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Hydrogen sulphide removal from landfill gas

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ABSTRACT

Control of odours should be considered to be a fundamental issue in order to site, design and manage sanitary landfills. With regard to construction and demolition (C&D) debris, landfilling was the mainly adopted solution in many European Countries; in particular, gypsum drywalls can produce high concentrations of hydrogen sulphide (H_2S) in landfill gas ranging from 7 ppm to 100 ppm. In some cases also dangerous concentrations until to 12,000 ppm were detected. In this paper H_2S removal efficiency in a lab-scale vertical packed scrubber was investigated. Hydrogen sulphide abatement was evaluated for inlet H_2S concentrations of 1000–100–10 ppm, adjusting scrubbing liquid pH in the range 9–12.5 by means of caustic soda (NaOH 2N solution). Moreover, best operating conditions for the system were defined as well as H_2S abatement along the tower and liquid recirculation effectiveness in case of inlet H_2S concentration of 10 ppm (typical odour concentration). Results showed that pH of 11.5 in scrubbing liquid could be considered the best value for removal of different inlet H_2S concentrations, also taking into account parasitical consumption of NaOH due to CO_2 absorption. Moreover, in case of continuous working of the system at H_2S concentration of 10 ppm, strong removal efficiency was already obtained with a packed bed height of about 70 cm. Significant performances were ensured after 1 h of constant activity, consuming about 3 ml of soda per cubic meter of polluted air. Subsequently liquid blowdown was necessary.

Keywords: Hydrogen sulphide; Odours; Construction and demolition wastes; Packed scrubber; pH adjustment

1. Introduction

In several European Countries landfilling is the mainly adopted solution for management of Construction and Demolition (C&D) wastes. Recycling is poorly developed and market of recycled aggregates coming from C&D wastes belongs principally to Germany (25.7%), UK (31.3%) and Netherlands (11.3%) (Bressi, 2002; UEPG, 2006; Mulder et al., 2007).

With regard to Italy, recycling of construction and demolition wastes is not developed yet. Analysis of ANPAR (2006), Italian association of recycled aggregates producers, revealed that C&D wastes production in 2004 was of about 40 million of tons but only an amount of 10% was recycled; as a result, the most quantity of inert wastes was landfilled.

USEPA Report(1998a) showed that gypsum drywall is present in C&D wastes in the weight of 21–27% and it is composed by 90% gypsum and 10% paper facing and backing. Unfavourable economics and poor market restrict gypsum recycling development (Jang and Townsend, 2001). As a result, considerable quantities of C&D wastes with gypsum can be found in landfills.

Particularly, disposal of gypsum drywalls could produce serious environmental impacts. Gypsum is a mineral composed by calcium sulphate ($CaSO_4$) and water (H_2O). It is a good fire barrier and it is used in buildings for interior walls. Under particular moisture conditions, part of gypsum sulphate dissolves in water in high concentrations: a contamination of groundwater due to leachate could occur (Jang and Townsend, 2001). Moreover, high concentrations of hydrogen sulphide (H_2S) could be recorded in C&D debris landfills. Under wet and anaerobic conditions sulphate reducing bacteria (SRB) use sulphate ($SO_4{}^{2-}$) as an oxygen source in order to assimilate the organic matter, producing sulphide (S^{2-}) as a by-product. Then, a dynamic equilibrium between four forms of sulphide as (S^{2-}), (HS^-), (H_2S_{aq}) and (H_2S_g) takes place immediately and equilibrium among these species depends on pH. For instance,

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only HS^- and S^{2-} exist in the solution for pH equal to 9, thus practically no sulphide specie can leave the aqueous phase like a free gas (USEPA, 1998b; Gostelow et al., 2001; Jang and Townsend, 2001; Stuetz and Frechen, 2001; Jefferson et al., 2002; Reinhart and Townsend, 2002; Mustafa, 2003; Takeshita et al., 2003; Bogner and Heguy, 2004; Lee et al., 2006).

H_2S is characterized by the classical rotten eggs smell and it can be detected by human in concentrations as low as 0.21 ppb on volume basis. Concentrations until 30 ppm result in a strong odour; higher concentrations affect the nervous and respiratory system of human. Gypsum drywalls could produce considerable concentrations of hydrogen sulphide gas ranging from 7 ppm to 100 ppm (Bogner and Heguy, 2004). In some cases more dangerous concentrations (5000-12,000 ppm) were also detected (Reinhart and Townsend, 2002; Haarstad et al., 2003; Lee et al., 2006). Council Decision 2003/33/EC prohibits gypsum disposal where organic matter is present, so to prevent H_2S production and impact. Nevertheless, some studies showed that landfilling of only gypsum drywall produces significant concentrations of hydrogen sulphide anyway, because drywall paper is enough organic matter for growth of bacteria. Moreover, concrete aids gas migration (Reinhart and Townsend, 2002).

Owing to H_2S dispersion, several C&D debris landfills were closed in many developed countries. Some facilities operators were required to install gas collection and recovery systems; others, on the contrary, resorted to odours masking agents around their sites (Townsend, 2003). While various experiments were carried out in order to improve C&D wastes management and reduce landfill dumping (Poon et al., 2001; John et al., 2004; Mulder et al., 2007), two burning problems remain to be faced with regard to odours problems:

- management and monitoring of old C&D debris landfills;
- correct design of gas collection, treatment and recovery systems in new plants anyway.

Five odours control methodologies were identified by the United States for National Resource Council (Otieno and Magagula, 2001):

- modification of the process;
- dilution in atmosphere;
- dissolution of odour in a liquid (wet scrubbing in a mist or packed tower);
- oxidation of odour with air;
- modification of odour perception.

Burgess et al. (2001) reported the following classification for odours treatment systems:

- biochemical systems: biofilters, bioscrubbers, activated sludge;
- chemical systems: chemical scrubbers, thermal oxidation, catalytic oxidation, ozonation;
- physical system: condensation, adsorption (activated carbon), absorption (clean waters scrubber).

Zappa (2001) showed that scrubbing and thermal oxidation are the most effective solutions in order to removal H_2S at different concentrations ranges (1–10 ppm; 11–50 ppm; >50 ppm). An economic comparison between those two technologies was presented by Calcaterra and Confalonieri (2002): total costs, covering capital and operating costs, are clearly higher for thermal oxidation than scrubbing system.

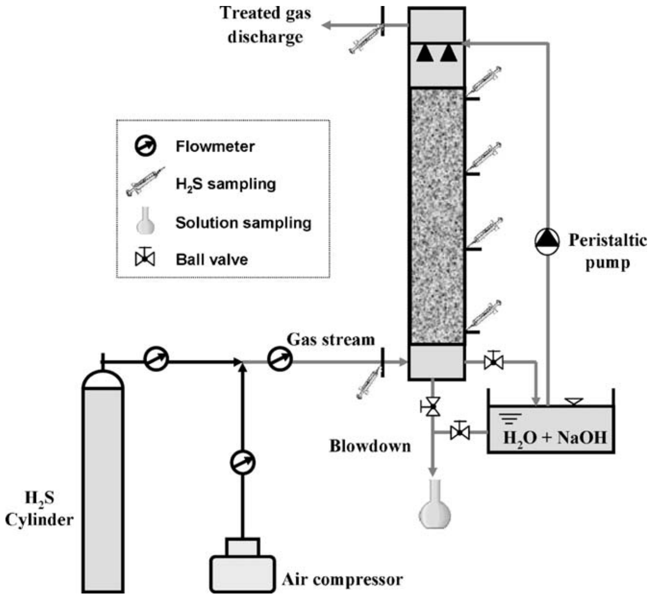

Fig. 1 – Lab-scale plant for H_2S abatement.

Aim of the paper was to assess H_2S removal in a vertical packed scrubber by means of pH adjustment in the scrubbing liquid using caustic soda. Best operating conditions and liquid recirculation effectiveness were assessed. Besides, H_2S abatement along the tower was evaluated.

2. Materials and methods

2.1. *Experimental apparatus*

The lab-scale plant for experimental activity is shown in Fig. 1.

The principle of H_2S removal was that compound in foul gas was absorbed by the countercurrent scrubbing liquid during a short contact time (1–2 s) and transferred from gas phase to water one. The absorption phenomenon was advanced by high pH values.

Hydrogen sulphide was supplied from a gas cylinder (0.5% purity) and it was diluted at different concentrations (1000–100–10 ppm) by means of air from a compressor. The flow rates of H_2S and air were controlled by flow meters. However flow rate of inlet gas was equal to 25 l/min. The reactor was a column made of PVC. The dimensions were 130 cm in height and 10 cm cylinder internal diameter. The effective packing height was 100 cm. The scrubber bed was composed of plastic Raschig rings (8 mm × 8 mm) and a sieve plate of stainless steel was placed at the bottom of the tower in order to support packing materials and allow an uniform and homogeneous distribution of gas. An inlet liquid (NaOH 2N solution) was introduced at the top of scrubber by means of a peristaltic pump at 30 cl/min. In case of continuous operation, scrubbing liquid was recirculated at 30 cl/min again, until to necessary discharge of blowdown on the grounds of pH value and H_2S removal efficiency of the system. Six H_2S sampling ports were located in the column, two at the inlet and outlet points and four along the tower respectively.

2.2. *Analytical methods*

All the hydrogen sulphide concentrations were measured by gas detection tubes (Gastec Co.). The measure ranges of H_2S gas tubes were 0.1–4% and 10–4000 ppm, 1–240 ppm, 1–40 ppm.