Research Article

Removal of Hydrogen Sulfide by Physico-Biological Filter Using Mixed Rice Husk Silica and Dried Activated Sludge

This study investigated the effectiveness of a new packing material, namely mixed rice husk silica with dried activated sludge for removing H₂S. Dried sewage sludge was collected from Putrajaya sewage treatment plant in Malaysia. Rice husk silica was prepared at temperature of 800°C, after acid leaching and mixed with dried sewage sludge to be utilized in a polyvinyl chloride filter. The system was operated under variable conditions of two parameters, different inlet gas concentration and different inlet flow rate. H₂S was passed through the filter with one liter of the packing material. More than 99.96% removal efficiency (RE) with empty bed residence time (EBRT) of 90–45 s and 300 ppm inlet concentration was observed. However, the RE decreased to 96.87% with the EBRT of 30 s. The maximum elimination capacity (EC) of 52.32 g/m³/h was obtained with the RE of 96.87% and H₂S mass loading rate of 54 g/m³/h, while at the RE of 99.96%, maximum EC was 26.99 g/m³/h with the H₂S mass-loading rate of 27 g/m³/h. A strong significant correlation between increasing of H₂S mass loading rate and pressure drop was also detected (p < 0.01). Maximum pressure drop was 3.0 mm H₂O after 53 days of operating time, the EBRT of 30 s, and 54 g/m³/h of H₂S loading rate. These observations suggest that the mixture of rice husk silica with dried activated sludge is a suitable physico-biological filter for H₂S removal.

Keywords: Air pollution; Elimination capacity; Empty bed residence time; Packing material; Removal efficiency

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1 Introduction

Industrial growth has lead to higher energy consumption and emis-
sion of different kind of pollutants into the air [1, 2]. It is important
to note that the malodor caused by hydrogen sulfide (H₂S) has been
an environmental and public nuisance in the recent years. There are
common places that produce and generate a variety of offensive
odors such as pulping sites, petroleum refinery plants, drug manu-
facturing processes, sewage treatment facilities, and livestock farms.
The majority of these odors are attributable to the production and
dissemination of sulfur compounds [3, 4]. In recent years, the influ-
ence of even low concentrations of air pollutants on human health
has re-emerged as an important scientific issue. Several studies have
found that various acute and chronic health impacts to air pollution
[5]. In order to solve environmental problems more successfully,
researchers need to focus on design and optimization of the treat-
ment processes for environmental pollution [6].

The biological treatment system becomes more and more popular
because it is a green technology, which does not use chemicals
and does not produce wastes potentially dangerous for the environ-
ment. This process is fundamentally based on the ability of micro-
organisms to transform organic and inorganic pollutants into less
toxic and odorless compounds [7]. Two mechanisms, namely absorp-
tion/adsorption and bio-oxidation, allow the biofilters to remove
odoriferous compounds. First, odoriferous gases pass through a biofilter,
and are absorbed into the moist surface layer of the biofilter media
particles and/or are adsorbed onto the surface of the particles.
Biodegradable components of the odoriferous gases are then metab-
olized by microbial floras [8]. There are mixtures of microorganisms
in activated sludge as Allen [9] reported there are probably several
hundred thousand million bacteria per gram of dry matter in acti-
ated sludge. Aerobic sulfur oxidizing bacteria can transform hydro-
gen sulfide to elemental sulfur and sulfate (SO₄²⁻) [10]. Table 1 shows
some studies for removal of malodoriferous gases by biofiltration
system.

The main goal of this study was to investigate the removal of
hydrogen sulfide in a physico-biological filter packed with a new
packing material, namely mixed rice husk silica, and dried activated
sludge. Furthermore, elimination capacity (EC), pressure drop and
pH changes versus operating time, different empty bed resi-
dence time (EBRT) and different inlet concentration of H₂S was
investigated. This study has been carried out in the laboratory of
Environmental Studies at University Putra Malaysia (UPM) in 2010.
2 Materials and methods

2.1 Preparation of packing material

Mixed rice husk silica with dried activated sludge was prepared as packing material in a physico-biological filter for removal of hydrogen sulfide. Rice husk silica was prepared according to Jamwal and Mantri’s method [19], available at http://www.nandinichemical.com/2007febjournal.html. The rice husk was first washed with tap water to remove the dirt and other contaminants occurred in them. Then, the rice husk was dried in an oven at 110°C for about 24 h; and subjected to acid leaching by reflux in 3% v/v hydrochloridric acid (HCL) and 10% v/v sulfuric acid (H2SO4) for 2 h, at a ratio of 50 g husk/L. The husk was then thoroughly washed with distilled water and dried in oven at 100°C for 4 h. Finally, the cleaned husk was burned inside a muffle furnace at 800°C. Incineration was done in a porcelain crucible for 4 h in static air. Dried activated sludge was collected from a sewage treatment plant (Putrajaya STP 2) in Malaysia and mixed with rice husk silica (50:50 v/v) to be used as physico-biological filter.

Chemical analysis including analysis of elements (CHNS) and chemical composition was carried out for rice husk silica and dried activated sludge. Moreover, Brunauer–Emmett–Teller (BET) specific surface area was performed by using a ThermoFinnigan Sorptomatic apparatus using nitrogen adsorption at –196°C for rice husk silica [20].

2.2 Experimental set-up and operation

The physic-biological filter was constructed using PVC cylinder with 50 cm in height (packed height 22 cm) and 7.5 cm in diameter. The schematic diagram of the pilot is shown in Fig. 1. The Malodorous gas, H2S was passed through the packing material as down flow from a H2S gas cylinder with 4000 ppm and 150 bars inside pressure. An air pump was used to prepare oxygen for aerobic bacteria in the filter and to produce different concentration of H2S in the mixing chamber.

Temperature and humidity of the filter’s packing material were controlled at 27 ± 5°C and 60 ± 5%, respectively using a humidifier tower. The experiment was carried out in two stages: in the first stage, system was operated at a constant EBRT of 60 s and different concentration of inlet H2S.

Jeong et al. [3] compared two kinds of packing materials, namely polypropylene fibrils and volcanic stone in biofilter system for the biological removal of hydrogen sulfide. They used low concentration of H2S during adaptation period. After this period, H2S concentration was increased in weekly increment. In this study, while microorganisms adapted, the tested H2S was introduced in the filter at low concentration (10 ppm) for 1 wk. After the adaptation period, H2S concentration at the inlet was increased in weekly increments, from 10 ppm to a final concentration of 300 ppm (10, 50, 100, and 300 ppm). In the second stage, inlet H2S concentration was fixed (300 ppm) and air flow rate was variable based on different EBRT of 30–90 s (EBRTs 90, 75, 60, 45, and 30 s was used).

2.3 Data analysis

Inlet and outlet hydrogen sulfide concentrations were measured using H2S detector model ppb RAE 3000, USA. Removal efficiency (RE), EC, and pressure drop (mm H2O) were calculated as follow:

\[
RE(\%) = \frac{C_{Gi} - C_{Go}}{C_{Gi}} \times 100
\]

\[
EC(g/m^3/h) = \frac{(C_{Gi} - C_{Go}) \times Q}{V_f}
\]

in which \(Q\) is the gas flow rate (m^3/h), \(V_f\) is the volume of the filter bed (m^3), \(C_{Gi}\) and \(C_{Go}\) are the inlet and outlet hydrogen sulfide concentration (ppm; g/m^3) [21–24].

3 Results and discussion

In this study, rice husk silica combined with dried activated sludge was used as physico-biological filter for removal of hydrogen sulfide.

Table 1. Some studies about gas pollutant removal by biofiltration in literatures

<table>
<thead>
<tr>
<th>Filter’s material</th>
<th>Height (cm)</th>
<th>Inner diameter (cm)</th>
<th>Packing material</th>
<th>Pollutants</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>PVC</td>
<td>100</td>
<td>10</td>
<td>Compost</td>
<td>H2S</td>
<td>[11]</td>
</tr>
<tr>
<td>PVC</td>
<td>80</td>
<td>10</td>
<td>Compost and Coconut fiber</td>
<td>NH3</td>
<td>[12]</td>
</tr>
<tr>
<td>Glass</td>
<td>140</td>
<td>10</td>
<td>Granulated sludge: BGSn and BGNs</td>
<td>NH3 and H2S</td>
<td>[13]</td>
</tr>
<tr>
<td>Acrylic tube</td>
<td>60</td>
<td>5.5</td>
<td>Peat</td>
<td>H2S</td>
<td>[14]</td>
</tr>
<tr>
<td>PVC</td>
<td>100</td>
<td>10</td>
<td>SLIR</td>
<td>H2S</td>
<td>[15]</td>
</tr>
<tr>
<td>Metal</td>
<td>91</td>
<td>5.6</td>
<td>Activated sludge</td>
<td>Odor and NH3</td>
<td>[16]</td>
</tr>
<tr>
<td>Glass</td>
<td>25</td>
<td>6</td>
<td>Cell-laden ca-alginate</td>
<td>H2S and NH3</td>
<td>[17]</td>
</tr>
<tr>
<td>Glass</td>
<td>10</td>
<td>8</td>
<td>Vibrio alginolyti-cus</td>
<td>NH3</td>
<td>[18]</td>
</tr>
</tbody>
</table>

PVC: polystyrene chloride; BGSn, column packed with granulated sludge and mainly supplied with hydrogen sulfide; BGNs, column packed with granulated sludge and mainly supplied with ammonia; SLIR, specialized engineering in recycling agricultural residues, its commercial name is ABONLIR.
As Papirer [25] described silica is one of the most important adsorbents. Some of the important characteristics of the rice husk silica and dried activated sludge are listed in Tables 2 and 3, respectively.

The study was carried out in two stages: (I) operation with different inlet concentration of hydrogen sulfide and (II) operation with different inlet flow rate.

### 3.1 Filter’s performance with different inlet concentration of H$_2$S

In the first stage, performance of the physico-biological filter was evaluated during the start up period with the EBRT of 60 s and different hydrogen sulfide inlet concentrations of 10–300 ppm.

System was operated with low inlet H$_2$S concentration of 10 ppm. After 5 days (adaptation period), the RE of physico-biological filter reached to the maximum amount of 100%. As shown in Fig. 2, after adaptation period, the RE was greater than 99.26% with the inlet H$_2$S concentration of 10–300 ppm.

In this stage, the maximum EC of 26.87 g/m$^3$/h with the inlet hydrogen sulfide concentration of 27 g/m$^3$/h was obtained (Fig. 3).

Maximum pressure drop reached 1.0 mm H$_2$O after 28 days of operating time and 300 ppm inlet concentration of hydrogen sulfide. The relation between different operating times (day) with different inlet concentrations of H$_2$S on pressure drop is shown in Fig. 4.

The system was started with pH of 6.8 and decreased to 3.8 after 28 days of operating time and 300 ppm inlet concentration of hydrogen sulfide. The result of changes in pH versus operating time and different inlet concentration of H$_2$S with fixed EBRT of 60 s is shown in the Fig. 5.

### 3.2 Filter’s performance with different flow rates (second stage)

In the second stage, the inlet H$_2$S concentration was fixed at 300 ppm, while flow rate was variable with the different EBRT of 30–90 s (EBRTs of 90, 75, 60, 45, and 30 s). In this stage, the physico-biological filter showed $>99.96\%$ RE with the EBRT of 90–45 s. However, the RE decreased to 96.87% with the EBRT of 30 s and system was unstable. The result of RE in the second stage is shown

### Table 2. Some of the physical and chemical characteristics of rice husk silica used in this study

<table>
<thead>
<tr>
<th>Properties</th>
<th>Unit</th>
<th>Value</th>
<th>Properties</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chemical properties</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SO$_3$ (%)</td>
<td>(%)</td>
<td>1.66</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Analysis of elements</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C (%)</td>
<td>(%)</td>
<td>0.05 ± 0.01</td>
<td>Physical properties</td>
<td></td>
<td></td>
</tr>
<tr>
<td>H (%)</td>
<td>(%)</td>
<td>0.27 ± 0.01</td>
<td>Density</td>
<td>(g/L)</td>
<td>52</td>
</tr>
<tr>
<td>N (%)</td>
<td>(%)</td>
<td>0.36 ± 0.05</td>
<td>Surface area</td>
<td>(m$^2$/g)</td>
<td>226.3</td>
</tr>
<tr>
<td>S (%)</td>
<td>(%)</td>
<td>0.03 ± 0.01</td>
<td>Median pore radius</td>
<td>(nm)</td>
<td>2.3747</td>
</tr>
<tr>
<td>Chemical composition</td>
<td></td>
<td></td>
<td>Cumulative pore volume</td>
<td>(cm$^2$/g)</td>
<td>0.3078</td>
</tr>
<tr>
<td>SiO$_2$ (%)</td>
<td>(%)</td>
<td>97.35</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### Table 3. Some important properties of dried activated sludge used as packing material

<table>
<thead>
<tr>
<th>Properties</th>
<th>Unit</th>
<th>Value</th>
<th>Properties</th>
<th>Unit</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particles size (Wt %)</td>
<td></td>
<td></td>
<td>Density</td>
<td>(kg/m$^3$)</td>
<td>634</td>
</tr>
<tr>
<td>&lt; 2 mm (%)</td>
<td>(%)</td>
<td>0</td>
<td>pH</td>
<td></td>
<td>6.8</td>
</tr>
<tr>
<td>2–4 mm (%)</td>
<td>(%)</td>
<td>22</td>
<td>C (%)</td>
<td></td>
<td>3.56</td>
</tr>
<tr>
<td>4–6 mm (%)</td>
<td>(%)</td>
<td>24</td>
<td>H (%)</td>
<td></td>
<td>6.98</td>
</tr>
<tr>
<td>6–8 mm (%)</td>
<td>(%)</td>
<td>21</td>
<td>N (%)</td>
<td></td>
<td>0.45</td>
</tr>
<tr>
<td>8–15 mm (%)</td>
<td>(%)</td>
<td>33</td>
<td>S (%)</td>
<td></td>
<td>0.043</td>
</tr>
<tr>
<td>&gt; 15 mm (%)</td>
<td>(%)</td>
<td>0</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
in Fig. 6. Masoudinejad et al. [26] reported 90% of RE with an inlet hydrogen sulfide concentration up to 93.34 ppm after 3 wk of the operating system. They used the *Thiobacillus thioparus* on seashell bed biofilter. Roshani et al. [27] reported an average RE of 98% with retention time of 60 s and inlet hydrogen sulfide concentration of about 5–265 ppm. They evaluated the performance of biofiltration in the removal of H_{2}S from gas flue. In their study, they used a mixture of municipal solid waste compost and PVC bits as a bulking agent in 1:1 volumetric ratio. Kang et al. [28] reported a RE of H_{2}S over 99% with a retention time of 400 s. They used activated sludge to remove H_{2}S in biogas.

The maximum EC was obtained 52.32 (g/m^3/h) with the RE of 96.87% and hydrogen sulfide mass loading rate of 54 (g/m^3/h). However, at the RE > 99.96%, maximum EC was 26.99 (g/m^3/h) with hydrogen sulfide mass loading rate of 27 (g/m^3/h). The result of EC versus different EBRT and different hydrogen sulfide mass loading rate is shown in Fig. 7. Roshani et al. [27] reported a maximum EC of the biofilter about 22 g/m^3/h during the operating time with maximum inlet H_{2}S concentration of 265 ppm. Kim et al. [29] recorded a maximum EC of 8 g/m^3/h at a loading rate of 13 g/m^3/h. In their study, they used biofilter packed with biomedia, encapsulated by sodium alginate and polyvinyl alcohol (PVA). Ramirez et al. [30] reported a critical EC of 14.9 g/m^3/h with RE of 99.8%. However, they found a maximum EC of 55.0 g/m^3/h with RE of 79.8% and EBRT of 150 s. They investigated removal of H_{2}S by immobilized *T. thioparus* in a biotrickling filter packed with polyurethane foam.

The result of changes in pH versus operating time and different mass loading rates (g/m^3/h) is shown in Fig. 8. System was started with a packing material’s pH of 6.8. It reduced to 3.4 after 53 days of operating time and 54 g/m^3/h of mass loading rates. The reduction of pH is because of the production of sulfuric acid by sulfur bacteria. As Shinabe et al. [31] described, sulfuric odorants such as H_{2}S and methanethiol, were oxidized into sulfuric acid by sulfur bacteria.

Figure 4. Result of pressure drop (mm H_{2}O) versus operating time and different inlet concentrations of H_{2}S.

Figure 5. Changes of pH versus operating time and different inlet concentrations of H_{2}S.

Figure 6. Removal efficiency (%) versus different empty bed residence times and different mass loading rates.

Figure 7. Elimination capacity (g/m^3/h) versus different empty bed residence times (s) with different H_{2}S mass loading rates.

Figure 8. Changes of pH versus operating time and different mass loading rates (g/m^3/h).
which used these compounds as energy sources. Yang and Allen [32] reported that the pH at the inlet to their biofilter decreased from 8.0 to 2.5 after 32 days operating time. Kowal et al. (1991) cited in McNevin and Barford [10], 2000 observed a decrease in pH from 6.5 to 3.5 after 60 days of operating time their activated sludge biofilter.

As Roshani et al. [27] described, any increase in pressure drop adds to the operating cost of the filter because odorous air must be supplied at a greater pressure to achieve the same flow rate. The result of pressure drop versus operating time (day) and different mass loading rate (g/m³/h) is shown in Fig. 9. According to statistic analyze of Pearson correlation, there was direct and very high relationship between increasing of hydrogen sulfide mass loading rate and the amount of pressure drop (r = 0.95, p = 0.008). The relationship between different mass loading rate and the amount of pressure drop is shown in the Fig. 10.

Maximum pressure drop reached to 3.0 mm H₂O after 53 days of operating time and 54 g/m³/h of mass loading rates. McNevin and Barford [10] reported increasing pressure drop from < 500 to > 2500 pa after 3 months of continuous operating time. A maximum pressure drop of 18 mm H₂O was reported in Roshani et al. [27] study. They evaluated performance of biofiltration in the removal of hydrogen sulfide from gas stream.

![Figure 9. Pressure drop (mm H₂O) versus operating time (day) and different loading rates (g/m³/h).](image)

4 Conclusions

The results of this study showed high RE of hydrogen sulfide (> 99.96%) with EBRT of 90–45 s. High EC, up to 52.32 g/m³/h obtained with the RE of 96.87% and hydrogen sulfide mass loading rate of 54 g/m³/h. Maximum pressure drop was 3.0 mm H₂O after 53 days of operating time, the EBRT of 30 s, and 54 g/m³/h of hydrogen sulfide mass loading rate.

Based on the results of this study, mixed dried activated sludge and rice husk silica could be considered as suitable packing material for physico-biological filter to removal of H₂S. High performance of the filter, inexpensiveness, and easiness of providing of the packing material are the most important advantages of this packing material.

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References


