



Removal of hydrogen sulfide with granulated coal ash under aerobic and anaerobic conditions



Satoshi Asaoka^{a,*}, Waqar Azeem Jadoom^a, Takamichi Ishidu^b, Hideo Okamura^a, Takahito Oikawa^c, Kenji Nakamoto^c

^a Research Center for Inland Seas, Kobe University, 5-1-1 Fukaeminami, Higashinada, Kobe, 658-0022 Japan

^b Faculty of Maritime Sciences, Kobe University, 5-1-1 Fukaeminami, Higashinada, Kobe, 658-0022 Japan

^c The Chugoku Electric Power Co., Inc., 4-33, Komachi, Naka-ku, Hiroshima, 730-8701, Japan

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ABSTRACT

Annual emissions of fly ash from the burning of coal have increased to approximately 750 million tons. It is hoped that new applications utilizing by-products from coal-fired power plants will contribute to further waste reduction and set the standard for industrial-scale recycling. The purpose of this study was to evaluate the removal rate of hydrogen sulfide by a new recycled material called granulated coal ash, which is produced from coal fly ash from coal thermal electric power stations and blast furnace cement. A batch experiment was carried out to evaluate the removal rate of hydrogen sulfide by granulated coal ash under both anaerobic and aerobic conditions. The granulated coal ash could remove hydrogen sulfide under both anaerobic and aerobic conditions. However, the maximum removal of hydrogen sulfide by granulated coal ash was only 5.1 mg-S g^{-1} under anaerobic conditions because the manganese oxide which oxidizes hydrogen sulfide to sulfur could not be regenerated under anaerobic conditions. In contrast, the rate constant for hydrogen sulfide under aerobic conditions was high due to the multiplier effect attributed to both increasing Eh and the regeneration of manganese oxide by dissolved oxygen.

1. Introduction

Annual industrial emission of fly ash by coal burning is approximately 750 million tons [1]. In 2011, coal-fired generation accounted for 29.9% of the world's electricity supply, and it is estimated that this will increase to 46% by 2030 [1]. Coal ash has been recycled for various uses—as a key ingredient in concrete and road base construction, as a soil amendment, for zeolite synthesis, as a raw material for extracting rare earth elements, and as an absorbent [1–5] and its utilization percentage is approximately 50–70% [1]. Therefore, the present utilization rate is insufficient for complete recycling of the increasing amounts of coal ash being produced. It is hoped that new and expanded applications for coal fly ash will contribute to further waste reduction and set the standard for other industrial-scale recycling.

In the past few years we have developed a recycled material for environmental remediation of coastal oceans called granulated coal ash, GCA [6]. GCA is produced through the granulation of coal fly ash from coal thermal electric power stations (Chugoku Electric Power) with added blast furnace cement as binder amounting to 13–15% of the final product. When coal fly ash is mixed with blast furnace cement, the

Pozzolanic reaction [7] occurs, namely, the silicon oxide and aluminum oxide contained in coal fly ash react with the calcium hydroxide in blast furnace cement, which increases the specific surface area and compression strength of the GCA. A previous study found that GCA can remove hydrogen sulfide from the liquid phase with oxidation by the manganese oxide present on the GCA [6].

GCA was used to remove hydrogen sulfide from eutrophic coastal marine sediments, as it has a harmful impact on aquatic ecosystems in the lower range of $2.93\text{--}59 \mu\text{M}$ [8]. Hydrogen sulfide is generated by sulfate-reducing bacteria under the anoxic conditions created by the consumption of dissolved oxygen (DO) by oxidative decomposition of the sediments. High concentrations of hydrogen sulfide are especially detected in marine sediments in enclosed or semi-enclosed water bodies located adjacent to large metropolitan areas [9–15].

GCA was scattered in the water or mixed with organically enriched sediment along the seacoast and was found to suppress hydrogen sulfide effectively [14,16]; moreover, the applied GCA in actual field sites need not be collected because the adsorption site on the GCA for hydrogen sulfide is regenerated through manganese oxidation under oxic conditions such as occur during the vertical mixing seasons [14].

* Corresponding author.

E-mail address: s-asaoka@maritime.kobe-u.ac.jp (S. Asaoka).

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In a previous study, batch experiments for hydrogen sulfide removal were conducted under conditions of dissolved oxygen (DO) less than 2 mg L^{-1} [17]. However, the DO at the sea bottom, which has significant organic matter loads, ranges from aerobic to anaerobic conditions. For example, DO ranges of $0.1\text{--}19.8 \text{ mg L}^{-1}$ at Dokai Bay, Japan [18], $3.78\text{--}9.11 \text{ mg L}^{-1}$ at Jinhae Bay, Korea [19], $0.14\text{--}4.4 < \text{ mg L}^{-1}$ at Helena Bay, South Africa [20] and $< 2.8\text{--}12 \text{ mg L}^{-1}$ at Tokyo Bay, Japan [21] have been reported. Hence, it is necessary to evaluate the removal efficiency of hydrogen sulfide by GCA under different DO concentrations to control hydrogen sulfide in coastal marine sediments. The purpose of this study was to evaluate the removal rate of hydrogen sulfide by GCA under aerobic and anaerobic conditions.

2. Experimental

2.1. The Granulated Coal Ash used in this study

Coal which is the origin of fly ash was mainly imported from Australia and Republic Indonesia. GCA tested in this study was produced through the granulation process of pulverized fly ash from Shinonoda thermal electric power stations (Chugoku Electric Power) with added blast furnace cement (B class: Nippon Steel & Sumikin Blast Furnace Slag Cement) as binder amounting to 13% of the final product. GCA granules with 5-mm diameters are mainly composed of silicon oxide, carbonate, aluminum oxide, calcium oxide, organic carbon and iron oxide with a quartz and aluminosilicate crystal phase. The chemical composition of the manufactured GCA, by weight, was 39.5% silicon, 13.3% carbonate, 12.6% Al_2O_3 , 5.5% CaO, 2.3% Fe_2O_3 , 0.8% MgO, 0.6% K_2O , 0.6% TiO_2 , 0.3% Na_2O , 0.2% P_2O_5 , and other trace substances Table 1 [6]. The environmentally regulated substances dissolved from the GCA used in this study were well below the standard levels for environmental criteria in Japan [22].

2.2. Hydrogen sulfide removal experiments

The conditions of the removal experiments for hydrogen sulfide with different DO levels and hydrogen concentrations are shown in Table 2. The experiments at $\text{DO} < 0.2 \text{ mg L}^{-1}$ and 8 mg L^{-1} represent anaerobic and aerobic conditions, respectively. DO of 4 mg L^{-1} represents an intermediate condition.

2.2.1. Anaerobic and intermediate conditions

The hydrogen sulfide solution was prepared as follows: An aliquot of 1 mol L^{-1} of Tris-HCl buffer (Kanto Kagaku) was diluted by pure water to be 30 mmol L^{-1} . The 30 mmol L^{-1} of Tris-HCl buffer was de-aerated with N_2 gas to a final DO of $< 0.2 \text{ mg L}^{-1}$ or 4 mg L^{-1} . The following procedure was conducted in a glove box which was substituted by N_2 gas. An aliquot of $\text{Na}_2\text{S}\cdot 9\text{H}_2\text{O}$ (Wako Pure Chemical Industries) was dissolved into the 30 mmol L^{-1} of Tris-HCl buffer solution to represent the possible range in the pore water of organically enriched sediments. The pH of the solution was adjusted to 8.2, which is the

Table 1

Trace substances composition of the GCA (mg kg^{-1} dw).

Ba	397	Ni	29.2
Zr	298	Rb	28.8
Mn	255	Co	28.6
N	200	Cr	27.2
V	111	Ga	20.6
Zn	88.9	Sc	14.5
Ce	69.7	Th	12.7
Cu	58.9	Hf	6.7
Y	52.6	W	5.2
Nd	34.4	U	4.2
La	34.2	Yb	3.9
Pb	29.3	Cs	3.2

Table 2

Conditions of removal experiment for hydrogen sulfide.

H_2S Concentration (mg-S L^{-1})	DO concentration (mg L^{-1})
2	< 0.2
10	< 0.2
	4
	approx. 8
130	< 0.2

typical pH of seawater, by adding HCl or NaOH as necessary. Fifty mL of the prepared hydrogen sulfide solution was slowly dispensed into a 100-mL vial bottle, and 0.2 g of the GCA was added to the solution. Thereafter, the bottle was plugged with a rubber cork and sealed with an aluminum cap. The bottle was taken from the glove box and vacuum packed. The bottle was agitated moderately at 100 rpm at 25°C in a constant-temperature oven. The batch experiments were conducted in triplicate and the bottles were individually prepared for each sampling time. Time courses of hydrogen sulfide concentration were measured using a detection tube (200SA or 200SB: Komyo Rikagaku Kougyo). Hydrogen sulfide solution was also prepared without the addition of GCA as a control, and experiments were conducted by the same protocol.

2.2.2. The aerobic condition

An aliquot of $\text{Na}_2\text{S}\cdot 9\text{H}_2\text{O}$ (Wako Pure Chemical Industries) was dissolved into 30 mmol L^{-1} of Tris-HCl buffer solution, which was not de-aerated with N_2 gas, and the DO was approximately 8 mg L^{-1} , to prepare 10 mg-S L^{-1} of hydrogen sulfide concentration. The pH of the solution was adjusted to 8.2, which is the general pH of seawater, by adding HCl or NaOH as necessary. Fifty mL of the prepared hydrogen sulfide solution was slowly dispensed into a 100-mL vial bottle, and 0.2 g of GCA was added to the solution. Thereafter, the bottle was plugged with a rubber cork, sealed with an aluminum cap and vacuum packed. The bottle was agitated moderately at 100 rpm at 25°C in a constant-temperature oven. The bottles for this batch experiment were individually prepared for each sampling time. The time courses of hydrogen sulfide concentration were measured using a detection tube (200SA or 200SB: Komyo Rikagaku Kougyo). The hydrogen sulfide solution was also prepared without the addition of GCA as a control, and experiments were conducted by the same protocol.

2.3. Other parameters

The Eh (Electric potential of the normal hydrogen electrode) and pH of initial conditions were measured by an Eh meter (HM-31P: DKK-TOA) and a pH meter (HM-32P: DKK-TOA), respectively.

An Eh-pH diagram of sulfur was drawn using the geochemical modeling software, Geochemist's Workbench 8.0 (RockWare). The parameters used in this thermodynamic calculation were as follows: the activities referred to the liquid-phase concentration of hydrogen sulfide. The pressure and temperature were set at 1.013 hPa and 25°C , respectively. The Eh-pH diagram of sulfur was almost the same in the hydrogen sulfide range of $2\text{--}130 \text{ mg-S L}^{-1}$.

Multiple-comparison analyses were carried out to detect significant differences in the obtained data by Fisher's least significant difference method using the statistical software Excel-Toukei (BellCurve).

3. Results and discussion

3.1. Removal kinetics of hydrogen sulfide by GCA under anaerobic conditions

In the case of hydrogen sulfide initial concentration of 2 mg L^{-1} under $\text{DO} < 0.2 \text{ mg L}^{-1}$, the H_2S concentration of control without GCA decreased to 1.3 mg-S L^{-1} after 24 h due to oxidation (Fig. 1). Because

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