Cryptogamic Packed Biofilter as Potential Adsorbent for CO₂, NH₃, and H₂S Impurities from Biogas

Norbert W. Temba,¹,² Thomas Kivevele,¹ and Tatiana Pogrebnaya¹

¹Department of Materials, Energy Science and Engineering, The Nelson Mandela African Institution of Science and Technology, Arusha, Tanzania
²Pangani District Council, Tanga, Tanzania

Correspondence should be addressed to Norbert W. Temba; temban@nm-aist.ac.tz

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1. Introduction

The increase in energy demands due to population growth, depletion of worldwide oil reserves, and the problem of harmful emissions emanating from fossil fuel have put pressure on the world’s countries to use renewable energy [1]. Biogas is one of the renewable energy sources. It provides the reliable option of a sustainable form of energy derived from energy crops, agricultural wastes, agroindustrial wastes, municipal waste, etcetera. The quality of biogas is an essential aspect for its energy and economic value; it depends on the percentage composition of methane, carbon dioxide, and hydrogen sulfide. Biogas produced from a digester is used mainly for cooking and rarely for heating, lighting, and electricity generation, simply because of the presence of hydrogen sulfide and other impurities which together lead to corrosion of metal components in engines and gas burners. Typical raw biogas consists of about 55–65% methane, 30–45% carbon dioxide, traces of hydrogen sulfide, fractions of water vapors, and other contaminant gases [2]. The composition depends on the type of feedstock and anaerobic conditions. The H₂S and NH₃ components in biogas are (i) corrosive, which damages engines, and (ii) toxic [3]. In order to meet the standard for clean gas by increasing CH₄ concentration, the H₂S and other impurity content in biogas must be reduced. To achieve this, biogas must go through purification process [4]. Current technologies of biogas purification by removing H₂S, CO₂, and NH₃ employ the use of membrane separation, biological filters or activated carbon, and chemical liquids. These methods not only are costly but also have low removal efficiency [5]. Using iron oxide- (Fe₂O₃-) rich material is a simple, efficient method to remove H₂S in biogas since iron oxide readily reacts with H₂S to form iron sulfide (Fe₂S₃) when biogas passes through it [6].
\[
\text{Fe}_2\text{O}_3 + 3\text{H}_2\text{S} = \text{Fe}_2\text{S}_3 + 3\text{H}_2\text{O} \tag{1}
\]

\[
2\text{Fe}_2\text{S}_3 + 3\text{O}_2 = 2\text{Fe}_2\text{O}_3 + 6\text{S} \tag{2}
\]

Soil can be considered the fine earth which covers land surfaces as a result of the in situ weathering of rock materials or the accumulation of mineral matter transported by water, wind, or ice. Soils rich in metallic oxides are found to be effective in the removal of \(\text{H}_2\text{S}\) [3]. A process of soil biofiltration involves absorption of gas dissolved in soil solution which occurs as discharge air (raw gas) passes through a series of moist, aerated biological material through perforated pipes arranged in a bed material [7]. Soil biofiltration is considered among the best available technology and more foolproof operation, because of lower investment and operating costs; they are most suitable where the pollutant loading is in low concentration or low volume discharge [8], whereby degradation rates typically range from 10 to 100 \(\text{g m}^{-2}\text{h}^{-1}\) for common air pollutants. Primarily, they have been used for the control of foul odors and reduction in potentially toxic trace gases. The design employs either soil medium or compost in a system that makes use of natural soil processes for dissolution, adsorption, and microbial metabolism of inorganic gases and volatile organics present in the effluent air [7]. The desired filter volume depends mostly on the rate of the air pollutant loading (flow rate) relative to its degradation capacity and on the concentration of pollutants in raw gas. Another element that has been investigated for air biofiltration is biochar. It is known to be an efficient and cost-effective sorbent for different kinds of pollutant removal. Biochar contains unmodified activated carbon which in the presence of water slows down the degeneration process by promoting deposition of sulfur by mechanically removing sulfur from the active sites [9]. Sethupathi et al. [10] identified biochar as potential absorbers of \(\text{CO}_2\) and \(\text{H}_2\text{S}\) from biogas. The study analyzed adsorption dynamics of biochar derived from four different types of oak materials.

Another practice of gas purification and air pollution control involves the use of active biological material. This process employs biomonitors, bioaccumulators, and bioindicators in filtering air impurities using naturally occurring plants (mosses, lichens, and liverworts) and inorganic materials. Lichens and mosses can be used as biomonitors of air pollution because they are highly dependent on atmospheric sources for nutrients and do not shed plant parts as readily as vascular plants [11]. These organisms are scientifically referred to as cryptogams. They are made up of photoautotrophic communities, consisting of cyanobacteria, algae, fungi, lichens, and bryophytes, that is, liverworts and mosses in variable proportions [12]. These communities can provide food webs by photosynthesis and nitrogen fixation, which is particularly essential in arid regions and other terrestrial environments with a low abundance of organic nutrients. Cryptogams are estimated to be responsible for almost half of the total terrestrial biological nitrogen fixation of \(~49\cdot10^8\text{kg year}^{-1}\) and carbon uptake of \(~3.9\cdot10^9\text{ton year}^{-1}\) which amounts to \(~7\%\) of the net primary production of terrestrial vegetation [13].

These organisms are also well known to accumulate and retain a variety of contaminants, which make them useful in recording relative spatial and temporal deposition patterns of these contaminants. Integrating plants and soil biofiltration for the elimination of technogenic and volatile organic gases (VOC) has been studied to provide means of biofiltration without the use of consumables [14, 15]. Incorporating different media in a packed bed filter has shown successful results in biofiltration for a wide range of air pollutants and VOCs [7, 16]. Natural organic medium primarily composed of peat, compost, leaves, wood bark, and soil has been studied for the removal of toluene-contaminated air and VOCs [17]. In this study, we use the already analyzed soil physical-chemical properties in order to determine the effective optimal composition for improved removal efficiency of the biofilter. The effects of soil type, soil mass variation, and analysis of each substrate for the removal of \(\text{CO}_2\), \(\text{H}_2\text{S}\), and \(\text{NH}_3\) in the biofilter were illustrated. Also, the effect of gas flow rate using the soil with high removal efficiency was evaluated. The design and fabrication of the filter were established by considering the high surface area for reaction, pressure drop, moisture retention capability, and production rate of the biodigester.

2. Material and Methods

2.1. Soil Collection. The soil samples and profile statistics were collected from Tanzania Agricultural Research Institute located in Tanga, Tanzania, as part of the soil survey profile for Kwaraguru Sisal Estate, Tanga. In total, four soil samples were identified as likely candidates for evaluation based on guidelines of soil classification using World Reference Base for Soil Resources 2014 Edition [18]. The selected soil sample contained a high amount of metallic micronutrients per kilogram (the amount of metallic nutrient a plant can absorb via its roots), percentage of soil organic carbon, and carbon to nitrogen ratio. The soil samples with the high composition of extractable micronutrients (Fe, Zn, Cu, and Mn) from each profile were selected for this study. Other properties, including the percentage of organic carbon (%C), carbon to nitrogen ratio (C/N), and pH, were also considered for the selection. The selected soils were named T1, T2, T3, and T4 as depicted in Table 1.

2.2. Filter Preparation. To construct a prototype of the filter, each substrate, i.e., soils T1, T2, T3, or T4; biochar; and moss, was pretreated after collection, in order to remove unwanted components like roots, plastics, and leaves. The filter was designed to provide a high specific surface area for gas reaction with a low-pressure drop over the packed column. A combination of high content of active ingredients favors a substantial sulfide-binding capacity [19]. Each filter contained one of the four types of soil (T1, T2, T3, or T4) sieved at less than 400 \(\mu\text{m}\) with variable mass (100 g, 150 g, or 200 g); 100 g of biochar (charcoal) dust sieved at less than 350 \(\mu\text{m}\); and 545 g of naturally grown moss plant collected from damp sidewalk during the rainy season. The packed bed arrangement of the substrates was adopted from Pham et al. [20]; the study demonstrated removal of \(\text{H}_2\text{S}\) using...
local soil materials from biogas produced by anaerobic digestion of animal wastes. In this study, 700 g layer of small and medium gravels was added above the aerator to prevent blockage by overlying filter material. Moss plant was added as a top layer, with soil and biochar dust as preceding layers below it in order to ensure active gas exchange, growth, and sustainability of the filter. The substrates were packed into polyplastic containers each with the dimensions (25 cm × 16 cm × 9 cm) with an airtight cap to cover the top part; the container was transparent to allow just enough light in the filter to aid photosynthesis reaction of moss plants. An inlet port for the introduction of unpurified gas was inserted at the bottom part connected to the aerator (20 cm long) to ensure homogeneous distribution of the inlet gas across the face of the bed. A step-by-step preparation of the filter is as shown in Figure 1.

The bed included soils with a high amount of micronutrient content and %C, biochar dust (charcoal) which contains unmodified activated carbon, and moss substrate. The substrates were arranged in layers based on the role of each substrate in gas sorption efficiency and regenerative capacity. Filter fabrication was conducted at room temperature to ensure all the humid condition present in the moss plant is preserved. The humid condition in the filter is essential to ensure microbial action and enhance the sorption of CO₂ and H₂S. Xie et al. [21] showed that the presence of soil moisture content results in increased removal capacity for H₂S in a soil biofilter. The presence of water has also shown a beneficial effect on overall temperature catalytic performance of activated carbon for the oxidation of H₂S to sulfur [9]. After fabrication, the filter was then closed and sealed to begin measurements. It was designed to require no energy during its operation.

### 2.3. System Operation.

Two biogas reactors L1 and L2 available at private premises were used for experimental testing of the biofilter. Table 2 depicts features of both digesters as well as the average composition of raw biogas which was comparable by methane and carbon dioxide contents but slightly different by ammonia and hydrogen sulfide concentrations. In our on-site tests, biogas flow was allowed to pass through the filter media contained in a rectangular plastic container. The block diagram of the experimental setup is shown in Figure 2.

### Table 1: Soil database and profile information.

| Coordinates (Lat-Long) | Appearance | Sample name | Depth (cm) | Clay (%) | Texture class | Bulk density (g/cm³) | pH H₂O | Exch. Cu (mg/kg) | Exch. Zn (mg/kg) | Exch. Mn (mg/kg) | Exch. Fe (mg/kg) | Organic C (%) | C/N |
|------------------------|------------|-------------|------------|----------|---------------|---------------------|--------|-----------------|-----------------|----------------|----------------|--------------|-----|
| -5.42057° S, 38.52185° E | Dark reddish-brown | T1 | 0-15/20 | 62 | Clay | 1.02 | 5.2 | 7.47 | 0.99 | 16.5 | 33.99 | 1.49 | 6.77 |
| -5.42519° S, 38.52794° E | Very dark greyish brown | T2 | 20/25-40 | 40 | Clay | 1.34 | 6.9 | 7.77 | 0.48 | 47.1 | 31.29 | 1.36 | 4.69 |
| -5.48036° S, 38.52422° E | Dark reddish-brown | T3 | 0-15/20 | 50 | Clay | 1.18 | 5.4 | 2.25 | 1.62 | 117.0 | 51.39 | 1.77 | 8.05 |
| T4 | | | 15/20-40 | 58 | Clay | 1.21 | 5.4 | 1.5 | 0.36 | 39.3 | 57.69 | 1.10 | 6.47 |

Figure 1: Preparation of the biofilter: (a) inlet through aeration; (b) gravel packs (200 g); (c) biochar dust (250 nm/100 g); (d) soil substrate (100 g); (e) moss substrate (545 g); (f) filter open.
The gas flow rate was controlled using the gas regulator and LZB-3WB rotameter. The gas composition was recorded before and after the filter using a portable gas analyzer (GA5000 gas, Geotech, England). The overall pH of the filter measured before the experiment was 6.8. An experimental analysis by Nelson et al. [7] revealed that for the regular operation of soil-based biofiltration for air purification, the pH levels should be maintained at around 7 to 8, to prevent the formation of acids from the degradation of inorganic gases.

The tests were divided into three parts. The first part was conducted to analyze the effect of soil type on biogas purification in order to determine the sorption capacity of the adsorbents. The second part was aimed at analyzing raw gas adsorption by each substrate, and the third part was at summarizing the role of each adsorbent in gas purification which was conducted with flow rates of 80 and 100 ml/min during times 0-150 min for each filter in ambient condition. The arrangement of these substrates in the filter is as shown in Figure 3.

In order to examine the performance of each substrate in a filter, raw biogas was allowed to pass through a 5 g sample packed in airtight 5 ml plastic tube at a constant flow of 100 ml/min; the inlet and outlet concentrations of H₂S, CO₂, and NH₃ were measured; and the readings were recorded in a 10 min interval for 80 min. A small sample mass had to be used in order to ensure the maximum surface area for adsorption and to avoid a very high-pressure drop which is not recommended for biofilter operation. For example, Wellinger and Lindberg [22] and Abatzoglou and Boivin [3] showed from various studies that commercially packed biofilters are engineered to provide high specific surface area, having a low-pressure drop, and together with a high content of active ingredient provide high sulfate-binding capacity.

2.4. Filter Performance Parameters. The performance of the developed biofilter was determined in terms of removal efficiency (RE) and sorption capacity (SC). The CO₂, NH₃, and H₂S concentrations of treated gas at the outlet were analyzed at 10 min intervals for each test; RE and SC were calculated using Equations (3) and (4), respectively, whereby \( C_i \) and \( C_f \) are the initial and final concentration of H₂S (CO₂ or NH₃) as measured before and after filtration, respectively;

| Digester type | Digester volume (m³) | Feedstock | Volume gas column (m³) | Max pressure (mbar) | Raw gas composition |
|---------------|----------------------|-----------|------------------------|---------------------|---------------------|
| L1            | 16                   | Domestic sewage and garden wastes | 4                   | 107.8               | 68–72% CH₄, 38–40% CO₂, 6–14 ppm NH₃, 498–913 ppm H₂S |
| L2            | 9                    | Rougher garden wastes and overflow from L1 | 0.8                 | 84.3                | 64–66% CH₄, 34–38% CO₂, 3–7 ppm NH₃, 80–110 ppm H₂S |

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$Q$ is the flow rate ($m^3 h^{-1}$), $M$ is the atomic mass of sulfur, $V_{mol}$ is the molar volume of the gas ($l \text{ mol}^{-1}$) under standard conditions. A breakthrough time (BT) in hours was recorded as the time when $C_f$ approached 50% of $C_i$, where no significant adsorption of sulfur by the substrate was observed.

$$RE\% = \frac{C_i - C_f}{C_i} \times 100$$

$$SC\left(\frac{g \text{ sulfur}}{100 g \text{ sorbent}}\right) = Q \times \frac{M}{V_{mol}} (C_i - C_f) BT$$

Table 3: Soil-type performance in biogas purification (RE) average over an operating time of 2.5 hrs ($Q = 80 \text{ ml/min}$).

| Gas analyzed | Initial concentration ($C_i$) | RE (%), average over time 2.5 hrs | T1 | T2 | T3 | T4 |
|--------------|------------------------------|----------------------------------|----|----|----|----|
| $CO_2$       | 36.4–40.0%                   | 36.0 42.0 76.1 75.1              |    |    |    |    |
| $NH_3$       | 7–13 ppm                     | 56.3 nd 71.4 56.4                |    |    |    |    |
| $H_2S$       | 498–836 ppm                  | 83.5 84.0 93.3 97.1              |    |    |    |    |

*nd: not determined.

Figure 4: Biogas purification by filters with different soil samples: RE versus operating time. Test conditions: $Q = 80 \text{ ml/min}$, $m = 100 g$, $T = 19^\circ C$, $C_0 = 36.4\%$ for $CO_2$, 13 ppm for $NH_3$, and 836 ppm for $H_2S$. 
In order to elucidate the role of each substrate, special tests were conducted as described in Section 2.3; the SC of each biofilter component was calculated by using Equation (4).

### 3. Results and Discussion

#### 3.1. Effect of Soil Type on Biogas Purification

This experiment was conducted to assess the performance of the filter in terms of removal efficiency for CO$_2$, H$_2$S, and NH$_3$ impurities present in the biogas. Figure 4 shows the removal efficiency for CO$_2$, H$_2$S, and NH$_3$ from each soil type at a constant flow rate, with the test conditions and results summarized in Table 3. The results indicate that filters with T3 and T4 soils had the most substantial average removal of CO$_2$ at 76% and 75%, respectively. T3 filter also showed a significant RE of NH$_3$ at 72% on average over the entire period than other soil types. For H$_2$S removal, filters T4 and T3 showed the largest RE at 97% and 93%, respectively. The reason for higher RE exhibited by soils T4 and T3 is presumably due to the highest total content of extractable iron compared to T1 and T2, as shown in Table 1. As has been discussed, according to reaction Equation (1), high iron contents in the soil contribute to more significant interaction with H$_2$S impurities. Presence of other extractable micronutrients (Zn, Cu, and Mn) also favors reaction with H$_2$S.
The test results accord well with an investigation by Rodriguez et al. [23] where it was shown that H₂S and S₂ reacted faster with CuO and ZnO than corresponding sites of alumina (Al₂O₃) based on the bandgap size and chemical reactivity. Also, it was further observed by Ko [24, 25] that iron-rich and zinc-containing soils effectively removed H₂S from coal-derived natural gas. The soil T3 showed higher interaction with H₂S. The dark reddish-brown coloration of the soil indicates richness in the crystalline structure of iron oxide minerals which have a strong influence on H₂S removal capacity [19]. Also, Lasocki et al. [26], observed that the iron oxide present in soils and activated carbon (charcoal) as beds in filters reduced almost 100% of H₂S in a 30 min exposure period in the laboratory-scale investigation.

Other factors that contributed to better performance of T3 soil were high levels of soil organic matter, organic carbon percentage, and C/N ratio of the soil compared to other soils. Presence of organic matter improves soil aggregate and structural stability which are significant for infiltration of water and aeration into the soil, thus enabling the high surface area of reaction [27]. Soil organic carbon is vital in improving soil structure and provides habitat for soil biota, which together ensures sustainable yields as they retain water and nutrients [28, 29]. The level of C/N in the soil determines the rate of

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decomposition of organic matter and soil productivity, which was studied by using paddy fields by Li et al. [30]. Therefore, T3 soil was subjected to further tests in the effect of soil mass and flow rate to the removal efficiency.

3.2. Effect of Varying Soil Mass on Removal Efficiency. Further tests were performed to identify how the mass of the soil affects the removal of CO$_2$, NH$_3$, and H$_2$S from raw biogas. The experiment was conducted only T3 soil, which exhibited better results than other types of soils. The masses of other substrates and flow rate were kept constant at 100 ml/min. The filters that differed in soil masses were used, whereby T3-100, T3-150, and T3-200 referred to T3 soil substrate with a mass of 100, 150, and 200 g, respectively. The initial concentration of raw gas was in the range of 36.4–37.5% for CO$_2$, 7-13 ppm for NH$_3$, and 498-836 ppm for H$_2$S. Soils had the same particle size of <400 μm.

During operation time of 150 min, the outlet impurities were recorded as the raw gas passed into the filter. Figure 5 shows the removal efficiency of CO$_2$, NH$_3$, and H$_2$S gases. For CO$_2$, the removal efficiency was approximately 99.7% for T3-200 over the operation time. When the mass of the soil was further lowered to 150 g, the removal efficiency dropped to about 75% and further to nearly 70% for the filter with 100 g soil.

For the case of NH$_3$, RE was 83% but dropped to 33% for T3-200 and as high as 83% to 77% drop for T3-150. The filter T3-100 had a maximum RE of 80% before saturation. After 120 min, no more NH$_3$ adsorption was observed. We suppose this sharp decrease could be due to the drop in the initial concentration of the gas to the lowest amount recorded of 7 ppm and that low ammonia concentration measurement occurred at the sensitivity limit of the gas analyzer. T3-100 has a large surface area of reaction, unlike other filters with substantial mass, which are highly compacted thus having less adsorption rate. As it was demonstrated by Kent et al. [31], filter beds with small particle size not only offer a high specific surface area for reaction and thus favoring nitrification and microbial activity but also tend to constitute a greater resistance to gas flow which increases as biomass grows in bed porosities [32]. On the other hand, large particles have low-pressure drop since the gas flow is not blocked but offer fewer surface sites for oxidation reaction; therefore, it can lead to lower elimination performances [33]. Also, NH$_3$ has a low Henry’s constant ($H_{20°C} = 5.6 \times 10^{-4}$) [34] and a protonation constant $pK_a$ of 9.23 [35]. These values make NH$_3$ in biofilters to be partly retained onto the surface of carrier material by adsorption and by absorption of water fraction of the material [36].

The T3-100 filter showed higher RE in H$_2$S than T3-150 and T3-200, which can be explained by the availability of high surface area of adsorption between the gas molecules and the soil adsorbent containing a high content of iron and other cations, e.g., Cu and Zn. T3-200 due to a large mass of soil content has the highest gas retention time of all the filters, as it takes longer for the gas reaction to complete. The large mass of soil may also lead to clogging. It can be effective if the soil is evenly dispersed to allow more gas retention time. The higher retention time was effective in H$_2$S elimination capacity as it was studied by Yang and Allen [32].

From the results, one can conclude that with T3-100, there was a steady flow of raw gas from the aerator to the subsequent layers, thus having quick reaction time and a greater RE for H$_2$S and NH$_3$. On the other side, a large mass of soil causes the filter to be nonporous since the particles are tightly compacted together, resulting in the reduced surface area of reaction and large pressure drop.

3.3. Effect of Biogas Flow Rate on the Biofilter T3 Removal Efficiency. The H$_2$S, CO$_2$, and NH$_3$ removal were further analyzed for filter T3 by considering variation in flow rates of 80 ml/min and 100 ml/min. The filter performance with the soil mass 100 g (T3-100) is shown in Figure 6. As is seen, quite steady RE behavior is upheld for all three impurities, except a small and casual variation for NH$_3$ that can be attributed to a small but changeable amount (7-13 ppm) in the raw gas. The CO$_2$ removal is affected by the flow rate, the RE lowered by 7% from 80 to 100 ml/min. In the case of H$_2$S, the change of flow rate practically does not influence the RE that remains constant ≈94% during the filter operating time. A summary of removal efficiency at both flow rates is given in Table 4. Also, the effect of the soil mass is taken into account.

The RE for CO$_2$ was high for the filter with the largest soil mass at both flow rates. This can be attributed to the effect of pressure drop. The pressure drop is high for the most substantial packed bed in comparison to other filters. This, as a result, makes CO$_2$ concentration as it passes through the packed material in a given time to be significantly low, thus allowing time for maximum adsorption of the gas by the moss substrate. The same can be explained for the RE of CO$_2$ to be higher for the same flow with T3-150 than

| Gas analyzed | Initial concentration ($C_i$) | Flow rate, $Q$ (ml min$^{-1}$) | RE (average over time) (%) |
|-------------|-----------------------------|-----------------------------|----------------------------|
|             |                             | T3-100                  | T3-150                  | T3-200                  |
| CO$_2$      | 36.4–40.0%                  | 80                      | 76.1                    | 75.9                    | 99.7                    |
|             |                             | 100                     | 68.6                    | 71.1                    | 99.7                    |
| NH$_3$      | 7-13 ppm                    | 80                      | 72.4                    | 73.3                    | 50.0                    |
|             |                             | 100                     | 77.2                    | 69.7                    | 57.8                    |
| H$_2$S      | 498-836 ppm                 | 80                      | 93.3                    | 97.7                    | 96.7                    |
|             |                             | 100                     | 93.6                    | 85.9                    | 98.6                    |

Table 4: Effect of soil mass and biogas flow rate on the biofilter T3 performance; H$_2$S removal efficiencies are given as average values over 2-hour operating time.
T3-100. Meanwhile, with a low flow rate for T3-100 and T3-150 filters, high RE is observed. This can also be as a result of having sufficient reaction time for the adsorption of CO₂ by the substrate.

On average, the RE for NH₃ is high with T3-100 at 100 ml/min and drops as the mass of the filter increases (T3-150 > T3-200). Moreover, with the flow of 80 ml/min, there is a slight difference in RE for T3-100 and T3-150, but it drops for T3-200. Shoda [37] and Joshi et al. [38] demonstrated on a field-scale peat biofilter that NH₃ removal along with sulfur compounds can also be attributed to the adsorption by the acidic nature produced during oxidation of sulfur compounds.

By increasing the flow rate, the gas reaction time increases for the filter with larger mass (T3-150 and T3-200) and lower risk of clogging. But for the filter with small mass, the effect is reversed as more gas will pass through without being adsorbed by the soil adsorbent. For the T3-100 filter, the RE was considerably high for a low flow rate than a high flow rate. This allows a large number of particles present to interact with H₂S gas, offering more time for oxidation to occur. With a low flow rate, maximum gas reaction time was observed. These results are in agreement with studies from Chung et al. [39] and Singh and Mandal [40], whereby data on H₂S removal by combined chemical and microbial solution showed that RE for H₂S increased with increasing gas reaction time. In the investigation on operational parameters of biofiltration design by Yang and Allen [32], it was observed that an increase in loading rate results in a decrease in gas retention time, due to insufficient reaction time between H₂S molecules.

![Figure 7: Sulfur removal by various adsorbents: (a) breakthrough curves, (b) H₂S absorption over breakthrough time, and (c) sorption capacities. Test conditions: Q = 100 ml/min, m = 5 g, C₀ = 307-819 ppm for H₂S, and time = 80 min.](image-url)
and the biomass in the filter. Thus, H\textsubscript{2}S RE can be improved by either reducing the flow rate or increasing the soil volume. As seen in Table 4, the best performing filter for all gases by considering average RE was T3-100. Other filters showed better RE for one type of pollutant but less for the other type measured at either of the flow rates. For example, T3-200 showed high RE for CO\textsubscript{2} and H\textsubscript{2}S but less with NH\textsubscript{3}.

3.4. Analysis of the Function of Each Substrate

3.4.1. Sulfur Adsorption Performance of Various Adsorbents. The breakthrough time is used to specify the saturation time of the substrate before it needs regeneration or replacement. Longer breakthrough time implies a better adsorption capacity of the particular substrate (adsorbent) [41]. It also implies a shorter reaction time. Variations of the adsorbate concentration against the operating time of a filter are known as breakthrough curves [10]. These curves are presented in Figure 7(a) for different biofilter substrates, the soil T3, biochar, and moss. The sample was considered saturated when the outlet concentration reached 50% of the inlet.

The H\textsubscript{2}S breakthrough time for adsorbents T3 and T4 was both about 80 min, after which a minimal variation in sulfur adsorption was observed. Also, for the case of biochar and moss, the H\textsubscript{2}S breakthrough time (BT) was 50 min and 20 min, respectively. For other adsorbents, T1 and T2, a small change in their concentrations was observed; thus, no significant adsorption was detected. The adsorption breakthrough time of T3 and T4 was higher than that of other adsorbents (substrates). A comparison between initial and final H\textsubscript{2}S concentrations is shown for the substrates in Figure 7(b). The sulfur sorption capacity of each substrate was calculated using Equation (4) and indicated as the amount of sulfur adsorbed per 100 g of the sorbent in Figure 7(c). As seen, the soils T3 and T4 retain the highest H\textsubscript{2}S amount, 20.8 and 20.1 g/100 g, respectively, compared to other adsorbents whereas T1 and T2 soils are practically unable to adsorb sulfur. Regarding biochar and moss substrates, H\textsubscript{2}S was partly retained with a sorption capacity of up to 6.0 and 1.8 g/100 g, respectively. The results are comparable with data by Sahu et al. [42], whereby the sorption capacity of red mud (RM) sample used for H\textsubscript{2}S removal was 2.1 g-H\textsubscript{2}S/100 g of RM. High sorption capacity by T3 and T4 is comprehensible, as they both have a high total content of extractable cation, specifically iron, and high C/N compared to other soil substrates as shown in Table 1.

3.4.2. Moss Substrate Performance in CO\textsubscript{2} and NH\textsubscript{3} Removal. The capacity of a moss substrate on removing CO\textsubscript{2} and NH\textsubscript{3} from biogas was tested under the same conditions as described in the previous subsection about H\textsubscript{2}S sorption. A 59% drop in CO\textsubscript{2} concentration and 38% for NH\textsubscript{3} were recorded during the 60 min of the experiment Figure 8. For the case of CO\textsubscript{2}, moss plants like most cryptogams account for the largest net carbon uptake (photosynthesis minus respiration) [43, 44]. The present study demonstrated moss to be very effective in CO\textsubscript{2} adsorption from raw biogas for 60 min. A low adsorption rate for NH\textsubscript{3} is mainly because moss is a nonvascular plant as most ammonia needs to be converted first to ammonium and nitrate by nitrifying bacteria, and absorption by the plant from the soil is done by roots [38].

Ammonium is usually toxic to plants in any appreciable quantity as it causes degeneration and morphological...
Table 5: Absorption properties of the cryptogamic filter compared to other biofilters.

| Bed type/ adsorbent | Gas          | Impurity gas | Loading rate | RE (%) | Elimination capacity | Disadvantage                                                                 | Reference |
|---------------------|--------------|--------------|--------------|--------|-----------------------|-------------------------------------------------------------------------------|-----------|
| Peat biofilter      | NH₃          | 20 ppmv      | 1.8 g m⁻³ hr⁻¹ | 80     | 41 g m⁻³ d⁻¹          | Overloads at a loading rate of 7.9 g m⁻³ hr⁻¹. Prone to channeling and maldistribution. Limited ability to neutralize acidic degradation products [49] |
| Perlite biofilter   | NH₃          | 20-50 ppmv   | 8.6 g m⁻³ d⁻¹ and 21.5 g m⁻³ d⁻¹ | 99.5   | NA                    | Production of NO and NO₂ at the outlet [38] |
| Cow manure compost  | H₂S          | ~ 1500 ppm   | 58 m³ h⁻¹     | 80     | 16–118 g m⁻³ h⁻¹      | High operating costs, dogging to deposition of elemental sulfur [50] |
| Red mud             | H₂S          | 200 ppm      | 0.0048        | NA     | 2.1 g H₂S/100 g       | Due to the small mass of adsorbent, H₂S removal was low [42] |
| Biomass ash         | CO₂ and H₂S | 100–600 ppm  | 0.56–1.25 kg H₂S t⁻¹ ash, 35 kg CO₂ t⁻¹ ash, and 135 kg CO₂ t⁻¹ ash | 50 for H₂S, not feasible for CO₂ | NA                    | Not feasible for CO₂ removal, ashes could not be reused for as fertilizer [51] |
| T3, T4 soil substrates | H₂S           | 771-819 ppm | 0.0048 m³ h⁻¹ | 93, 97 | 20.8, 20.1 g S/100 g | A constant flow rate < 0.006 m³/hr to prevent large pressure drop This study |
| Moss substrate      | CO₂, NH₃, H₂S | CO₂ 31.6%, H₂S 307 ppm, NH₃ 13 ppm | 0.006 m³ h⁻¹ | 59 CO₂, 38 NH₃, 68 H₂S | 6.0 g S/100 g of sorbent | The flow rate < 0.0048 m³/hr to prevent intoxication of microbes by NH₃ and H₂S present This study |
| Cryptogamic filter  | CO₂, NH₃, H₂S | CO₂ 38%, NH₃ 8.7 ppm, H₂S 819 ppm | 0.006 m³ h⁻¹ | 99.7 CO₂, 77.2 NH₃, 98.6 H₂S | 11 g S/100 g | It does not work on high pressure, high flow rate operation, and requires water scrubber for removal of excess oxygen This study |

*NA: not applicable/addressed.
abnormalities [45, 46]. However, studies from Burkholder [47] showed that cultured moss had a high tolerance to ammonium ion (NH$_4^+$). It also enhanced the growth of some species of moss, e.g., fast-growing aquarium moss [48]. The NH$_4^+$ absorption can also be attributed to the thin layer of soil by which the moss is partially rooted.

3.5. Comparison of Biogas Purification with Other Biofilters of Different Media. Biological techniques can be the best alternative in the reduction of impurities in biogas, because the use of chemicals is limited, and it is considered economical and environmentally friendly. Investigations of different media for biogas purification in comparison to our study are summarized in Table 5.

In this study, NH$_3$ was removed to up to 77.2% at a constant flow of 0.006 m$^3$ h$^{-1}$ (100 ml/min). For CO$_2$ and H$_2$S removal, the physical and chemical properties of biochar in the filter have significantly improved the adsorption capacity. Sethupathi et al. [10] observed that the adsorption performance of a fixed bed column was significantly improved by physical and chemical properties of biochar on CO$_2$ and H$_2$S removal. It also revealed that biochar has a relatively low effect on CH$_4$ adsorption. Therefore, CH$_4$ content is unaffected during the whole adsorption process. Fernández-Delgado Juárez et al. [51] in their study on biogas purification with biomass ash found that the ash was successful in removing both CO$_2$ and H$_2$S, but to an extent, CO$_2$ removal was affected by high filter rates. Our study has addressed the flow rate to ensure maximum gas reaction time to allow considerable adsorption of CO$_2$ by biochar and moss substrates. The presence of moisture by which cryptogams thrive provides a humid condition for further removal of CO$_2$ and H$_2$S gases. The filter has integrated the physical and chemical properties of these substrates to facilitate the removal of these impurities from the gas. Other multiple gas removal filters were designed for the removal of more than one pollutant from the gas; results have shown significant removal for one gas but relatively poor for the other gases [51]. The primary advantages of biological filters were low energy requirements, mild operations, and regeneration capability of the materials [52, 53]. The disadvantage of using these methods was the low flow rate required for operation, additional nutrients were required, clogging of the bed due to sulfur deposit, and a small amount of O$_2$, N$_2$, and moisture were left in the treated biogas [6].

4. Conclusion

Biofiltration system integrated with four types of soils, T1, T2, T3, or T4, with biochar and cryptogamic cover, was designed and tested on-site for removal of biogas impurities in the form of CO$_2$, H$_2$S, and NH$_3$. The function of each substrate was elaborated via a special experimental arrangement. Among the samples, the T3 and T4 soils demonstrated the best H$_2$S removal with sorption capacities of 20.8 and 20.1 g S/100 g and RE of 93% and 97%, respectively. This good performance of the soils attributed to their high iron content, large cation exchange concentration, and organic carbon percentage. Moss and carbon-based material (biochar) showed inadequate H$_2$S removal, indicating that pure physical adsorption was not efficient on digesters operating at low pressures such as in typical farm. Moss substrate had significantly high CO$_2$ RE of 59% and as low as 38% for NH$_3$ during the 60 min of the experiment.

Integrating these substrates in a bed arrangement of a packed filter improved the overall adsorption of CO$_2$, NH$_3$, and H$_2$S. The most important advantage of a cryptogamic biofilter over other physical-chemical technologies is that it is applicable in a wide range of pollutants (impurities) and effective in low concentration, can be used under ambient conditions (pressure, temperature, and pH), and consumes very little energy, and materials are easy to obtain, simple to operate, and economic. Another advantage is flexibility, as they are quick to respond to the changing pollutant characteristics because of the wide range of microbial metabolic pathways.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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