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

2

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14

15 Abstract

16 Ground granulated blast furnace slag (GGBS) amended soil has been found able to remove

17 gaseous hydrogen sulfide (H<sub>2</sub>S). However, how H<sub>2</sub>S is removed by GGBS amended soil and

18 why GGBS amended soil can be regenerated to remove H<sub>2</sub>S are not fully understood. In this

19 study, laboratory column tests together with chemical analysis were conducted to investigate

20 and reveal the mechanisms of H<sub>2</sub>S removal process in GGBS amended soil. Sulfur products

21 formed on the surface of soil particle and in pore water were quantified. The test results

22 reveal that the reaction between H<sub>2</sub>S and GGBS amended soil was combined process of

23 oxidation and acid-base reaction. The principal mechanism to remove H<sub>2</sub>S in GGBS amended

24 soil was through the formation of acid volatile sulfide (AVS), elemental sulfur and

25 thiosulfate. Soil pH value decreased gradually during regeneration and reuse cycles. It is

26 found that the AVS plays a significant role in H<sub>2</sub>S removal during regeneration and reuse  
27 cycles. Adding GGBS increased the production of AVS and at the same time suppressed the  
28 formation of elemental sulfur. This mechanism is found to be more prominent when the soil  
29 water content is higher, leading to increased removal capacity.

30

31 Keywords

32 H<sub>2</sub>S, GGBS, sulfur, AVS, regeneration

33

34 1. Introduction

35 Ground granulated blast furnace slag (GGBS) is a by-product of iron and steel industry.  
36 Large amounts of GGBS are generated each year (e.g., production capacity of 10 million tons,  
37 “K. Wah Construction Material,” 2016). GGBS is rich in minerals, finely granulated and  
38 highly alkaline. Its most popular use is to replace cement in concrete to improve strength,  
39 durability, decrease permeability and retard setting (Oner and Akyuz, 2007). GGBS has also  
40 be sometimes used for soil solidification and stabilization (Kogbara and Al-Tabbaa, 2011).

41

42 Landfill is a source of odorous gas, mainly in the form of H<sub>2</sub>S. The odorous gas would  
43 migrate through landfill cover soil and cause serious environmental problems. GGBS has  
44 been shown to be an effective soil conditioner to reduce H<sub>2</sub>S concentration (Ng et al., 2016).  
45 The laboratory study shows that GGBS amended soil could reduce H<sub>2</sub>S to a level lower than  
46 the olfactory threshold of 0.02 ppm (i.e., the lowest H<sub>2</sub>S concentration that human nose could  
47 sense), and it can be regenerated multiple times to maintain its removal capacity. The  
48 mechanisms involved in H<sub>2</sub>S removal and its regeneration/reuse are, however, unclear.  
49 Factors that control the capacity of GGBS for H<sub>2</sub>S removal are not known.

50

51 Linz-Donawitz Steel Slag (LD) and Steel Making Slag (SMS), which have similar  
52 composition with GGBS, have been also found effective in removal of H<sub>2</sub>S (Kim et al., 2012;  
53 Montes-Morán et al., 2012). The existing studies show that elemental sulfur S(0) can be  
54 found as a product of the LD-H<sub>2</sub>S reaction. Kim et al (2012) estimated sulfur transformation  
55 during the removal of aqueous H<sub>2</sub>S by SMS and found that the major products were S(0) and  
56 manganese sulfide (MnS). Sulfur transformation in unsaturated soil condition (which is often  
57 the case for a landfill cover), on the other hand, would be substantially different because soil  
58 water content may play a major role. This is because water could influence the physical state  
59 of reactant (e.g., gaseous or aqueous), hence the reaction kinetics.

60

61 The objective of this paper is to quantify the sulfur transformation and phase transfer upon  
62 H<sub>2</sub>S removal by GGBS amended unsaturated soil. Sulfur products in soil samples before/after  
63 reaction and during each regeneration/reuse cycles were measured. Influences of soil water  
64 content on the removal mechanisms are then investigated.

65

## 66 2. Materials and methods

### 67 2.1 Material properties

68 Loess soil (silty clay) was collected from Xi'an, China. GGBS was provided by K. Wah  
69 Construction Company, Hong Kong. Loess soil samples were amended with 0% and 30% (by  
70 mass) GGBS. pH values of loess and GGBS are 8.36 and 11.67, respectively. pH value of  
71 loess amended with 30% GGBS is 11.74. Measurements show that after adding 30% GGBS  
72 (mean particle size of GGBS is 9.33 μm), the mean particle size of amended soil shifts from  
73 35.36 μm to 27.87 μm. Metal contents of loess soil and GGBS were obtained using X-ray  
74 fluorescence (XRF), and they are summarized in Table 1. Chemico-physical properties of

75 loess, GGBS and their mixtures were measured and are listed in Table 2. Water used in all  
76 the tests in this study was ultrapure water. Chemicals were provided by Sigma-Aldrich.

77

## 78 2.2 Sample preparation and analysis methods

79 Dynamic H<sub>2</sub>S removal tests and regeneration tests were carried out. Loess soil was amended  
80 with 0% and 30% GGBS, compacted to the same bulk density (1.54 g/cm<sup>3</sup>) in a soil column.

81 A concentration of 1000 ppm H<sub>2</sub>S was supplied from the bottom of each column at a constant  
82 rate of 50 mL/min. The tests would stop when H<sub>2</sub>S breakthrough took place. H<sub>2</sub>S

83 breakthrough is defined when the H<sub>2</sub>S concentration at the column outlet reached the

84 olfactory threshold of 0.02 ppm. H<sub>2</sub>S removal capacity is defined as the maximum sulfur

85 (sulfur in H<sub>2</sub>S, unit mg) that can be removed by 1 g of soil (bulk mass) before H<sub>2</sub>S

86 breakthrough. Regeneration method was air ventilation. Detailed test procedures are reported

87 by (Ng et al., 2016). In order to investigate the effects of soil water content on H<sub>2</sub>S removal

88 capacity and removal mechanisms, GGBS amended soils with different gravimetric water

89 contents (i.e., 0%, 10% and 20%) were tested. For the GGBS amended soil with water

90 content of 20%, three regeneration and reuse cycles were applied. The testing program is

91 shown in Table 3.

92

93 After each column test, two soil samples (around 4 g each) were collected from the lower part

94 of the soil column. These two samples were placed into two separate 250 ml pyrex glass

95 bottles, namely A and B. Bottle A was used for the measurements of the concentration of

96 soluble sulfide, sulfate and thiosulfate in soil water, while bottle B was used to measure the

97 concentration of elemental sulfur S(0) and acid volatile sulfide (AVS) on soil particle.

98 Detailed measurement procedures of these chemicals are given in the next section. Both

99 bottles A and B contained 30 ml of ultrapure water and 5 drops of 10 N sodium hydroxide

100 (NaOH), aiming to increase the pH to prevent sulfide ion from forming H<sub>2</sub>S. Soil in the bottle  
101 A was agitated by magnetic stirrers to facilitate soluble sulfide, sulfate and thiosulfate to  
102 dissolve in the water. After agitation, the soil-water mixture was allowed to stand and  
103 segregate. A flow chart of chemical measurements can be found in Fig. S1 in the  
104 supplementary information (SI). Each condition was tested for two replicates.

105

#### 106 2.2.1 Measurements of soluble sulfide, sulfate and thiosulfate in soil pore water

107 Supernatant from the bottle A was filtered through 0.45 µm filter (Sartorius Stedim), and the  
108 filtrate was collected. The filtrate was firstly taken for measuring the concentration of soluble  
109 sulfide using the methylene blue method (APHA, 2005). In this method: 1 drop of 10N  
110 NaOH was added into 6 ml filtrate sample, and then 0.4 ml amine sulfuric acid and 0.12 ml  
111 ferric chloride (FeCl<sub>3</sub>) solution were added to filtrate sample. The filtrate was mixed and  
112 stood for 5 min, and then 1.28 ml diammonium hydrogen phosphate solution was added to  
113 the filtrated sample. Subsequently the sample stood for 20 min to let precipitates to settle  
114 down, and then the supernatant was collected and measured with methylene blue absorbance  
115 at 664 nm using a UV/Vis spectrophotometer (Lambda 25, Perkin Elmer Inc., USA) with a  
116 cuvette providing a light path of 10 mm, and a sulfide measuring range of 0 to 1 mg/L.

117

118 5 ml filtrate from the bottle A was also collected and added with a drop of 1 N zinc acetate  
119 (Zn(Ac)<sub>2</sub>) and a drop of 6N NaOH, and it was mixed and allowed to stand for 10 min for the  
120 precipitates of ZnS to settle). Then the supernatant was filtered through 0.45 µm filter again,  
121 and the filtrate was used for the measurements of soluble sulfate and thiosulfate, by an ion  
122 chromatograph (100, Dionex, USA) equipped with a conductivity detector and an IonPac  
123 AS9-HC analytical column.

124

125 2.2.2 Measurements of insoluble AVS, elemental sulfur S(0) on soil particle surface

126 Measurements of AVS were performed by acidifying samples (USEPA, 1991). Bottle B was  
127 firstly purged with nitrogen gas (N<sub>2</sub>). Then 20 ml concentrated hydrogen chloride (HCl) was  
128 added to the soil sample, followed by agitating using a magnetic stirrer. Gas generated from  
129 the acid-treated soil was stripped into two serial traps filled with 1 N NaOH solution. After  
130 the acid treatment, N<sub>2</sub> gas was injected into the acid-treated soil for one hour continuously to  
131 remove any remaining H<sub>2</sub>S. Details of the testing apparatus are provided in Fig. S2 in the SI.  
132 After an hour of N<sub>2</sub> injection, H<sub>2</sub>S absorbed in the NaOH solution was quantified using the  
133 methylene blue method (APHA, 2005). The amount of sulfide available in AVS was obtained  
134 by subtracting the concentration of soluble sulfide obtained from the previous step (section  
135 2.2.1) from the concentration of sulfide measured in this procedure.

136

137 S(0) in the soil samples was measured by the revised method suggested by McGuire and  
138 Hamers (2000). After the acid treatment and one hour of N<sub>2</sub> purging, the sealed bottle B was  
139 added with 20 ml Tetrachloroethylene (C<sub>2</sub>Cl<sub>4</sub>), and then shaken continuously for four hours  
140 in a spinning shaker. Subsequently, C<sub>2</sub>Cl<sub>4</sub>, which carried extracted S(0), was collected and  
141 filtered through 0.2 μm membrane (Sigma-Aldrich). S(0) was measured using a high  
142 performance liquid chromatography (HPLC, LC-30AD, Shimadzu, Japan) equipped with a  
143 Waters symmetry C18 column (4.6 mm × 150 mm, 5 μm particle size) and a UV detector set  
144 at 254 nm. An eluent of 90% acetonitrile + 10% water was used at a flow rate of 1 mL/min.

145

146 It should be noted that any thiosulfate available in the bottle B would turn into sulfur dioxide  
147 (SO<sub>2</sub>) and S(0) once the soil was treated with concentrated HCl (see Equation [1]). Therefore,  
148 in order to analyze S(0) produced during H<sub>2</sub>S removal, it is required to subtract S(0)  
149 generated from thiosulfate from that measured by HPLC.

150



151

152 Soil samples were collected from each soil column for chemico-physical characterizations.

153 pH measurement was carried out according to the standard ASTM D4972-13 (ASTM, 2001)

154 using a pH meter (Oakton Instruments). Surface elements were identified by an X-ray

155 photoelectron spectroscopy (XPS, Axis Ultra DLD model). Microstructure was investigated

156 using a Scanning electron microscope (SEM, JSM 6300 (JEOL) model).

157

### 158 3. Results and discussions

159 In this study, the initial sulfur components of soil samples before reacting with H<sub>2</sub>S were

160 measured. Therefore, for the results presented herein, all sulfur products refer to the net sulfur

161 product formed by H<sub>2</sub>S reaction.

162

163 The major sulfur products in the soil during reaction, regeneration and reuse were identified

164 by the XPS spectrum (see Fig. S3 in the SI). According to the XPS spectra data of sulfur

165 species reported by Moulder (1992), the peaks of mineral sulfide, elemental sulfur,

166 thiosulfate and sulfate could be identified in a S(2p) XPS spectra at binding energies of 162.6

167 eV, 164 eV, 167.8 eV and 168.8 eV (see x-axis in Fig. S3), respectively. Therefore, the major

168 reaction products of H<sub>2</sub>S removal by the GGBS amended soil were metal sulfide, elemental

169 sulfur, thiosulfate and sulfate. This suggests that the chemical analysis were able to cover

170 most of the reaction products.

171

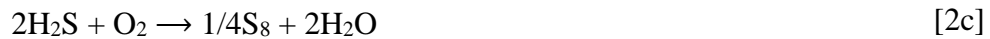


172 Fig. 1 shows that for LH and L30GH, H<sub>2</sub>S input is almost equal to the sum of all the  
173 measured sulfur products. This means that the mass balance of sulfur was almost achieved,  
174 and most of the sulfur products have been captured in these tests. For LH, almost all H<sub>2</sub>S was  
175 transferred into elemental sulfur, S(0). This was attributable to the oxidation of H<sub>2</sub>S by the  
176 minerals available in loess. For L30GH, H<sub>2</sub>S was transferred into 68% AVS, 13% S(0), 11%  
177 thiosulfate and 6% sulfide. It was found that the form of H<sub>2</sub>S present in soil was controlled by  
178 the pH value (Haimour et al., 2005; He et al., 2011). S<sup>2-</sup> is dominant when pH value is higher  
179 than 10. On the contrary, when pH is between 8 and 9, HS<sup>-</sup> is dominant, whereas no S<sup>2-</sup>  
180 exists in solution for any pH lower than 8. Since the pH value of L is 8.36 (Table 2), HS<sup>-</sup> was  
181 likely to be the main form of H<sub>2</sub>S existed in soil. As shown in Fig. 1, H<sub>2</sub>S in the sample LH  
182 was mainly oxidized into S(0), and very few sulfide was found. On the other hand, pH value  
183 of the soil sample L30G was 11.74 (Table 2), so it is not surprising to find more sulfide in the  
184 soil sample. This is similar to LD slag that its strong alkalinity would trigger dissociation of  
185 dissolved H<sub>2</sub>S into S<sup>2-</sup> and HS<sup>-</sup> (Montes-Morán et al., 2012). The results also show that  
186 adding GGBS could suppress the formation of S(0). This seems to indicate that the use of  
187 GGBS may be beneficial for improving the H<sub>2</sub>S removal capacity in regeneration cycles  
188 because precipitation of S(0) on the surface of soil particles could block the reactive sites  
189 substantially (Sun et al., 2014). Moreover, adding GGBS to soil increased the production of  
190 AVS, probably because of the higher mineral content in GGBS (Table 1) and increased  
191 surface activity due to its high alkalinity. The significance of having high AVS production is  
192 discussed later.

193

194 Fig. 2 (A) shows that sulfide ions decreased during regeneration, and increased during reuse.  
195 This was because of the dissolution of H<sub>2</sub>S in pore water during the reuse, and the oxidation  
196 of sulfide by O<sub>2</sub> during regeneration, according to the following chemical Equation [2].

197



198 (Chen and Morris, 1972; Davydov et al., 1998)

199

200 It should be noted that sulfide would be oxidized into different products when supplying  
201 different amount of O<sub>2</sub>, as indicated in Equation [2]. Although the formation of thiosulfate,  
202 sulfate and elemental sulfur through sulfide oxidation also contributed to the overall sulfur  
203 transformation, this was only at very small scale because the total sulfide in pore water was  
204 low (<0.06 mg/g).

205

206 Fig. 2 (B) shows that AVS decreased during regeneration, and increased during reuse.

207 Previous studies show that some AVS were very sensitive to O<sub>2</sub>. For instance, any exposure  
208 of AVS to O<sub>2</sub> would change the nature of some potential AVS minerals such as mackinawite

209 and greigite (Rickard and Morse, 2005). Possible mechanisms are given in Equations [3-4]

210 (which use iron mineral as an example; other minerals may apply). It can be seen from Fig. 2

211 (B) that although AVS changed during several regeneration and reuse, its content was within

212 a relatively constant range between 0.3 mg/g – 0.6 mg/g. This may be because part of the

213 AVS acted as a catalyst to remove H<sub>2</sub>S (see Equation [3-4]). During regeneration, AVS

214 turned into mineral oxide/hydroxide (e.g., Fe(OH)<sub>3</sub>), as shown in Equation [3a-d]. While

215 during reuse, mineral oxides/hydroxide reacted with H<sub>2</sub>S and AVS was formed again, as

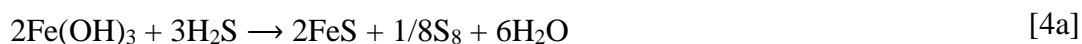
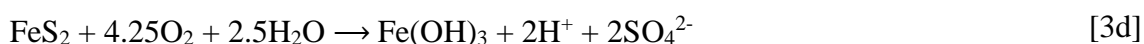
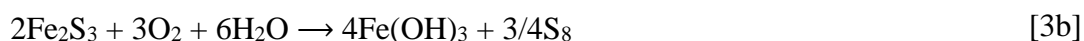
216 shown in Equation [4a-d]. This is the reason why GGBS amended soil could be regenerated

217 to remove H<sub>2</sub>S through air ventilation. This is similar to the mechanism when Linz-Donawitz

218 Steel Slag is used to remove H<sub>2</sub>S, the reaction during which the transition metal oxides and/or

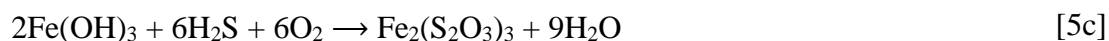
219 hydroxides would act as active catalysts to oxidize H<sub>2</sub>S to elemental sulfur (Montes-Morán et  
 220 al., 2012). It should be noted that for Equations [3a-c] and [4a], during both regeneration and  
 221 reuse, S(0) would be formed, and this may be the reason that caused continuous increase in  
 222 S(0) from L30GH to R3 (see Fig. 2 (B)).

223



224 (Davydov et al., 1998; Schippers and Jorgensen, 2002).

225



226 (Cantrell et al., 2003)

227

228 In Fig. 2 (A), the measured increase in sulfate ion during regeneration is likely to be  
 229 attributable to the oxidation of pyrite (Equation [3d]), as well as the oxidation of sulfide  
 230 (Equation [2b]). Biological oxidation of other reduced sulfur products might be another  
 231 reason that caused the increase in sulfate through R2 to R3. Sulfur oxidizing bacteria (SOB)

232 would use the energy of reduced sulfur compounds (e.g., H<sub>2</sub>S, thiosulfates, sulfites, and  
233 elemental sulfur), and then convert the reduced sulfur compounds to sulfate. The optimum  
234 pH values for SOB growth are typically between 1 and 9 (Pokorna and Zabranska, 2015). As  
235 can be seen in Fig. 3, during regeneration and reuse cycles, the pH value showed a decreasing  
236 trend from about 12 to 9. Thus, it is likely that SOB has been activated and produced sulfate  
237 through biological oxidation. The observed pH drop in Fig. 3 was attributable to the  
238 dissolution of H<sub>2</sub>S during reuse, and oxidation of sulfide ion and pyrite during regeneration.  
239 As indicated in Equation [2b] and [3d], both processes would produce hydrogen ion, hence  
240 results in decrease of pH value. At R3, the pH value was still around 9, indicating that HS<sup>-</sup>  
241 and S<sup>2-</sup> both existed and they were stable in the pore water.

242

243 On the other hand, the observed increase in thiosulfate during both regeneration (except for  
244 R2) and reuse (see Fig. 2 (B)) is likely to be the consequence of the oxidation of sulfide  
245 (Equation [2a]) and the aerobic reaction between mineral hydroxide and H<sub>2</sub>S during reuse  
246 (Equation [5c]). Since oxygen might dissolve in pore water during regeneration, aerobic  
247 reactions might have taken place between H<sub>2</sub>S and the soil, hence leading to the production  
248 of sulfate and thiosulfate during reuse. When comparing the reaction products from initial  
249 H<sub>2</sub>S removal (i.e., L30GH) and the removal after regeneration (i.e., R1H and R2H), it  
250 demonstrates that there was a change in removal mechanism. For the initial H<sub>2</sub>S removal, the  
251 main reaction product was AVS (Fig. 1), indicating that most H<sub>2</sub>S was bonded with the  
252 minerals in the soil. For H<sub>2</sub>S removal in the subsequent regenerated cycles, S(0) was  
253 accumulated while AVS remained at 0.3 mg/g – 0.6 mg/g (Fig. 2 (B)). This indicates that  
254 during regeneration/reuse cycles, the major removal mechanism of H<sub>2</sub>S was through the  
255 oxidization to S(0).

256

257 SEM images depicted in Fig. 4 show that for R3 (where most sulfur products were found  
258 compared to others), needle-like elemental sulfur crystal could be identified and it spreads  
259 around the surface of soil particles. Small amount of octahedral pyrite can also be seen. This  
260 further confirms the proposed removal mechanisms discussed in Equation [3], [4] and [5].  
261 Moreover, because the metal hydroxide/oxide generated from oxidation of AVS acted as a  
262 catalyst during the removal of H<sub>2</sub>S, the H<sub>2</sub>S removal capacity would not change much during  
263 each reuse cycle due to relatively constant range of AVS (Fig. 2 (B)). However, precipitation  
264 of elemental sulfur on particle surface would gradually reduce the availability of reactive  
265 sites and hinder further regeneration and reuse (Sun et al., 2014). Therefore, the H<sub>2</sub>S removal  
266 capacity was gradually reduced at the subsequent regeneration/reuse cycles (Ng et al., 2016).  
267

268 Fig. 5 shows the effects of different soil water contents on the removal capacity and removal  
269 mechanisms. The removal capacity increased with an increase in soil water content from 0%  
270 to 20%. This appears to agree with the findings reported by Montes-Morán et al (2012) who  
271 showed that the relative humidity of the slags particles changed dramatically with the H<sub>2</sub>S  
272 removal capacity. The water solubility of H<sub>2</sub>S is relatively high: 7100 mg/L at 0 °C, and 3925  
273 mg/L at 20 °C (Bergersen and Haarstad, 2008). Therefore, higher soil water content would  
274 result in more H<sub>2</sub>S dissolving in the pore water, and hence more H<sub>2</sub>S could be removed from  
275 its gas phase. Also, for a given soil dry density, increasing soil water content would decrease  
276 the pore air ratio. This would hence (i) reduce the effective diffusion coefficient, and  
277 therefore more effectively limit H<sub>2</sub>S migration and (ii) extend the retention time of H<sub>2</sub>S in the  
278 soil, resulting in a higher H<sub>2</sub>S removal capacity (Xu et al., 2014). Moreover, it can be seen in  
279 Fig. 5 that higher soil water content facilitated AVS formation and suppressed the formation  
280 of elemental sulfur. The AVS content increased from 0.166 to 0.527 mg/g (in percentage:  
281 from 36% to 68%), while the elemental sulfur decreased from 0.259 to 0.0988 mg/g (from

282 56% to 13%). Since AVS plays an important role in H<sub>2</sub>S removal during regeneration/reuse  
283 cycles (see Fig. 2 (B)), increasing AVS content by increasing soil water content is likely to be  
284 able to improve H<sub>2</sub>S removal capacity during regeneration. Because the accumulation of  
285 elemental sulfur would block the availability of reactive sites on the surface of soil particle,  
286 reducing elemental sulfur production by increasing soil water content may also improve H<sub>2</sub>S  
287 removal capacity in regeneration cycles. The test results imply that GGBS amended soil  
288 would be suitable to be used as in a landfill cover located in humid regions, because the  
289 increase in soil water content due to rainfall could improve the H<sub>2</sub>S removal capacity for not  
290 only the initial removal but also probably the removal in the subsequent regeneration/reuse  
291 cycles.

292

293 In Figs 5 and 6, it can be seen that for the first two regeneration cycles of the sample with  
294 20% of soil water content (i.e., from L30GH to R2 in Fig. 6), the removal capacities were  
295 almost equal to the sum of the measured sulfur products. For the third regeneration cycle of  
296 the samples (i.e., R2H and R3 in Fig. 6) and samples with lower water content (i.e., 0% and  
297 10% in Fig. 5), however, the sum of the measured sulfur products were higher than the  
298 removal capacities. This inconsistency may be associated with the reduction of reaction  
299 kinetics between H<sub>2</sub>S and those samples. It was demonstrated by Xu et al (2014) that in a  
300 diffusion-advection-reaction system, any change of reaction kinetics could affect the  
301 distribution of H<sub>2</sub>S in a soil bed. Hence, a non-uniform distribution of sulfur products along  
302 the soil column would be resulted, where the lower part would contain higher sulfur products,  
303 whereas the higher part contains less. Since the soil samples tested in the present study were  
304 taken at the lower part of the soil columns, their sulfur products content would be higher than  
305 the average sulfur content calculated from H<sub>2</sub>S input.

306

307 4. Conclusions

308 This study presents a set of comprehensive laboratory testing that provides insights into the  
309 pathways and mechanisms of how H<sub>2</sub>S would be removed by GGBS amended soil. The test  
310 results show that gaseous H<sub>2</sub>S could be removed by the GGBS in soil through oxidation and  
311 acid-base combined reactions. Using GGBS to react with H<sub>2</sub>S caused an increase production  
312 of acid volatile sulfide (AVS) and suppressed the formation of elemental sulfur. AVS has  
313 shown to play an important role in H<sub>2</sub>S removal during regeneration and reuse cycles. Soil  
314 pH value gradually decreased during regeneration and reuse cycles. Precipitation of  
315 elemental sulfur on particle surface was unfavorable for H<sub>2</sub>S removal. Increasing water  
316 content of GGBS amended soil up to a 20% (by weight) is favorable for H<sub>2</sub>S removal  
317 because this promoted H<sub>2</sub>S dissolution, simultaneously facilitating the formation of AVS and  
318 suppressing the formation of elemental sulfur.

319

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325

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**Table 1. Metal content (weight percentage of dry matter) obtained from XRF analyses**

Type	CaO	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	MgO	TiO <sub>2</sub>	K <sub>2</sub> O	MnO	Fe <sub>2</sub> O <sub>3</sub>
Loess	22.8	61.1	7.6	10.5	0.8	1.3	0.0	3.6
GGBS	37.9	34.2	13.8	8.1	1.0	0.6	0.5	0.3

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**Table 2. Properties of loess and amended loess tested in this study**

Soil condition	ID	G <sub>s</sub> (kg/m <sup>3</sup> ) <sup>a</sup>	pH value <sup>b</sup>	Mean particle size (μm) <sup>b</sup>	S <sub>s</sub> (m <sup>2</sup> /g)
Loess	L	2690	8.36	35.36	22.58
Loess+30% GGBS	L30G	2760	11.74	27.87	15.95
GGBS	-	2924	11.67	9.33	1.28

388 <sup>a</sup>G<sub>s</sub> is specific gravity  
389 <sup>b</sup> Mean value of three repeated tests  
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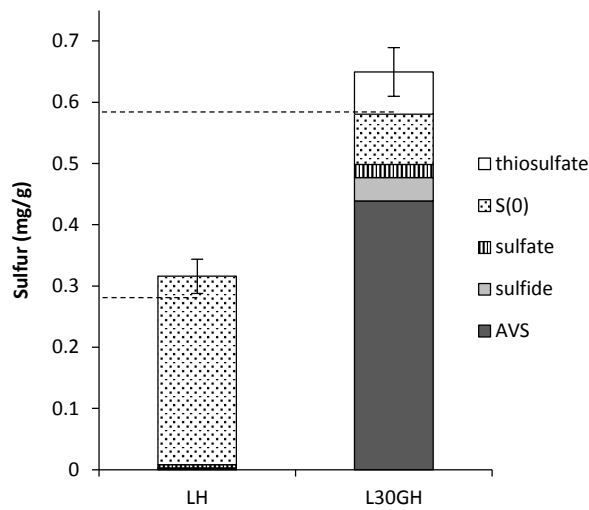
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**Table 3. Testing program**

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Soil condition	Water content	Regeneration cycle	Sample ID*	
			Before H <sub>2</sub> S	After H <sub>2</sub> S
Loess	15%	-	L	LH
Loess + 30%GGBS	20%	-	L30G	L30GH
		1	R1	R1H
		2	R2	R2H
Loess + 30%GGBS	20%	3	R3	-
		0%		
Loess + 30%GGBS	10%			
	20%			

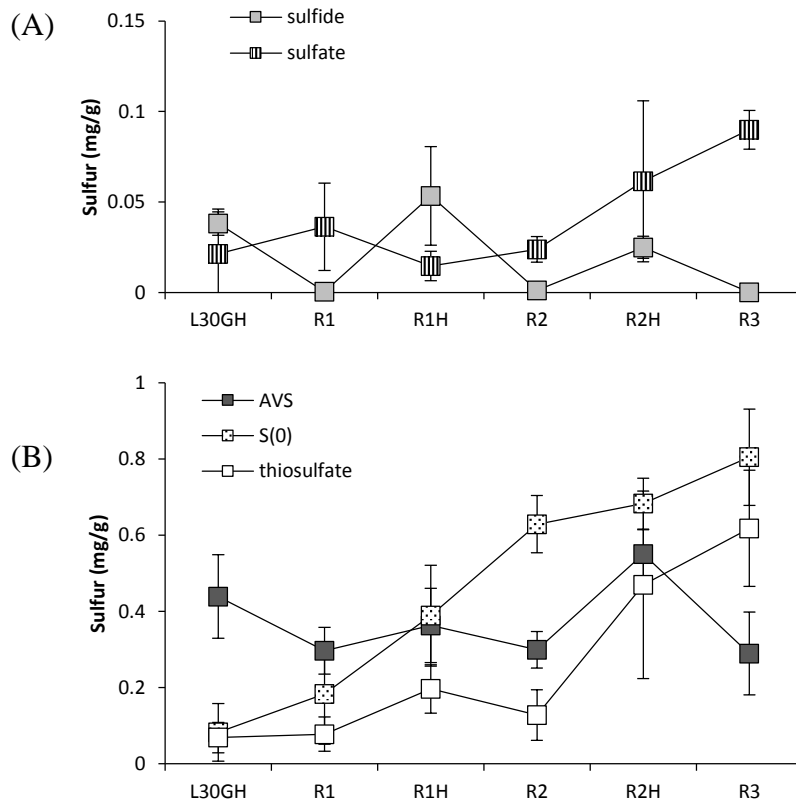
\* RX is the soil sample after the X<sup>th</sup> cycle of regeneration, RXH is the soil sample RX after reacting with H<sub>2</sub>S. For example, R1 is the soil sample L30GH after the 1<sup>st</sup> cycle of regeneration. R1H is the soil sample R1 after reacting with H<sub>2</sub>S. R2 is the soil sample R1H after the 2<sup>nd</sup> cycle of regeneration.



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Fig. 1. Sulfur products in soil after reaction with H<sub>2</sub>S. Dotted lines represent H<sub>2</sub>S input calculated from column tests, error bars represent mean absolute deviation. Sulfur content in y-axis is expressed as mg of sulfur per 1 g of bulk soil.

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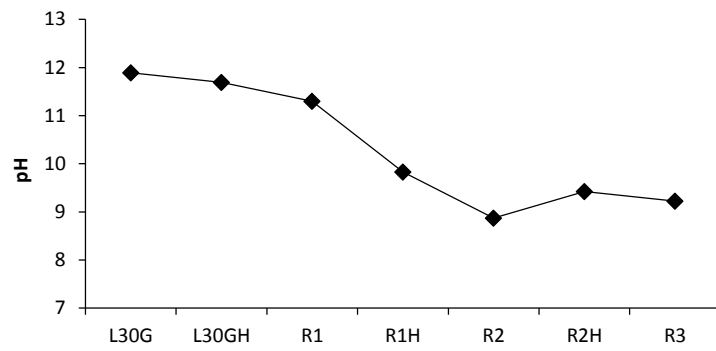
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Fig. 2. Sulfur transformation in regeneration/reuse of L30G. Error bars represent mean absolute deviation. Sulfur content in y-axis is expressed as mg of sulfur per 1 g of bulk soil.

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Fig. 3. pH value of L30G during regeneration and reuse

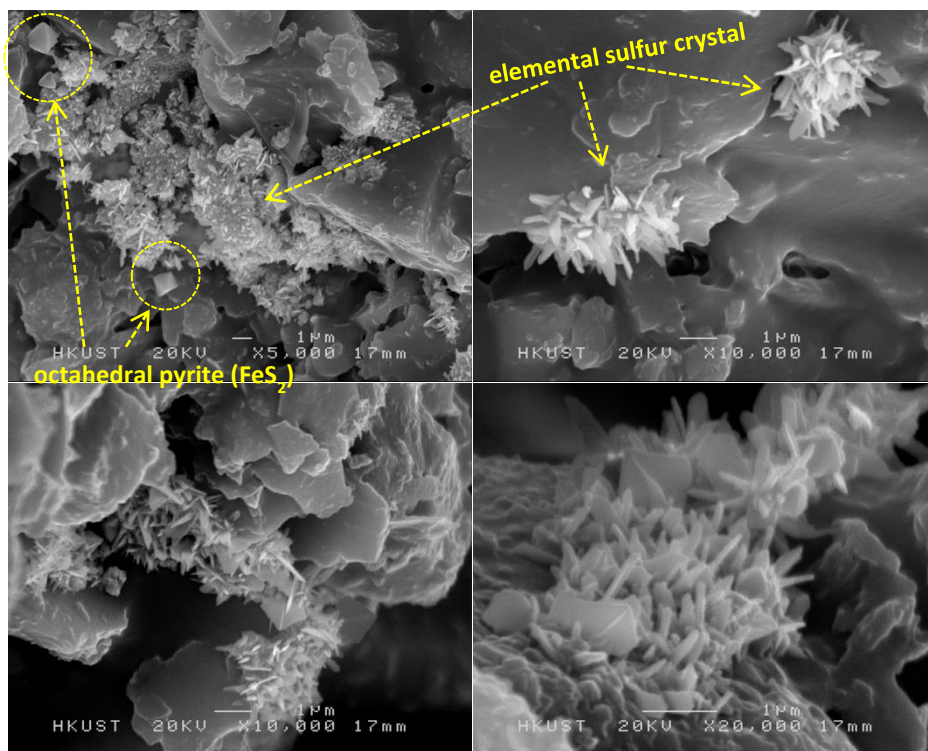
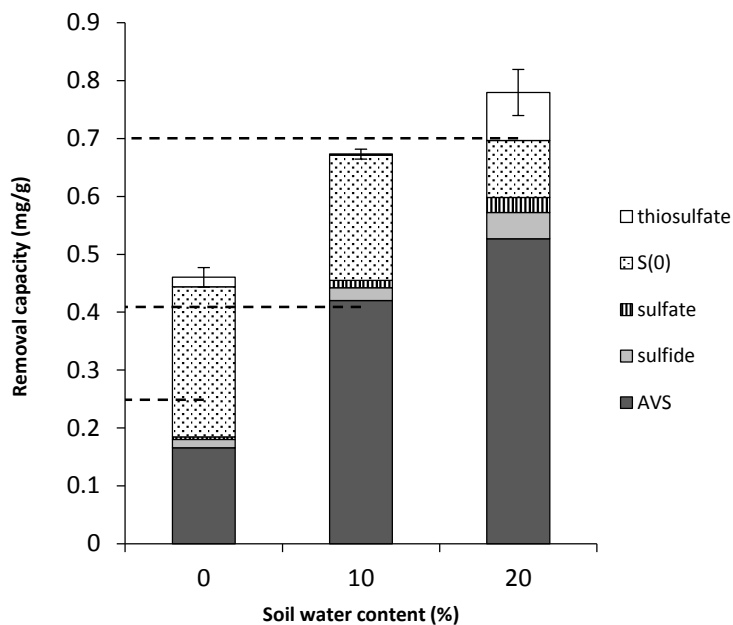


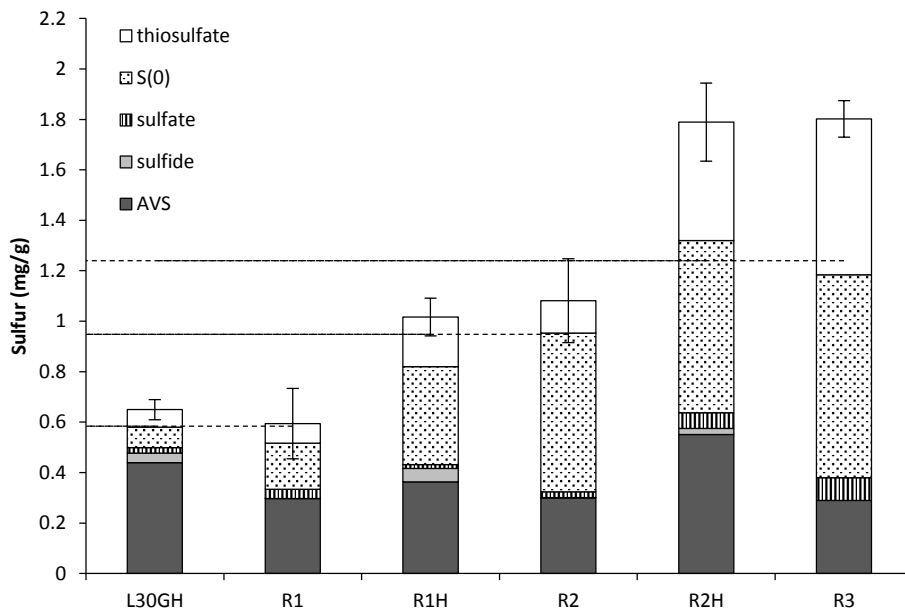
Fig. 4. SEM image of R3 to show the formation of elemental sulfur crystal and octahedral pyrite after reaction

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Fig. 5. Influence of water content on sulfur product. Dotted lines represent H<sub>2</sub>S input calculated from column tests. Error bars represent mean absolute deviation. Sulfur content in y-axis is expressed as mg of sulfur per 1 g of dry soil.

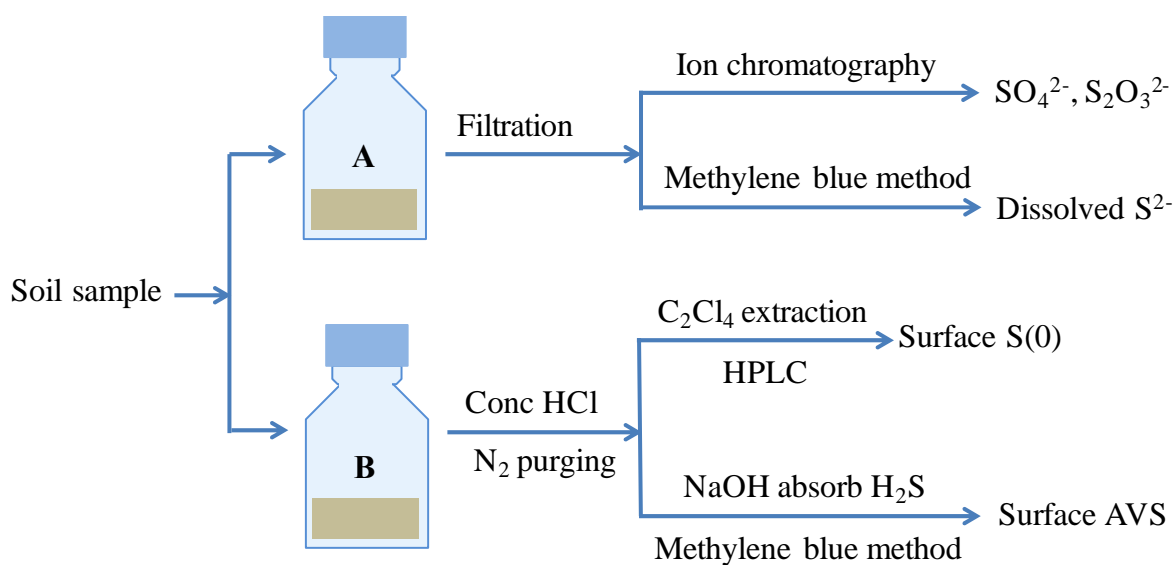


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Fig. 6. Sulfur products in L30G regeneration and reuse. Dotted lines represent H<sub>2</sub>S input calculated from column tests. Error bars represent mean absolute deviation. Sulfur content in y-axis is expressed as mg of sulfur per 1 g of bulk soil.

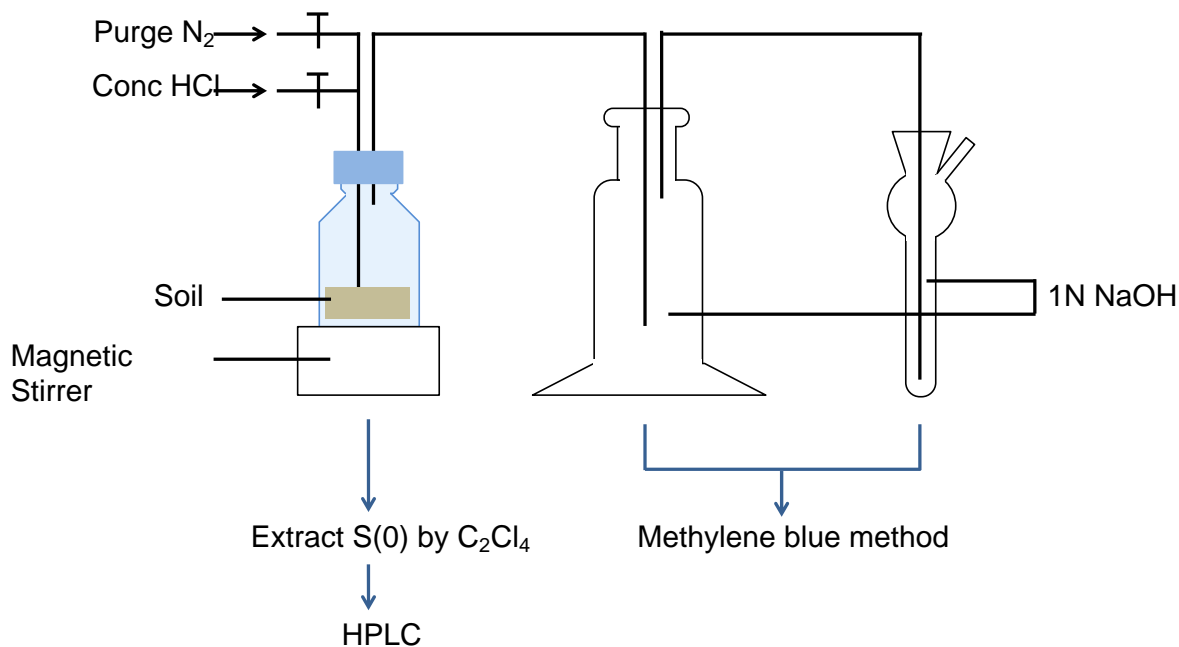
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### Supporting information



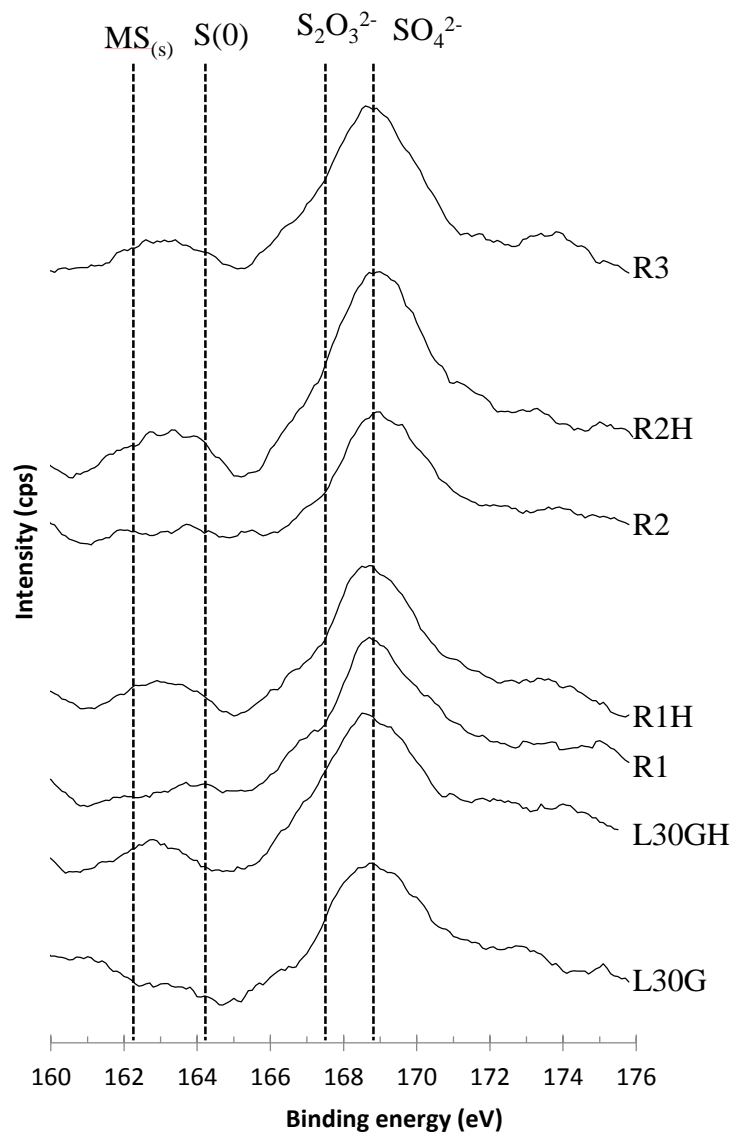
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**Fig. S1.** Flow chart of chemical measurements



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**Fig. S2.** Test setup for measurement of  $\text{S}(0)$



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**Fig. S3.** XPS results of sulfur products in soil samples