



Sulfate and organic matter concentration in relation to hydrogen sulfide generation at inert solid waste landfill site – Limit value for gypsum



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ABSTRACT

In order to suggest a limit value for gypsum (CaSO_4) for the suppression of hydrogen sulfide (H_2S) generation at an inert solid waste landfill site, the relationship between raw material (SO_4 and organic matter) for H_2S generation and generated H_2S concentration, and the balance of raw material (SO_4) and product (H_2S) considering generation and outflow were investigated. SO_4 concentration should be less than approximately 100 $\text{mg-SO}_4/\text{L}$ in order to suppress H_2S generation to below 2000 ppm. Total organic carbon (TOC) concentration should be less than approximately 200 mg-C/L assuming a high SO_4 concentration. The limit value for SO_4 in the ground is 60 $\text{mg-SO}_4/\text{kg}$ with 0.011 wt% as gypsum dihydrate, i.e., approximately 1/10 of the limit value in inert waste as defined by the EU Council Decision (560 $\text{mg-SO}_4/\text{kg-waste}$). The limit value for SO_4 in inert waste as defined by the EU Council Decision is high and TOC is strictly excluded. The cumulative amount of SO_4 outflow through the liquid phase is much larger than that through the gas phase. SO_4 concentration in pore water decreases with time, reaching half the initial concentration around day 100. SO_4 reduction by rainfall can be expected in the long term.

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1. Introduction

Hydrogen sulfide (H_2S) gas at 20–30 ppm produces a strong offensive odor likened to rotten egg (Reiffenstein et al., 1992), and its inhalation at 750–1000 ppm leads to abrupt physical collapse (Milby and Baselt, 1999) (Hereafter, H_2S concentration exceeding 1000 ppm is called high concentration.). H_2S is generated at landfill sites globally (Lee et al., 2006; Eun et al., 2007; Xu et al., 2010; Fang et al., 2012; López and Lobo, 2014; Yue et al., 2014). In 1999, three workers at an inert solid waste landfill site in Japan died after exposure to H_2S at 15,000 ppm. In Japan, as inert solid waste landfill sites accept waste plastic, rubber, metal, glass, ceramic, and construction and demolition debris (stones, bricks, and blocks), it is not necessary to install gas bent pipes, liners for leachate, or water treatment facilities. Around the time of the accident, waste gypsum board without protective paper sheet was also accepted. H_2S gas is produced at landfills when sulfate-reducing bacteria (SRB) consume and convert sulfate (SO_4) under anaerobic conditions (Lee et al., 2006; Townsend et al., 2004, 2005). The source of SO_4 in construction and demolition debris is predominantly gypsum (CaSO_4) (Jang and

Townsend, 2001). SRB also consume organic matter. At inert solid waste landfill sites in Japan, not biodegradable organic matter but waste gypsum board from which protective paper sheet had been removed had been accepted. However, because organic matter in the base material of waste gypsum board (gypsum part) could be a source of H_2S (Inoue, 2005), the Ministry of the Environment in Japan prohibited the disposal of waste gypsum board at inert solid waste landfill sites in 2006 (Notice: Treatment of waste gypsum board from which protective paper sheet had been removed). Measures to prevent H_2S generation or diffusion at landfill sites include suppressing the creation of anaerobic conditions (by draining water and/or introducing oxygen) (Masamoto et al., 2012) and trapping generated H_2S (by conversion into iron sulfide using iron-containing material (Naruoka and Ono, 2004; Bergersen and Haarstad, 2014) and adsorption by activated carbon (Adib et al., 1999) or zeolite (Karge and Raskó, 1978)). The Ministry of the Environment in Japan recommended the appropriate installation of cover soil (to prevent rainwater percolation and trap H_2S by iron contained in cover soil) and gas bent pipes (to supply oxygen). If waste gypsum board was properly presorted at construction and/or demolition sites, it would be possible to dispose gypsum-free construction and demolition waste (CDW) at inert solid waste landfill sites. However, in regard to CDW contaminated with waste gypsum board due to poor presorting at the site, the

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disposal at inert solid waste landfill sites would be allowed only after the removal of waste gypsum board by sorting at an intermediate waste treatment facility. Although waste gypsum board removal may be difficult because gypsum board is fragile and easily disintegrates into fine particles, no directive, such as a limit value for gypsum, has been enforced in Japan. In order to suppress H_2S generation at inert solid waste landfill sites, a directive for sorting at construction, demolition and/or sorting sites, i.e., a limit value, should be set. Ono and Tanaka (2003) investigated the relationship between eluted substances and generated H_2S by immersing ten actual waste samples (CDWs and waste gypsum board) in water, and suggested that total organic carbon (TOC) concentration should be below 30 mg-C/L to suppress H_2S generation at a high concentration. However, as only ten samples were used, the relationships of SO_4 concentration with generated H_2S and H_2S generation potential when both SO_4 and TOC concentrations were varied widely were unknown.

In EU, pursuant to Council Directive (EC, 1999), landfills were classified into three types: landfill for hazardous waste, that for non-hazardous waste, and that for inert waste, according to the environmental pollution risk posed by the accepted waste. The EU Council Decision (EC, 2002) states: “2.2.3. Gypsum waste; Non-hazardous gypsum-based materials should be disposed of only in landfills for non-hazardous waste in cells where no biodegradable waste is accepted.” Therefore, no waste gypsum board is accepted at the landfills for inert waste. As only selected CDWs, such as concrete, brick, tile, ceramic, and their mixtures, are accepted at the landfills for inert waste, waste gypsum board should be removed. Furthermore, the limit values for SO_4 and dissolved organic carbon are applied to waste acceptable at the landfills. However, the H_2S generation potential of waste having SO_4 at the limit value is unknown.

At first, the SO_4 concentration that generates H_2S at a high concentration should be investigated experimentally. However, if SO_4 outflow by diffusion or rainfall was rapid, the limit value for SO_4 would be high. In this regard, not only SO_4 concentration but also SO_4 balance is important.

In this study, in order to suggest the limit value for gypsum ($CaSO_4$) for the suppression of H_2S generation at inert solid waste landfill sites in Japan and other countries, the following two points were investigated:

- Relationship between raw material (SO_4 and organic matter) for H_2S generation and generated H_2S concentration.
- Balance of raw material (SO_4) and product (H_2S) considering generation and outflow.

2. Materials and methods

SRB, which are mesophilic bacteria, were cultured with various concentrations of raw material (SO_4 and organic matter) for H_2S generation at 36 °C in an incubator, and the concentration of generated H_2S was measured. Specifically, first, the required time for H_2S generation was investigated in order to determine the incubation period. Then, after preparing various concentrations of raw material, the concentration of generated H_2S was measured at the required time.

2.1. Materials

As basic experiments were conducted in this study, the actual sample, i.e., leachate from landfill site, was not used. Calcium sulfate (gypsum dihydrate ($CaSO_4 \cdot 2H_2O$), Japanese Food Additives, Wako Pure Chemical Industries) was used as SO_4 . Dried yeast (Dried Yeast Extract D-3, Wako Pure Chemical Industries) and lactose (Lactose Broth DAIGO, Wako Pure Chemical Industries) were

equivalently mixed and used as organic matter. As nutrient for microorganisms, BOD Nutrient Buffer (Pillows for 6 L sample, APHA Formulation, Hach Co.) was used. One pillow was mixed with pure water to make 40 mL (high-concentration nutrient). Silica sand (Toyoura Keiseiki Kogyo) was used as microorganism carrier and black sediment in an ornamental pond was used for seeding. Five grams of black sediment and 1000 mL of pure water were mixed and shaken for five minutes, and nitrogen gas (N_2) was bubbled for five minutes. The supernatant was used as seeding liquid.

The required amounts of SO_4 and organic matter were set based on the experimental conditions. Two milliliters of high-concentration nutrient and 25 mL of seeding liquid were introduced into a plastic bottle and pure water was added to make 500 mL. The mixture was shaken for five minutes and N_2 gas was bubbled for five minutes, and this is called H_2S generation solution hereafter.

2.2. Experimental methods

2.2.1. H_2S generation system

Two spoonful of silica sand, 80 mL of H_2S generation solution, and 10 mL of N_2 gas were introduced into a 100 mL plastic syringe. A silicone tube with a silicone inlet plug for gas chromatography was attached to the tip of the plastic syringe, and this is called

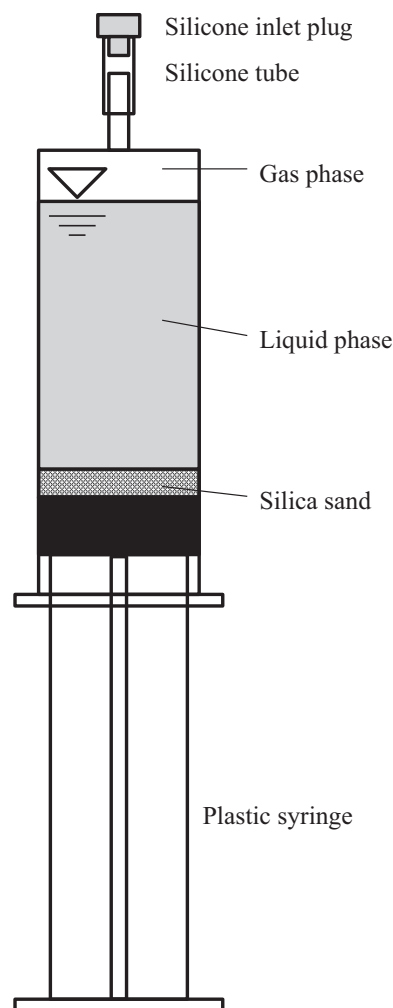


Fig. 1. Schematic diagram of H_2S generation system.

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