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Research Article

Comparison Study of Biogas Upgrading Removal of H₂S by Using Adsorption, Absorption and **Bio-filtration** Techniques

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Abstract

Energy is a critical need for our planet, and as the population and demand grow, it will be important to find new renewable energy sources. Biogas is a by-product of the decomposition of organic waste materials under anaerobic condition by using a digester, since all biogas streams commonly contain harmful impurities, such as Hydrogen Sulfide (H₂S), a chemical compound that is widely referred to as extremely poisonous to humans, corrosive, and very flammable. This study demonstrates how to purify biogas by removing the H_2S concentration. The H₂S was removed by means of Absorption, Adsorption and Biological filtration unit which converts H₂S into elemental Sulphur (S). These results are compared with those three H₂S removal techniques and the results demonstrate that, a larger percentage age of H_2S eliminated by adsorption technique have better removal efficiency (94%) the second one is Bio-trickling filter (85%) and the third one that have lowest efficiency is absorption technique (82.7%). Further research may be conducted to improve or enhance the efficiency of these techniques.

Keywords: Absorption; Adsorption; Biogas Upgrading; Biotrickling; H₂S removal

Introduction

Energy is a critical need for our planet, and as the population and demand grow, it will be important to find new renewable energy sources [1]. The depletion of non-renewable source of energy has become very high as a result of the growing of life style, technology, and industrialization, so that there are various renewable energy sources available, of which biogas is one. Biogas is a by-product of the decomposition of organic waste materials under anaerobic condition by using a digester. It contains mainly of Methane (CH₄) and Carbon Dioxide (CO₂). Few other components, such as Water (H₂O) in various proportions, Carbon Monoxide (CO), Hydrogen Sulfide (H₂S), Halogenated Hydrocarbons, Oxygen (O₂), Nitrogen (N₂) Siloxanes and Ammonia (NH₃) can be present and might be

inconvenient when not removed. Biogas production has many benefits for the atmosphere, businesses, and individuals involved. It's a renewable energy source for the local grid, providing both electricity and heat. Benefits to the environment include recirculation of organic waste from business and households in an environmentally sustainable manner. When spreading slurry on the fields, there are less odour issues.

Because of the cheap start-up costs and the abundance of waste materials, biogas could be particularly beneficial in rural or poorer areas. It can be made from a variety of organic materials, so biogas digesters are available in broad variety of sizes and forms. Agricultural waste, urban wastewater, industrial wastewater and municipal solid waste are also handled by certain industrial systems. Animal waste is normally digested using small-scale systems.

The separation of methane from carbon dioxide and other gases in biogas is known as biogas upgrading [2]. Biomethane, or renewable natural gas, is a concentrated biogas that contains up to 100% methane. This gas may be pumped into the gas distribution network or may be utilized as a vehicle fuel. Wastewater treatment plants and anaerobic digesters will all get benefit from biogas upgraded technology. Biomethane is biogas that has been improved to a quality that is comparable with natural gas that is fossil and contains methane content of 90% or higher. Biomethane is used as a truck fuel as well as a natural gas grid injection. The grid entry unit in a natural gas network controls the flow of gas into the system and normally requires that the gas quality be of adequate quality for entry.

Anaerobic digestion

Anaerobic Digestion (AD) is a phenomenon wherein the bacteria decompose organic materials without the requirement of oxygen, e.g. cow dung, wastewater bio solids, and agricultural waste. Anaerobic digestion for the generation of the biogas follows in a sealed container called a reactor that may be designed and built in a variety of forms and dimensions depending upon the location and feedstock circumstances. The waste is broken down or digested in these reactors, which produces end-products of the AD process i.e. the biogas or digestate.

Factors affecting biogas production

Biogas development, and thus bacterial activity, is influenced by a number of parameters. Its output is influenced by a number of factors that influence gas yields from various substrates. Feed stock properties like substrate accessibility, Carbon to Nitrogen (C/N) Ratio, Temperature value, Loading Rate, Hydraulic Retention Time (HRT), Agitation, Additives are some of the factors. The control of these factors is crucial for maintaining a proper balance between the accumulation of volatile acids and the growth of methanogenic bacteria in a digester.

Biogas cleaning and upgrading

The removal of trace components such as NH3, H2S etc. as well as bulk contaminant CO2 gas drying and compression are all part of the biogas upgrading phase, which results in a biomethane commodity gas stream with a methane content of usually more than 95% and very low levels of hazardous contaminants [3]. Purification enables a broader range of applications, including heat and power, as well as automotive



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fuels. Purification to extract CO_2 and H_2S is needed before it can be used as a fuel because H_2S corrodes critical mechanical components. If it is not removed, it will become a component of engine generator sets and car engines.

Hydrogen Sulphide gas removal

For all but the simplest burner applications, Hydrogen Sulfide in biogas must be extracted. Sulfuric acid (H_2SO_4) is formed when Hydrogen Sulfide reacts with water vapour in raw biogas. It is extremely corrosive to engines and components. H_2S is extremely toxic at concentrations greater than hundred parts per million by volume (ppmv). Activated carbon can be used to get rid of odours. H_2S is catalytically converted to elemental Sulphur using activated carbon. Biogas can also be scrubbed for Hydrogen Sulfide using Sodium Hydroxide (NaOH), water, or iron salt solutions. Dosing a biogas stream with O_2 , which converts H_2S to elemental Sulphur, is a simple and low-cost technique. The following are biogas purifying processes that are feasible.

- Absorption into liquid (Physical or Chemical)
- Membrane separation
- · Cryogenic separation
- Biological removal
- · Adsorption on solid surface

Adsorption via activated carbon

Activated carbon has a huge surface area, porosity, and surface chemistry, and that is why researchers have been interested in studying adsorption through it. Activated carbon has a high potential for separating contaminants from liquids and gases in a contaminated atmosphere due to its high porosity and surface region. A certain amount of water vapour must be present for the oxidation of H₂S, and it can be used interchangeably with water, which is adsorbed and forms a water layer on the pore surface. H₂S and O₂ migrate down into the carbon pores, where they are trapped. The O2 binds to the carbon surface and decomposes into radicals. The oxygen radicals react with the dissolved Hydrogen Sulfide ions to produce elemental Sulphur and O2 impregnated and un-impregnated. The term "impregnated activated carbon" refers to the addition of cations to aid in the adsorption process as a catalyst. When compared to unimpregnated activated carbon, this impregnated activated carbon has the best ability to remove H₂S. This complex process limits the rate of reaction since un-impregnated activated carbon is a poor catalyst.

Absorption technique

A contaminant existing in a gas stream is absorbed into a liquid *via* absorption techniques used in air pollution control. A number of organic solvent were utilised in some kind of a rack or closed-bed arrangement to preferentially capture H_2S as well as CO_2 from CH_4 and lighter alkanes. Based on the intensity of the engagement between gas particles mostly with solvent as well as the type of the absorbing isotherm, such liquids can be categorised under physical and chemical groups. The dissolution of such gases inside the liquids grows faster and at a lower pressures, and then plateaus at more pressures in chemisorption, the peak reflects the stoichiometric capability of the process involving chemicals.

Biological filtration unit

Bio-scrubbers, Bio-filtration, Bio-Trickling Filters (BTF), and the Air Injection are all examples of biological-based H_2S removal technologies. The bio-filter is a fixed bed bioreactor where the microorganisms utilised in the degrading process are immobilised in a packing media. Inoculation of microorganisms is used in bio-desulfurization. Contaminated gases are allowed to travel through the porous medium. Contaminants enter the biofilm that surrounds the packing material and microbial colony, where they are degraded. Open design bio-filters and enclosed design bio-filters are two forms of biofiltration. The open design necessitates a big space and is typically positioned outside with just rising gas flow. The closed bio-filter has a smaller volume and the flow can be rising or decreasing.

Materials and Methods

In recent years, there has been an increase in biogas production. As a result, biogas purification and upgrading have been extensively researched in recent years based on those literatures. We'll look at a few of them in this paper.

The H_2S elimination from the biogas was investigated utilising a novel adsorbent, laterite, due to their high iron (II) oxide content (Ferrous Oxide) [4]. As a result, laterite has the potential to be one of the most effective materials for biogas clean-up. The study's purpose was to see if entrapping CO₂ and H_2S could be used to clean small-scale biogas facilities [5]. It is possible to conclude that how the biomass ashes may be utilized to eliminate H_2S in either larger, medium or smaller biogas systems. The experimental work by the authors seeks to spread H_2S elimination procedure among the users by integrating a cheap chemical absorption technology with closed reactors [6]. At a rate of up to 92.41%, single purification columns were capable of eliminating Hydrogen Sulfide. They were able to achieve a 96.84% purity using multiple purification columns.

By using experimental methodologies, the absorbing compounds that are generated from the waste are pre-activated to remove H_2S from a biogas stream [7]. Purifying a biogas stream by eliminating H_2S with waste-derived adsorbent materials holds a lot of potential. In a study, a bench-scale and pilot-scale iron-redox biological method was investigated to remove H_2S [8]. The system comprised of a BTF with useable content of 47 L along with an Absorption Bubble Column (ABC) with useable contents of 3 L. H_2S was eliminated at a rate of more than 99.5% as a result. A activation process of the compounds generated from the waste was investigated experimentally by the authors at a temperature of about 700°C in a study [9]. The activated biochar's adsorption capacity was comparable to that of commercial sorbents, whereas the adsorption capacity of ashes is still insufficient.

An experimental BTF was employed to treat genuine biogas in the investigation, which took place in a wastewater treatment plant [10]. The statistical findings demonstrated a high degree of prediction and a great concordance seen between the models and the analytical data. The 'C model' and the 'EC model' respective values for the R-squared was 95.77% and 99.63%. The dynamic behaviour of a reaction chamber holding treated sludge that was derived from the sewage for biogas desulfurization is studied by the author utilising two modelling approaches. The first uses Bohart and Adams' fundamental model, while the second utilizes Linear Driving Force (LDF) model, which is calculated methodically using the Klinkenberg formula and arithmetically using Aspen Adsorption Simulation.

Activated carbons produced from pine tree uses sawdust pellets as adsorbents for NO₂ and H₂S removal in this investigation [11]. Textural characteristics, surface acid-base character, and lastly the adsorption characteristics of the activated carbons were studied in relation to activation temperature. The partition coefficients (K) of H₂S and CO₂ in an H₂S, CO₂ or CH₄ combination were investigated using activated carbon produced from Brazilian babassu coconut husk at various pressures and temperatures in this work [12]. The results reveal that by using activated carbon as an adsorbent, H₂S and CO₂ may be separated from a mixture that also contains CH₄.

As per this research, following the sorption of Hydrogen Sulfide on heating a modified zeolite 13X at 350°C, the chemisorbed amounts of Hydrogen Sulfide were quite less [13]. The process includes the adsorption of Hydrogen Sulfide on the surface of zeolite, H_2S solubility and dissolution within pore fluid, including oxidation for generating Sulphur which might be polymerized. Researchers compared biological and chemical desulfurization utilizing identical iron filtration to evaluate the minimization of H_2S [14]. The results show that iron filters removed 32.91% of H_2S , with a maximum of 70.21%. Iron filters have a substantially lower average H_2S removal effectiveness than micro aeration, two and four hours after retention (90.8% and 98.9%, correspondingly). The increased retaining period led to greater average removal effectiveness as compared to the previous one.

The investigators wanted to explore how different residues and treatment methods affected bio-char yield and reactivity [15]. Magnitude of the particle, hotness, and inorganic substance concentration were all factors in the treatment. Biochar created using the husk of the olives was much more aggressive during the gasification compared to that of the biochar made with the corncob due to its greater ash content. The H₂S adsorption to form activated carbon has been simulated using models that comprised the sorption isotherm, kinetic models, mass balance, according to the researchers [16]. The discontinuity mostly in the size of the sorption bed post tuning under homogeneous circumstances without any drop in pressure and continuous velocity is examined using real data given by the industry owing to the greater rate of flow and concentration of H₂S. Using corn stalk as a raw material and zinc chloride as an activator, this study looked at the best conditions for making desulfurizing stalk carbon [17]. H₂S adsorption was aided by the increase in micropores and the abundance of oxygen-containing functional surface groups. The principal desulfurization products discovered were elemental Sulphur and Sulphite.

Design

Agriculture waste Collection (Crop residue, Slaughter house and another residue).

Mechanical pre-treatment

Carried out by mills which breaks open the cellulose structure and increases the specific surface area of the biomass Particle size ranging from 1 to 2 mm (effective hydrolysis)

Design of the homogenizing tank

Rectangular domain that have two dimensional with an impeller or shaft at the middle of it in vertical axis. The cylindrical vessel diameter D=0.3 m and height H=0.375 m and speed of impeller 0.5 Rotation Per Minute (RPM).

Location

Direction of the digester facing to the sun help to increase the temperature of the biomass before they fed into the digester and it will resist thermal shock when cold water added.

Inlet and Outlet

The input and output pipes lead straight into the digester at a steep angle. The diameter of the pipe of the liquid medium is between 10 to 15 m.

Design of the Digester

The volume of the digester can be calculated as

Volume=flow rate (m³/day)*retention time (days)

Design of Gas Collecting Dome

The gas dome depends on the volume of gas to be produced. Thus, Volume of the gas dome=volume of gas to be produced. Total Chemical Oxygen Demand (COD) to be digested for the producer of the gas is given by: COD*daily input. Calculation of methane gas (CH₄) equivalent to that of the oxygen O_2 .

Equivalent Chemical Oxygen Demand (COD) for the production of CH₄ through complete AD:

- Calculation of Chemical Oxygen Demand (COD) equiv of CH4
- CH+2O=CO+2HO
- (16 g) (64 g) {atomic weight}
- Thus, 16 g CH=64 g O (COD)
- Thus, 1 g CH=64/16 g of O (COD)
- Thus, 1 g CH=4 g COD

The change of CH_4 mass equiv. to volume equiv. According to the Gas Law by Avogadro, one mole of any gas at standard temperature and pressure holds a volume equal to 22.4 liters. Thus,1 mole CH_4 (16 g)=22.4 L, CH_4 1g=1.4 L CH_4 .

The CH₄ amount of formation per unit of Chemical Oxygen Demand (COD) removal from above, we can observe that,1g CH₄=4g COD~1.4 L CH₄

Since, biomass contains 60% (approx.) of CH₄

Thus, volume of gas dome=total biogas production of the day since, biomass contain 2% (approx.) of H₂S thus, required amount of H₂S is 2% of total biogas production.

Hydrogen Sulfide (H₂S) removal comparison techniques

Adsorption method calculation: The quantity of H_2S trapped on the adsorbent was determined as the value of the difference between the initial value of the gas concentration and the desorbed amount of concentration of gas. The adsorption efficiency (in %) and the adsorption capacity (in mg) were determined using equations.

- Adsorption Capacity (q) is given by the equation
- (q)=[(Ci-Cf) *V]/M.(1)
- Where,
- Ci: inlet concentration,Co: outlet concentration.
- V: bed volume, and
- M: activated carbon mass.

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- And Adsorption Efficiency is given by:
- (η)=[(Ci-Cf)/Ci*100](2)
- And also,
- Filtration rate (FR)=Q(Flow rate)/A (cross-sectional area),
- τ (Empty bed contact time)=Volume of bed/Q (Flow rate)
- Breakthrough time=Amount consumed/AC consumption rate

Absorption method calculation

According to the reaction, chelated iron oxidizes sulfide, resulting in the creation of Sulphur.

 $S^{-2} + 2Fe^{+3} \Leftrightarrow S^0 + 2Fe^{+2}$

The oxygenation allows it to regenerate.

 $\frac{1}{2}O_2(g) + H_2O(l) \rightarrow \frac{1}{2}O_2(aq)$

 $\frac{1}{2}O_2(aq) + 2Fe^{+2} \rightarrow 2Fe^{+3} + 2OH$

The equation expresses the entire process.

 $H_2S + \frac{1}{2}O_2(g) \rightarrow S^0 + H_2O$

From the ideal gas equation of state:

PV = RT(3)

T where $Ru = \frac{8.314kJ}{kmol.k}$ and m is the species molecular mass

The gas constant R is defined as:

$$R = \frac{Ru}{m}$$
Then,
$$R = \frac{8.314kj/kmol.}{34.08kg/kmol} = \frac{0.244kJ}{kg} \cdot k$$

$$VH_2S = \frac{RT}{PH2S} = \frac{RT}{xH2SPtot} = \frac{\left(\frac{0.244kJ}{kgk}\right)(298K)}{xH2SPtot} \qquad (4)$$
% Efficiency (η)=[(Cin-Cout)/Cin] *100

Bio-trickling filter method calculation

Some metrics are necessary to evaluate the operating conditions and efficiency of a bio-filtration system. Some are specific to bioreactors for pollution control, and they've been used to characterise all process designs.

Mass Loading Rate (L) is input mass fed to bioreactor in unit time and carrier volume $[g/(m^3h)]$. It can be stated as follows:

$$L = \frac{Cin.QV}{V} \tag{5}$$

Were, Cin representing the gas inlet concentration, QV representing the volumetric flow rate, and V representing the bed volume. Empty Bed Residence Time (EBRT) is the time it takes for gas to pass through all of the packing volume, and is defined as follows:

$$EBRT = \frac{v}{q_v} \tag{6}$$

Elimination Capacity (EC) is just the proportion of contaminant reduced per unit of time and filter volume, and it is calculated as: $Ec = \frac{(Cgin - Cg.out).Qv}{v}$

Removal Efficiency (RE) can be expressed as follows: $RE = \frac{Cgin-Cg.out}{1} * 100\%$

Results and Discussion

Waters from Agriculture that includes crop residue, slaughter house and other residue wastes are processed to produce biogas. The preconditioning of the feed materials is the first step in the biogas generation process. Water is added to the feed material, which includes a substantial portion of recirculated flow from the output (digestate) and the daily inflow is 36 L/day or 0.36 m³/day, retention time or detention time

(7)

(8)

is 20 days, temperature in the digester is 35° C, potential of Hydrogen (pH) is 6.5, volume of the digester is 0.72 and we get total biogas production is 0.84 m³, amount of Hydrogen Sulfide (H₂S) is 0.0168 m³ and methane (CH₄) is 0.504 m³.

The second objective of this thesis is to select three biogas upgrading technologies (absorption, adsorption and bio-trickling filter) where these upgrading technologies are identified and described and it was found that different upgrading technologies have different H₂S removal efficiency as shown in Figure 1, Figure 2 and Figure 3.



Figure 1: Illustrates the relation between flow rate and volume of treated gas in adsorption of Hydrogen Sulfide (H₂S).

The study discovered that raising the flow rate of biogas increases the adsorption capacity. This phenomenon could be explained as "increasing biogas flow rate causes a slightly better discharge ability and the gas residue in activated carbon is lower". However, as described above and shown in Figure 1, in overall, the total adsorption capacity of the activated carbon is better at a higher flow rate. *Citation:* Mahlet T, Aravindan MK (2022) Comparison Study of Biogas Upgrading Removal of H₂S by Using Adsorption, Absorption and Bio-filtration Techniques. J Nucl Ene Sci Power Generat Technol 11:6.



Figure 2: Illustrates the relation between time and volume of treated gas in adsorption of Hydrogen Sulfide (H_2S).

The amount of adsorbate adsorbed at any given time is at the area just above breakthrough curve and was directly related to contact time, so it is essential in the adsorption process since the equilibrium contact time must be calculated as depicted in Figure 2. May be adsorption capacity can increase with increase in contact time if the researcher have not performed the process until it reaches its equilibrium.

When calculating adsorption capacity, the adsorbent dosage is the most important factor to consider. As the mass of adsorbent in a fixed volume reaction mixture increases, the number of active sites possible for adsorption increases, but the adsorption capacity declines, as they are inversely proportional. Although elimination value will increase. If we decrease the initial concentration the lower concentrations adsorption capacity will surely decrease as again the sufficient adsorbate is not able to get adsorbed on the active sites and there is also the interference of the other ions.

Therefore from the above method and calculation the amount of removal of H_2S is 94% so it has higher adsorption capacity.



Figure 3: Illustrates the relation between temperature and volume of treated gas in absorption of Hydrogen Sulfide (H_2S).

To investigate the impact of the pressure ratio on the absorption rates were conducted with different Pressures Ratios (PR) at constant temperature as depicted in Figure 3. The absorption rates for at the pressure ratios of Pr=101.325, 202.325, 303.325, 404.325 and 505.325 pa at 25°C. Based on the above trend, it was concluded that the

absorption rates were found to be higher for high absorption and high amount of pressure, so that they were pressure dependent. Temperature is a significant element in the H₂S absorption process, as H₂S absorption reduces at high temperatures due to temperature's direct influence on several reactive species such as corrosion and volatility as shown in Figure 4. The rise in temperature of the solution in the absorption column is directly related to solvent deterioration, corrosion, and the creation of stable salts.



Figure 4: Illustrates the relation between temperature and volume of treated gas in absorption of Hydrogen Sulfide (H_2S).



Figure 5: Illustrates the relation between outlet concentration and removal efficiency in Bio-Trickling Filters (BTF).

The characteristics of the bio-desulfurization of H_2S concentration biogas in a BTF were 85% investigated as shown in Figure 5, and in this process the amount of outlet concentration increases and the removal efficiency will be decrease. From the above method and results adsorption technique has better removal efficiency (94%) the second one is Bio-trickling filter (85%) and the third one that have lowest efficiency is absorption technique (82.7%). *Citation:* Mahlet T, Aravindan MK (2022) Comparison Study of Biogas Upgrading Removal of H₂S by Using Adsorption, Absorption and Bio-filtration Techniques. J Nucl Ene Sci Power Generat Technol 11:6.

Conclusion

The type of material used to make biogas has a big impact on the composition of the gas production and the purification process. There are a variety of biogas upgrade methods available and this research compares three biogas upgrading technologies. Adsorption by activated carbon absorption by chemicals and Bio-filtration methods are the main techniques. The results showed that the CH_4 concentration is improved by reducing the H₂S content of the biogas to a considerably lower concentration and results adsorption technique have better removal efficiency (94%) the second one is Bio-trickling filter (85%) and the third one that have lowest efficiency is absorption technique (82.7%) Another aspect to notice is that though most literatures indicates that temperature has no influence on absorption but in my finding temperature does have a small effect on the absorption process.

Finally, the operating cost, power, water and chemicals, scale, quality of biogas generation, targeted quality of biogas, site application, and process economics are all factors that influence a technology's suitability. The technology chosen, the quality and quantity biogas used, and the target output quality all influence the capital and operational expenses of biogas upgrading technology, and the unique investment costs of different upgrading technologies. This study has a higher output or value because it was based on an experimental study and also used software such as Aspen HYSYS and Cumulative Distribution Function (CDF) model to predict the concentration of impurities removed from a produced volume of methane gas in order to achieve the required methane purity in an upgrading process.

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