried out. Loess soil was amended with 0% and 30% GGBS, compacted to the same bulk density (1.54 g/cm³) in a soil column. A concentration of 1000 ppm H₂S balanced in nitrogen was supplied from the bottom of each column at a constant rate of 50 mL/min. H₂S concentration at column outlet was measured using a gas chromatograph equipped with a flame photometric detector (GC-FPD, GC-2014C Shimadzu Scientific Instruments). The tests would stop when H₂S breakthrough took place. H₂S breakthrough is defined when the H₂S concentration at the column outlet reaches the olfactory threshold of 0.02 ppm. H₂S removal capacity is defined as the maximum sulfur (sulfur in H₂S, unit mg) that can be removed by 1 g of soil (bulk mass) before H₂S breakthrough. Regeneration method was air ventilation. Detailed test procedures are reported by (Ng et al., 2017). In order to investigate the effects of soil water content on H₂S removal capacity and removal mechanisms, GGBS amended soils with different gravimetric water contents (i.e., 0%, 10% and 20%) were tested. For the GGBS amended soil with water content of 20%, three regeneration and reuse cycles were applied. The testing program is shown in Table 1.

After each column test, two soil samples (around 4 g each) were collected from the lower part of the soil column. These two samples were placed into two separate 250 mL pyrex glass bottles, namely A and B. Bottle A was used for the measurements of the concentration of soluble sulfide, sulfate and thiosulfate in soil water, while bottle B was used to measure the concentration of elemental sulfur S(0) and acid volatile sulfide (AVS) on soil particle. Detailed measurement procedures of these chemicals are given in the next section.

Table 1
Testing program.

Soil condition	Water content	Regeneration cycle	Sample ID ^a	
			Before H ₂ S	After H ₂ S
Loess	15%	—	L	LH
Loess +30%GGBS	20%	—	L30G	L30GH
Loess +30%GGBS	20%	1	R1	R1H
		2	R2	R2H
		3	R3	—
Loess +30%GGBS	0%	—		
	10%			
	20%			

^a RX is the soil sample after the Xth cycle of regeneration, RXH is the soil sample RX after reacting with H₂S. For example, R1 is the soil sample L30GH after the 1st cycle of regeneration. R1H is the soil sample R1 after reacting with H₂S. R2 is the soil sample R1H after the 2nd cycle of regeneration.

Both bottles A and B contained 30 mL of ultrapure water and 5 drops of 10 M sodium hydroxide (NaOH), aiming to increase the pH to prevent sulfide ion from forming H₂S. Soil in the bottle A was agitated by magnetic stirrers to facilitate soluble sulfide, sulfate and thiosulfate to dissolve in the water. After agitation, the soil-water mixture was allowed to stand and segregate. A flow chart of chemical measurements can be found in Fig. S1 in the SI. Each condition was tested for two replicates.

2.2.1. Measurements of soluble sulfide, sulfate and thiosulfate in soil pore water

Supernatant from the bottle A was filtered through 0.45 μm filter (Sartorius Stedim), and the filtrate was collected. The filtrate was firstly taken for measuring the concentration of soluble sulfide using the methylene blue method (APHA, 2005). In this method: 1 drop of 10N NaOH was added into 6 mL filtrate sample, and then 0.4 mL amine sulfuric acid and 0.12 mL ferric chloride (FeCl₃) solution were added to filtrate sample. The filtrate was mixed and stood for 5 min, and then 1.28 mL diammonium hydrogen phosphate solution was added to the filtrated sample. Subsequently the sample stood for 20 min to let precipitates to settle down, and then the supernatant was collected and measured with methylene blue absorbance at 664 nm using a UV/Vis spectrophotometer (Lambda 25, Perkin Elmer Inc., USA). The device has a cuvette that provides a light path of 10 mm, and has a measuring range of sulfide from 0 mg/L to 1 mg/L.

5 mL filtrate from the bottle A was also collected and added with a drop of 1 M zinc acetate (Zn(Ac)₂) and a drop of 6 M NaOH. The mixture was then allowed to stand for 10 min for the precipitates of ZnS to settle. Then the supernatant was filtered through 0.45 μm filter again, and the filtrate was used for the measurements of soluble sulfate and thiosulfate, by an ion chromatograph (100, Dionex, USA) equipped with a conductivity detector and an IonPac AS9-HC analytical column.

2.2.2. Measurements of insoluble AVS, elemental sulfur S(0) on soil particle surface

Measurements of AVS were performed by acidifying samples (USEPA, 1991). Bottle B was firstly purged with nitrogen gas (N₂). Then 20 mL concentrated hydrogen chloride (HCl) was added to the soil sample, followed by agitating using a magnetic stirrer. Gas generated from the acid-treated soil was stripped into two serial traps filled with 1 M NaOH solution. After the acid treatment, N₂ gas was injected into the acid-treated soil for one hour continuously to remove any remaining H₂S. Details of the testing apparatus are provided in Fig. S2 in the SI. After an hour of N₂ injection, H₂S absorbed in the NaOH solution was quantified using the methylene blue method (APHA, 2005). The amount of sulfide available in AVS

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