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Removal of Hydrogen Sulfide from Biogas by the *Acacia Auriculeaformis* Activated Carbon

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Abstract: Biogas is one of the most attractive renewable resources because of its ability to convert waste into energy. Biogas is produced during an anaerobic digestion process from various organic waste resources. It is composed of mainly CH_4, CO_2, and some trace gases such as hydrogen sulphide (H_2S) which is a very toxic, deadly and corrosive gas. Therefore, raw biogas must be cleaned of hydrogen sulphide (H_2S) before being used in many applications. Activated carbon is commonly used for adsorption due to its high surface area, micro porosity, thermal stability, high removal capacity and low cost compared to other adsorbents. The general objective of this work was to study the removal efficiency of hydrogen sulphide (H_2S) by the *acacia auriculeaformis* activated carbon. The *acacia auriculeaformis* is a tree that can be exploited for wood charcoal because of its rapid growth, even on infertile sites, and its tolerance to very acidic and alkaline soils. The carbonization of the *acacia auriculeaformis* branches were done using an oven at 550°C for four hours and activated by a 1 mol/L sodium hydroxide solution. The physicochemical parameters such as Iodine adsorption number, ash content, point zero-charge pH (pH_{ZPC}), and tapped density were determined to characterize the synthesized activated carbon. The tests of H_2S elimination by adsorption on activated carbons were carried out at the poultry farm FONDATION BRIN, located in the village YAOKOKOROKO, sub-prefecture of TABAGNE in the GONTOUGO region. This farm has an anaerobic digestion with a capacity of $15m^3$ for the treatment of the chicken manure it produces. Two types of filtration columns were used: a 15 cm column with a capacity of 15 g of carbon and a 30 cm column with a capacity of 30 g of carbon. The iodine value, ash content, moisture content, pH_{ZPC}, tapped density of the prepared activated carbon were 609.12 mg/g, 2.38%, 11.16%, 7.73 and 1.51 respectively. These results indicate that the prepared activated carbon is microporous (0-2 mm), of good quality and lightweight. Furthermore, the prepared activated carbon samples have a removal efficiency (RE) of H_2S, during the working time (10 h), higher than 97% for both types of columns used with H_2S output concentrations lower than 10 ppm which is the tolerance threshold for prolonged exposure. These results are similar with commercial activated carbon. The *acacia auriculeaformis* activated carbon can be used to remove hydrogen sulphide from biogas.

Keywords: Biogas, Hydrogen Sulphide, Activated Carbon, Adsorption, *Acacia Auriculeaformis*

1. Introduction

Biogas is formed by the anaerobic microbial decomposition of organic substances that produces not only the potential component methane (CH_4), but also undesirable impurities such as hydrogen sulphide (H_2S) and carbon dioxide CO_2 [1, 2]. Hydrogen sulphide (or H_2S) is a pollutant present in most biogas. Its purification is necessary to preserve the equipment from premature corrosion and also to protect humans and the environment [3].

Several methods have been investigated to remove hydrogen sulphide from the biogas stream. These methods include chemical methods [4, 5]; biological methods [6] and physical methods [7]. In addition, scrubbing is one of the methods for hydrogen sulphide removal that employs the use of various scrubbing agents such as water and chemicals. However, a large quantity of scrubbers is required, so they are expensive. Some scrubbers, especially chlorinated chemicals, produce secondary pollutants [3].

The adsorption method is one of the most practical technologies to remove hydrogen sulphide from biogas [8]. To avoid the problem of cost, especially in low-income settings, researchers are focusing on finding cheap adsorbents using available natural resources [9].

The activated carbon is among the adsorbents for hydrogen sulphide removal from biogas as a result of its surface properties that make it effective in the adsorption process. In a recent study, authors prepared activated carbon from water hyacinths that achieved hydrogen sulphide removal efficiencies up to 93% [7].

The Acacia auriculeaformis is an exploitable tree for charcoal (stem and branches larger than 4 cm in diameter) because of its rapid growth, even on infertile sites, and its tolerance to highly acidic and alkaline soils [10-12]. It is used for stabilization and revegetation of mines [13]. In Côte d'Ivoire, the National Centre for Agronomic Research has a plantation of this species near Abidjan (ANGUELEDOU forest) [14].

The general objective of this work is to study the performance of activated carbons for the adsorption of hydrogen sulphide from biogas, prepared from a local biomass that can be exploited for coal production. Specifically, this will involve: (i) synthesize activated carbons from acacia, (ii) characterize the obtained carbon, (iii), study the removal efficiency of hydrogen sulphide generated by the *acacia auriculeaformis* activated carbon.

2. Materials and Methods

2.1. Synthesis Protocol of Activated Carbon

The preparation of activated carbon from the *acacia auriculeaformis* branches was done according to the usual method [15, 16]. The dried *acacia auriculeaformis* branch pieces (figure 1) were carbonized at 350°C for 4 hours using the muffle furnace. Any carbonization residues were removed by washing thoroughly with distilled water. The obtained materials were oven dried at 105°C for 24 h, then ground to have particles with diameters between 125 µm and 2 mm and dispersed in a 1 mol/L sodium hydroxide solution. After stirring for 30 min, the mixture was kept at rest for 24 h. Finally, the resulting slurry was filtered and oven dried at 105°C for 24 h. The dry residue was washed with distilled water until the wash water was neutralized. The material was then oven dried at 105°C for 24 h.

Figure 1. *The dried acacia auriculeaformis branch pieces.*

2.2. Characterization of the Acacia Auriculeaformis Activated Carbon (AAC)

2.2.1. Humidity Content

The humidity content is determined by drying the adsorbent in an oven. For this, 0.5 g of activated carbon (AAC) is introduced into a ceramic crucible and the whole is weighed. After drying in an oven at 105°C for 24 h [16], the assembly is cooled to room temperature and weighed again. The humidity content (% H) is calculated from the following formula:

$$\%H = \frac{(m_2 - m_3)}{m_1} \times 100 \quad (1)$$

m_1: the initial mass of the AAC used (in g).
m_2: the mass of the crucible + AAC before drying (in g).
m_3: The mass of the crucible + AAC after drying (in g).

2.2.2. Ash Content

The ash content is the inorganic, inert, amorphous and unusable part present in the activated carbon. Thus, a 3 to 4 g sample of activated carbon is placed in a ceramic crucible. The sample is weighed and then introduced into the oven set at 650°C for 3 hours. After cooling down to room temperature, the sample is weighed again [16]. The ash content (C) is calculated from the following formula:

$$C\ (\%) = \frac{m_2 - m_0}{m_1 - m_0} \times 100 \quad (2)$$

m_0: the initial mass of the AAC used (in g).
m_1: The mass of the crucible +AAC before carbonization (in g).
m_2: The mass of the crucible +AAC after carbonization (in g).

2.2.3. Iodine Adsorption Number

The Iodine adsorption number is an indicator of the

mesoporosity of an activated carbon. For its determination, a mixture of 0.05 g of activated carbon and 15 mL of a 0.1N iodine solution is stirred for 4 min. After filtration, 10 mL of the filtrate was titrated with a 0.1N sodium thiosulfate solution in the presence of two drops of the starch solution. A blank test was performed under the same conditions in the absence of activated carbon. The iodine value can be calculated from the following formula:

$$I_d = \frac{(V_b - V_S) \times N \times 126.9 \times (\frac{15}{10})}{m} \quad (3)$$

Id: Iodine adsorption number (mg/g).

(V_b-Vs): difference of the results of the blank and adsorbent titration (in mL of sodium thiosulfate).

N: normality of the sodium thiosulfate solution in (eq.g/L).

126.9: atomic mass of iodine (in g/mol).

m: the mass of activated carbon in (g).

2.2.4. Determination of Point Zero-Charge pH (pH_{ZPC})

The pH_{ZPC} or pH of zero point charge corresponds to the pH value for which the net charge at the activated carbon surface is zero. A stock solution of NaCl (0.1 mol/L) was prepared. Different NaCl solutions (0.1 mol/L) at different (initial) pH (2, 4, 7, 9 and 10) were prepared. The pH was adjusted with NaOH (1M) or HCl (1M). Then 0.1 g of carbon was added to the different solutions and the whole was stirred with a magnetic stirrer for 48h. After stirring, the solutions are filtered and the (final) pH of the filtrate is noted. Finally, the isoelectric point (zero charge pH) was obtained by plotting the ΔpH curve according to Equation (4) [16].

$$\Delta pH = pH_f - pH_i \quad (4)$$

2.2.5. Tapped Density

The tapped density of the materials was calculated by measuring the volume of a compacted sample mass in a graduated cylinder. However, this measurement is not very precise. It can be calculated by the following formula:

$$d = \left(\frac{m2 - m1}{V}\right) / \rho \quad (5)$$

m_1 and m_2 the respective masses of the empty and filled test tube.

V: The volume of the graduated cylinder (10 mL).

ρ: The density of the water (1 g /cm^3).

2.3. H_2S Removal Tests by Adsorption with Activated Carbon Based on Acacia Auriculeaformis

The collection of biogas was carried out at the poultry farm FONDATION BRIN, located in the village YAOKOKOROKO, sub-prefecture of TABAGNE in the GONTOUGO Region. This farm has an anaerobic digester with a capacity of $15m^3$ for the treatment of the manure it produces.

The unfiltered biogas is stored in the air chamber (tank). It is conveyed through the pipes to the filtration column. Two types of filtration column were used:

1) a column of 15 cm with a capacity of 15g of carbon;

2) a column of 30 cm with a capacity of 30 g of carbon.

The H_2S concentration is determined at the inlet and outlet of the filter column using a portable biogas detector (figure 2). During the test period; the biogas flow rate was kept constant with a value of 0.146 m^3 /min or 0.00244 m^3/s. The figure 3 shows the adsorption test setup.

Figure 2. *Portable biogas detector.*

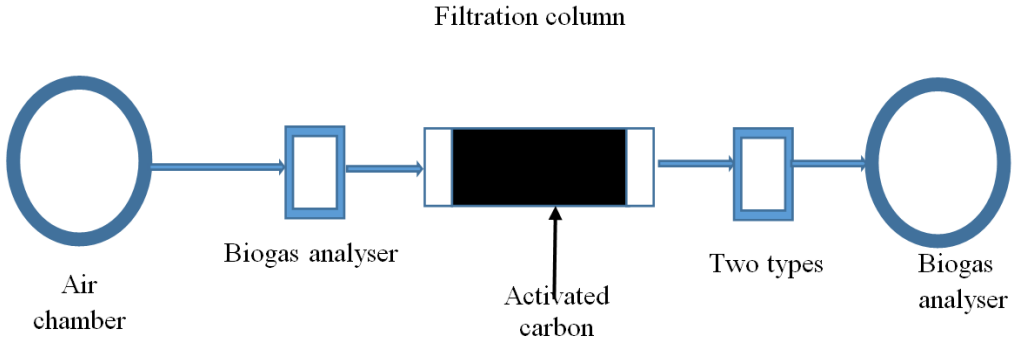

Figure 3. *Schematic of the adsorption test.*

3. Results and Discussion

3.1. Characteristics of Activated Carbon

The knowledge of activated carbon characteristics is necessary to contribute to the understanding of many phenomena such as adsorption, desorption, exchange, etc.

Table 1 shows some of the characteristics of the prepared activated carbon.

The Humidity content of the *acacia auriculeaformis* activated carbon (AAC) is 11.16 %. With a low Humidity content, this activated carbon could have a high higher heating value (HHV) [17, 18]. The ash content is 2.38%. The ash content for AAC is low. One of the parameters influencing the adsorption properties of carbon is its ash content. This parameter has a significant effect on the quality of the activated carbon. It appears that a high ash content decreases the specific surface area. Therefore, the ash content of a good adsorbent should not be too high, i.e. below 20% [19]. Too high an ash content (>20%) reduces the activity of the carbon, its reactivation potential and may generate impurity (mineral salt) leakage. The ash content reported in this study is an indication of the good adsorption capacity of this activated carbon [19]. The pH value of the zero charge point (pH_{ZC}) was obtained using the graph in Figure 4. The pH_{ZC} of the activated carbon is 7.73. The *Acacia auriculeaformis* activated carbon has a basic character. This would indicate a low oxygen content in this carbon [20]. The pH_{ZC} corresponds to the pH value for which the net charge of the adsorbent surface is zero. It makes it possible to determine the acid or basic character of the activated carbon and to know, according to the pH of the solution, its net surface charge. The density value is 1.51. The density varies according to the type of materials used. This value indicates that the *Acacia auriculeaformis* activated carbon is lightweight.

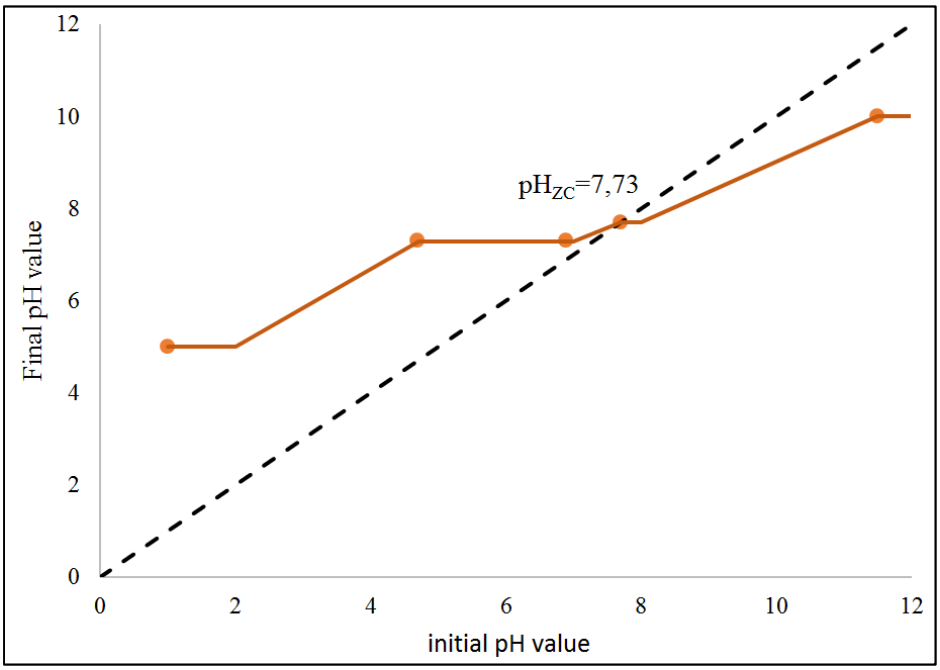

Figure 4. *PH_{ZC} of the acacia auriculeaformis activated carbon.*

Table 1. *Characteristics of activated carbon.*

Iodine adsorption number (mg/g)	point zero-charge pH pH_{ZPC})	Ash content (%)	Humidity content (%)	Tapped density
609.12	7.73	2.38	11.16	1.51

The activated carbons with sodium hydroxide (NaOH) contain pores accessible to iodine molecules. The iodine value is 609.12 mg/g. Lower iodine values (319.67 and 286.26 mg/g) have been reported in the characterization of acacia activated carbons prepared with basic agents [21]. The iodine adsorption number depends on the surface porosity and is thus useful in characterizing the surface area of carbon black. [21]. In the case of our study, the *acacia auriculeaformis* carbon activated with sodium hydroxide presents better results (values higher than 500 mg/g) [21, 22].

3.2. Study of Hydrogen Sulphide (H_2S) Removal from Biogas by the Acacia Auriculeaformis Activated Carbon

The installation includes a anaerobic digester which produces a biogas composed of methane (CH_4), carbon dioxide (CO_2), carbon monoxide (CO) and hydrogen sulphide (H_2S), as shown in Table 2. Changes in H_2S concentration before adsorption were also monitored during the working time and showed no change in the initial H_2S concentration (Table 2). This means that the initial H_2S

concentration is constant during the working time.

Table 2. *Biogas composition.*

Constituent	Measure 1	Measure 2	Measure 3	Measure 4
CH_4	85-90 %	85-90 %	85-90 %	85-90 %
CO	10-15 %	85-90 %	85-90 %	85-90 %
H_2S	80 – 100 ppm	80 – 100 ppm	80 – 100 ppm	80 – 100 ppm

The AAC was used to remove hydrogen sulphide (H_2S) from the biogas. Figure 5 shows the H_2S concentrations at the outlet of the filtration column. This graph is a function of time for a 15 g mass of CAA. Using a 15 g mass, the AAC did not reach their piercing time within the working time (10 hours). The piercing time is the time for which, the H_2S concentration of filtration column outlet becomes half of the initial concentration (40-50 ppm). Furthermore, the CAA samples have a removal efficiency (RE) of H_2S greater than 97% (Figure 6). This indicates development of pores essential for adsorption [23, 24].

Using a mass of 30 g, the ACC did not reach their piercing time as in the 15 cm filtration column in the working time (figure 7). Furthermore, CAA has a very high H_2S removal efficiency (RE) of over 98 % (figure 8). It is due to the increase of the number of adsorption sites with the increase of the activated carbon mass.

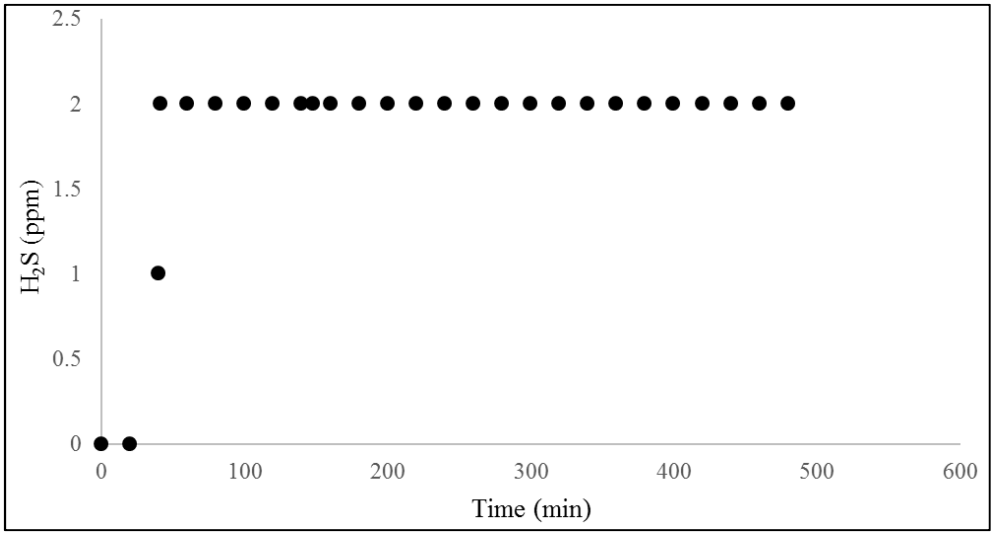

Figure 5. *Variation of H_2S concentration for 15 g of CAA.*

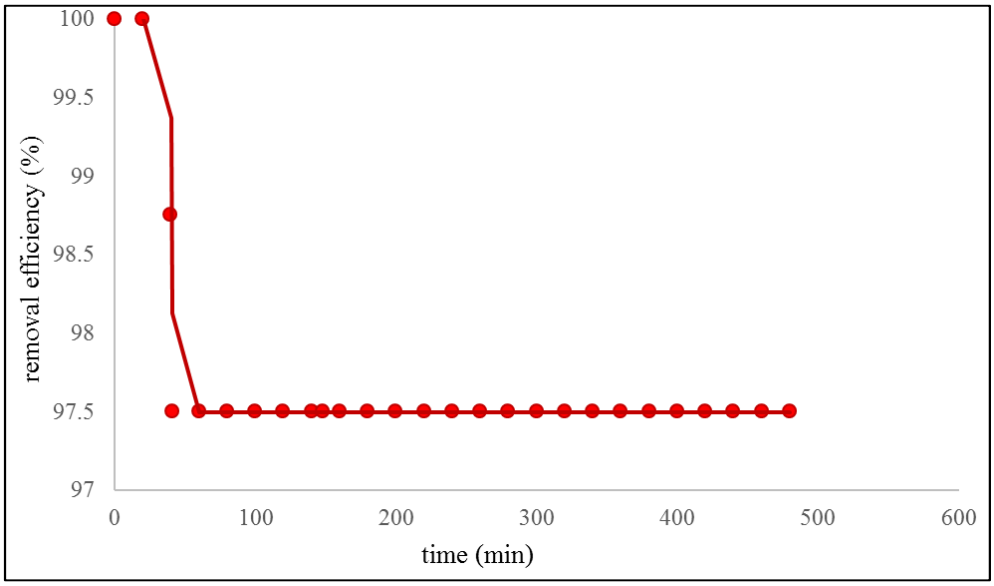

Figure 6. *H_2S removal efficiency (RE) for 15g of CAA.*

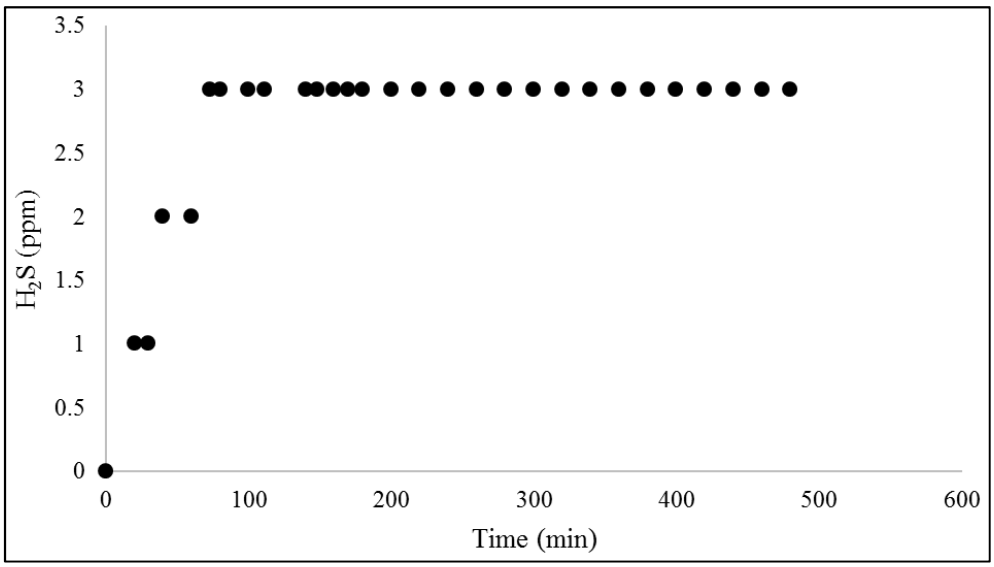

Figure 7. *Variation of H_2S concentration for 30 g of CAA.*

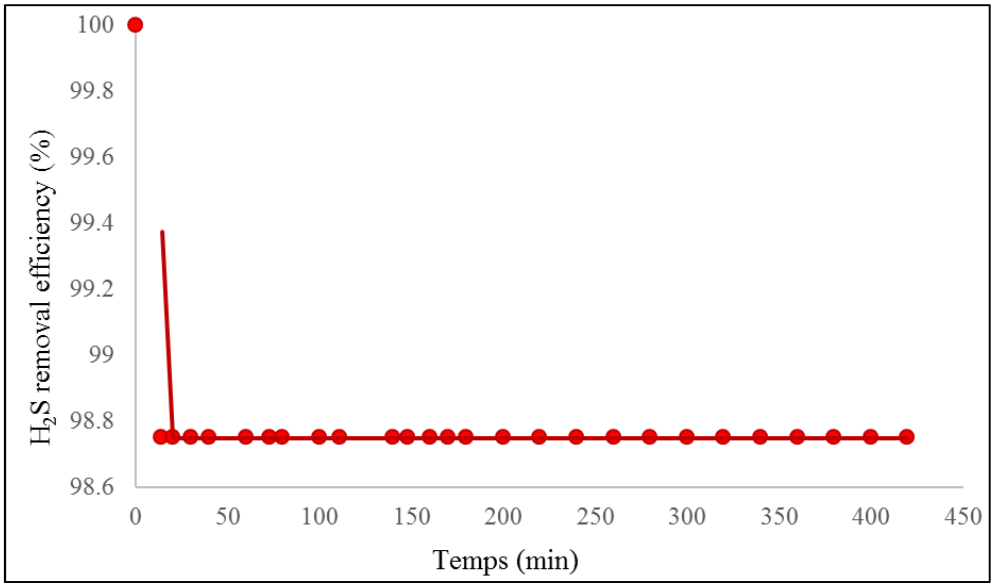

Figure 8. *H_2S removal efficiency (RE) for 30 g of CAA.*

4. Conclusion

This study aims to investigate the removal efficiency of hydrogen sulphide (H_2S) by the activated carbon of *acacia auriculeaformis*. The carbonization of *acacia auriculeaformis* is done using an oven at 550°C for four hours and activated with a 1 mol/L sodium hydroxide solution. The iodine value, ash content, humidity content, point zero-charge pH, tapped density of activated carbon were 609.12 mg/g, 2.38%, 11.16%, 7.73 and 1.51 respectively. These results indicate that this activated carbon is microporous (0-2 mm), of good quality and lightweight. Furthermore, the activated carbon samples have a removal efficiency (RE) of H_2S, during the working time (10 h), higher than 97% for both types of columns used with H_2S output concentrations lower than 10 ppm which is the tolerance threshold for prolonged exposure. The *acacia auriculeaformis* activated carbon can be used to remove hydrogen sulphide from biogas. However, the results obtained in this study should be completed. For this purpose, it is envisaged to perform the biogas filtration test over a long period of time in order to determine the breakthrough time and to carry out other tests with different masses of activated carbon in order to determine the optimal mass.

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