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Optimization of Amine-Based Absorbent Solutions for Biogas Purification from Cow Manure

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Abstract: The growing demand for energy and the depletion of fossil fuel resources have increased the urgency of finding renewable and sustainable energy sources. Biogas produced from cow manure through anaerobic digestion offers a promising option because it can turn agricultural waste into usable energy. However, raw biogas contains a high proportion of carbon dioxide (CO_2), which reduces its calorific value and must be removed to improve its quality. This research evaluates the effectiveness of four amine based absorbents, namely monoethanolamine (MEA), diethanolamine (DEA), triethanolamine (TEA) and methyldiethanolamine (MDEA), for purifying biogas in a packed absorption column. The column was filled with Pall rings to enhance gas and liquid contact and operated at room temperature (around 25 °C) and atmospheric pressure with a constant flow rate of biogas. Each amine solution was prepared at concentrations between 10% and 50%. The methane and carbon dioxide contents were measured before and after the purification process to assess removal efficiency. The findings indicate that MEA achieved the highest level of carbon dioxide removal, raising methane concentration from 58.3% to 78.2% at a 50% solution. DEA also showed good performance, although not as high as MEA, while TEA and MDEA produced lower removal efficiencies. These results demonstrate that amine based absorption can significantly improve the quality of biogas. In summary, MEA proved to be the most effective absorbent under the conditions tested, offering a practical and cost effective approach for small to medium scale biogas purification systems. These findings provide useful information for improving chemical absorption techniques to support wider use of renewable energy.

Keywords: Amine solutions; Biogas purification; Chemical absorption; Methane enrichment; Packed bed column.

Introduction

Energy is essential for human comfort and daily life. However, in many parts of the world, especially in developing countries, energy shortages remain a major concern due to the continued reliance on fossil fuels (Gürsan & de Gooyert, 2021; Kabeyi & Olanrewaju, 2022; Mohammad et al., 2021). In addition to creating an imbalance in energy supply, the extensive use of fossil fuels has also led to serious environmental consequences, particularly the rise of greenhouse gas emissions. Among these gases, carbon dioxide (CO_2) is recognized as the primary contributor to global warming, accounting for approximately 80% of all greenhouse gas emissions (Ochedi et al., 2021). Therefore, the development of renewable energy sources that can help reduce CO_2 emissions has become increasingly important in achieving energy sustainability.

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Biogas represents a promising alternative as it is a renewable energy source that can partially replace fossil fuels for heat, electricity, and power generation (Akkarawatkhoosith et al., 2019). Biogas is a combustible gas mixture produced through the anaerobic digestion of organic materials such as agricultural residues, food waste, and wastewater (Fatin et al., 2021; Nur Daiyan et al., 2020).

This biological process, driven by microbial activity, generates gas primarily composed of methane (60–65%) and carbon dioxide (35–55%), with smaller amounts of hydrogen, hydrogen sulfide, nitrogen, and moisture (Kapoor et al., 2020; Sihlangu et al., 2024). The presence of carbon dioxide in biogas decreases its calorific value, causes corrosion in equipment, increases ignition difficulty, and enlarges storage volume requirements for the same energy yield (Soehartanto et al., 2021; Ullah Khan et al., 2017). For instance, biogas with a methane to carbon dioxide ratio of 60:40 has a heating value of about 21–23.5 MJ/m^3 (Kabeyi & Olanrewaju, 2022), which is considerably lower than the 35.64 MJ/m^3 of pure methane (Akkarawatkhoosith et al., 2019). When carbon dioxide is effectively removed, the resulting high-purity methane, or biomethane, has similar characteristics to natural gas and can serve as a cleaner fuel alternative (Jensen & Skovsgaard, 2017).

Various techniques have been developed to remove carbon dioxide from biogas, including physical absorption (Tantikhajorngosol et al., 2019), chemical absorption (Abdeen et al., 2016; Daiyan et al., 2020; Maile et al., 2017), adsorption (Fourqoniah et al., 2023), cryogenic distillation (Yousef et al., 2018), and membrane separation (Baena-Moreno et al., 2020). Among these, chemical absorption is widely applied because of its low operational cost, high efficiency, and solvent regeneration capability (Akkarawatkhoosith et al., 2019).

In the absorption process, a liquid absorbent is used to separate certain gas components based on their solubility (Kalsum et al., 2022). During biogas purification, the biogas flows through the absorbent solution, where impurity gases are absorbed while methane remains in the gas phase due to its very low solubility (Singhal et al., 2017). Therefore, selecting an absorbent that can selectively remove unwanted gases without absorbing methane is crucial. Amine-based solutions such as monoethanolamine (MEA), diethanolamine (DEA), methyldiethanolamine (MDEA), and triethanolamine (TEA) are commonly used because they can remove carbon dioxide both physically and chemically while enhancing gas–liquid mass transfer through chemical reactions (Tamhankar et al., 2015). This method generally operates at ambient temperature and atmospheric pressure with a short residence time (Kumar Gupta & Tuohy, 2020).

Theoretically, chemical absorption is based on gas and liquid equilibrium and the chemical reaction between acidic gases such as carbon dioxide and amino functional groups in amine solutions, forming carbamate or bicarbonate compounds. This concept reinforces the scientific foundation of this research, as the effectiveness of carbon dioxide absorption depends on the type and concentration of the amine used.

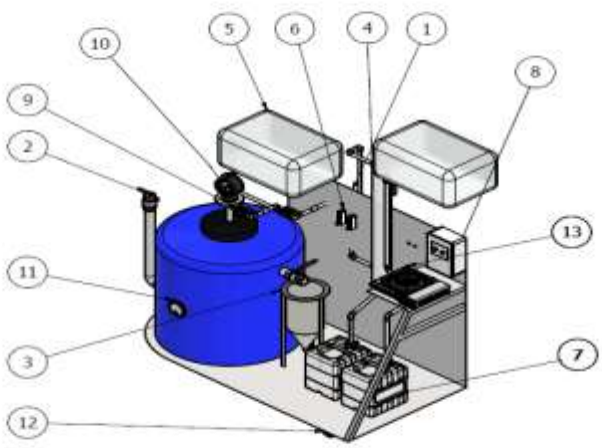
This research is conducted because improving the efficiency of biogas purification is essential to support the broader use of renewable energy at both local and national levels. Moreover, identifying the most effective amine solution can reduce operational costs and enhance biogas quality, making it more suitable as a substitute for fossil fuels.

This study aims to upgrade biogas through chemical absorption using different amine solutions, specifically by comparing MEA, DEA, TEA, and MDEA as absorbents. In this process, the amine solution is introduced at the top of the absorption column, while biogas is fed from the bottom. As both phases interact along the packing material, carbon dioxide is absorbed into the amine solution, resulting in an increase in methane concentration in the purified biogas.

Method

Location and Time of Research

The study was carried out at the Chemical Engineering Laboratory, Sriwijaya State Polytechnic, from April to June 2025.



1	Frame structure	8	Control panel
2	Digester tank	9	Valve
3	Slurry line	10	Pressure gauge
4	Column absorber	11	Temperature gauge
5	Gas storage bag	12	Wheel
6	Rotameter	13	Stove
7	Liquid container		

Figure 1. Biogas purification unit

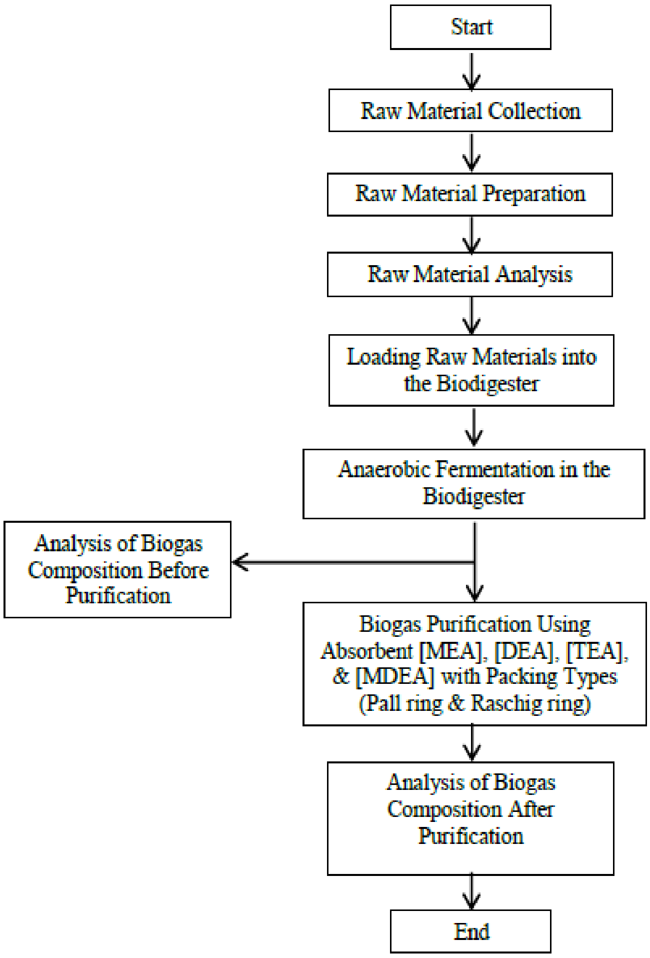


Figure 2. Flowchart of the Research Procedure

Biogas Production from Cow Manure

Biogas in this study was produced using fresh cow manure as the primary material. The manure was mixed with clean water in a 1:2 ratio, forming a slurry that could flow easily into the digester. The mixture is carefully stirred, then 50 mL of prebiotic is added to help boost biogas production. In this study, a fixed dome biodigester with a total volume of 250 litres is used. The digester is filled with slurry to 80% of its volume, while the remaining 20% is left empty to provide space for biogas accumulation.

Biogas purification was performed when production reached its peak, typically around day 21 of the fermentation process, consistent with (Tetteh et al., 2018), who reported that the optimal time for anaerobic digestion generally occurs on day 20. Experiments with tomato waste have shown that maximum methane production (0.42 m^3/kg VS) can be achieved at a Hydraulic Retention Time (HRT) of 24 days, indicating that peak biogas production is influenced by substrate type, organic loading rate, and temperature.

Generally, HRT for the co-digestion of fruit–vegetable waste and industrial organic waste tends to exceed 20 days; however, anaerobic co-digestion is commonly operated at HRTs between 10 and 20 days (Tufaner & Avşar, 2016). These findings underscore that the timing of peak biogas production depends on substrate composition and operational conditions, necessitating careful monitoring to determine the optimal point for biogas collection and subsequent purification.

Biogas Purification Using a Packed Column

This research employed a chemical absorption technique to purify biogas, utilising a vertically positioned packed column as the primary unit. The column was constructed using clear acrylic, allowing for visual inspection during operation. Packing materials were inserted into the column to expand the contact surface between the gas and liquid phases. Packing section in the absorption process plays important role providing surface area for the gas and liquid phases to contact upon (Arachchige et al., 2012). Two types of packings were examined: Raschig rings and Pall rings. Both types are widely used in gas treatment systems due to their support for effective mass transfer.

Raschig rings are simple cylindrical pieces with a height equal to their diameter. In this study, stainless steel Raschig rings were used because of their chemical resistance and mechanical durability, particularly when exposed to amine-based solutions. Their structure allows random flow distribution and provides a suitable surface area for general gas absorption. Produced in large numbers, Raschig rings form what is known as random packing, commonly applied in chemical engineering processes such as distillation, transformer oil filtration, and other applications requiring efficient gas–liquid contact. Named after the German chemist Friedrich Raschig, these rings are also used in devices where gas and liquid interact for absorption, stripping, or chemical reactions, and they can serve as support for biofilms in biological reactors, enhancing mass transfer and process efficiency (Ramesh & Moorthi, 2017a).

Pall rings are a modified version of Raschig rings, featuring side openings and internal supports that enhance fluid contact and reduce flow resistance. In this study, Pall rings made from plastic were also assessed. Their design improves liquid dispersion and airflow, which is particularly beneficial at higher flow rates. Despite their advantages, stainless steel Raschig rings were chosen as the standard packing due to better availability and reliable performance throughout the trials. The decision to use Raschig rings over Pall rings was based on their proven performance in similar applications and their compatibility with the experimental setup.

A support plate was placed inside the column to ensure even distribution and secure placement of the packing material. Biogas from the digester was supplied to the base of the column with the help of a compressor. Before entering the column, it flowed through a rotameter, which measured and regulated its flow rate. For the absorbent, amine solutions were stored in a container positioned below the column. These solutions were pumped upward through a rotameter, used to monitor and control the flow, before being introduced at the top of the column. Within the column, the gas rose from the bottom while the liquid absorbent moved downward through the packing. This countercurrent arrangement facilitated effective interaction between the two phases, allowing the amine solution to absorb impurities such as carbon dioxide and hydrogen sulfide.

The purification process was conducted at room temperature and atmospheric pressure, without additional heating or pressurization. The biogas flow rate was maintained at 0.4 L/min, while the absorbent (amine solution) flow rate was set at 0.8 L/min. These flow rates were selected to ensure adequate gas and liquid contact time, allowing efficient absorption of CO_2 and H_2S along the countercurrent packed column. A higher gas flow could reduce residence time, lowering absorption efficiency, whereas a lower absorbent flow might lead to premature saturation and insufficient gas scrubbing. The chosen absorbent flow rate of 0.8 L/min was also supported by the findings of (Putra et al., 2023) in their study, which demonstrated that 0.8 L/min provided the optimal absorption performance, balancing mass transfer efficiency and system stability. Flow rates were continuously monitored and adjusted using rotameters to maintain consistent and reproducible conditions throughout the experiments.

After passing through the column, the purified gas exited from the top and was collected in a gas bag. The used absorbent, which exited from the bottom, was collected in a separate container. Before each run, the entire system, including pumps, tubing, valves, and clamps, was meticulously checked for leaks or irregularities. The system was also rigorously rinsed between trials to eliminate residue from previous runs. The column was secured to a sturdy vertical frame, and all connections were tightly sealed to prevent any leaks, ensuring the accuracy of the results.

This packed column system was selected for its reliable performance in supporting gas–liquid mass transfer within a compact and manageable setup. The stainless steel Raschig rings promoted turbulence and liquid film formation, thereby enhancing contact between phases. This arrangement enabled repeated trials under steady conditions, providing consistent results throughout the experimental procedures.

Effect of Amine Type on Biogas Purification

To investigate the influence of amine choice on purification performance, four different amine solutions were prepared and tested: monoethanolamine (MEA), diethanolamine (DEA), triethanolamine (TEA), and methyldiethanolamine (MDEA). Each solution was mixed using distilled water as the solvent.

The absorbents were stored in individual containers and introduced to the packed column through flexible tubing. All test conditions, including flow rates and column setup, remained consistent during the trials to ensure that differences in performance were attributed solely to the type of amine used. At the end of each experiment, the used absorbent was collected in a separate container. The system was flushed and cleaned thoroughly before beginning a new trial with a different amine solution. This approach helped eliminate the influence of residual chemicals and ensured accurate comparisons between amines.

Effect of Amine Consentration on Biogas Purification

The influence of amine concentration on biogas purification was also evaluated. Each type of amine solution was prepared at five different concentration levels: 10%, 20%, 30%, 40%, and 50% by volume. Solutions were freshly mixed before use to maintain consistency. All other operational variables, such as biogas composition, temperature, flow rates, and equipment configuration, were kept constant during the tests. This allowed the observed effects to be directly related to changes in concentration.

After each test, the system was drained and rinsed to prevent cross-contamination. The equipment was allowed to stabilise before the subsequent trial commenced. This ensured that the outcome of each concentration test reflected only the solution used, not residue or leftover from the previous run. By applying a structured and systematic procedure, the study thoroughly examined and compared the influence of different amine concentrations on the purification process, utilising a repeatable and well-controlled experimental setup. This approach instils confidence in the audience about the study's methodology.

Result and Discussion

Table 1. Biogas Composition Results Before Purification

Day	Sampling date	Composition					Parameter			
		CH_4	CO_2	N_2	O_2	H_2	TS	RD	GHV	NHV
5	April 18, 2025	13.7	6.0	62.7	17.6	0.0	0.298	0.968	138	125
10	April 23, 2025	28.1	18.0	46.0	7.9	0.0	0.318	1.072	284	256
15	April 28, 2025	29.1	20.5	41.8	8.6	0.0	0.667	0.972	294	265
20	Mei 3, 2025	30.0	21.3	40.5	8.2	0.0	1.220	0.972	303	273
25	Mei 8, 2025	58.3	18.0	16.9	6.7	0.1	1.477	0.834	589	530
30	Mei 13, 2025	48.2	32.9	14.2	4.6	0.1	1.667	0.955	487	439
35	Mei 18, 2025	48.0	34.9	13.9	3.1	0.1	1.733	0.965	485	437
40	Mei 23, 2025	47.7	22.7	23.8	5.7	0.1	1.636	0.902	482	434

Table 2. Biogas Composition Results After Purification

Amine Absorbent	Packing Type	Concentration (%v/v)	Composition					Parameter			
			CH_4	CO_2	N_2	O_2	H_2	TS	RD	GHV	NHV
MEA	Pall ring	10	63.7	0.0	23.2	13.1	0.0	0.828	0.722	643	579
		20	66.0	0.0	25.0	9.0	0.0	0.808	0.707	667	600
		30	67.8	0.0	23.3	8.9	0.0	0.612	0.699	685	617
		40	73.3	0.0	18.8	7.9	0.0	0.603	0.675	740	667
		50	78.2	0.0	16.2	5.6	0.0	0.595	0.652	790	711
	Raschig ring	10	61.2	0.0	31.1	7.6	0.1	0.923	0.724	618	557
		20	64.0	0.0	28.6	7.3	0.1	0.859	0.712	647	582
		30	70.8	0.0	22.9	6.2	0.1	0.845	0.682	715	644
		40	72.9	0.0	21.2	5.8	0.1	0.620	0.673	737	663
		50	75.5	0.0	19.0	5.4	0.1	0.596	0.662	763	687
DEA	Pall ring	10	58.4	0.2	30.0	11.4	0.0	0.993	0.743	590	531
		20	63.4	0.2	26.7	9.7	0.0	0.979	0.720	640	577
		30	69.5	0.2	20.7	9.6	0.0	0.774	0.694	702	632
		40	69.8	0.2	21.0	9.0	0.0	0.695	0.692	705	635
		50	70.9	0.2	20.0	8.9	0.0	0.701	0.688	716	645
	Raschig ring	10	59.3	0.2	32.9	7.6	0.0	0.782	0.734	599	539
		20	63.5	0.2	29.2	7.1	0.0	0.731	0.716	641	577
		30	62.5	0.2	30.2	7.1	0.0	0.680	0.720	631	568
		40	69.8	0.2	21.0	9.0	0.0	0.689	0.692	705	635
		50	71.8	0.2	22.4	5.6	0.0	0.662	0.679	725	653
TEA	Pall ring	10	60.0	0.1	30.5	9.3	0.1	1.192	0.732	606	546
		20	62.8	0.1	27.3	9.7	0.1	1.284	0.721	635	571
		30	63.0	0.1	26.6	10.2	0.1	1.285	0.721	637	573
		40	65.9	0.1	21.1	12.8	0.1	1.162	0.712	666	600
		50	68.5	0.1	23.0	8.3	0.1	0.946	0.695	692	623
	Raschig ring	10	59.9	0.1	28.9	11.0	0.1	0.725	0.734	605	545
		20	60.2	0.1	26.2	13.4	0.1	0.738	0.736	608	548
		30	60.3	0.1	25.0	14.5	0.1	0.696	0.738	609	549
		40	61.9	0.1	27.1	10.8	0.1	0.685	0.726	626	563
		50	67.7	0.1	20.3	11.8	0.1	0.622	0.703	684	616
MDEA	Pall ring	10	58.8	0.0	30.4	10.8	0.0	0.642	0.736	594	535
		20	59.1	0.0	30.4	10.5	0.0	0.621	0.737	597	537
		30	60.1	0.0	29.8	10.1	0.0	0.615	0.733	607	547
		40	69.1	0.0	20.5	10.4	0.0	0.591	0.696	698	628
		50	68.6	0.0	21.0	10.4	0.0	0.459	0.698	693	624
	Raschig ring	10	58.3	0.0	33.1	8.6	0.0	0.614	0.738	589	530
		20	62.3	0.0	28.2	9.5	0.0	0.611	0.738	589	530
		30	65.1	0.0	28.5	6.4	0.0	0.608	0.723	629	567
		40	68.4	0.0	25.3	6.3	0.0	0.575	0.693	691	622
		50	70.2	0.0	24.5	5.3	0.0	0.560	0.684	709	638

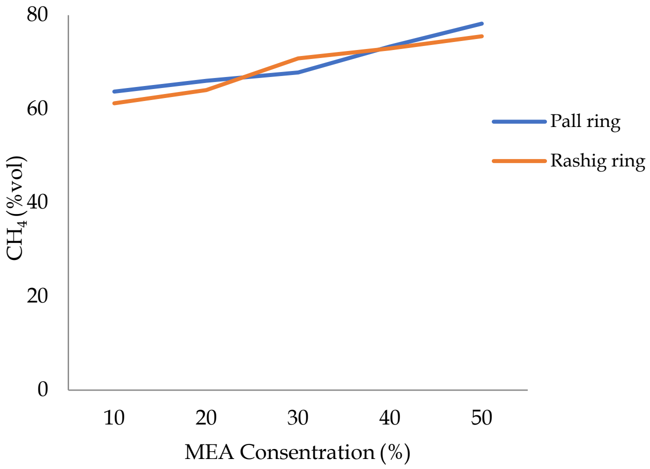


Figure 3. Effect of MEA Concentration and Packing Type on Methane Content in Upgraded Biogas

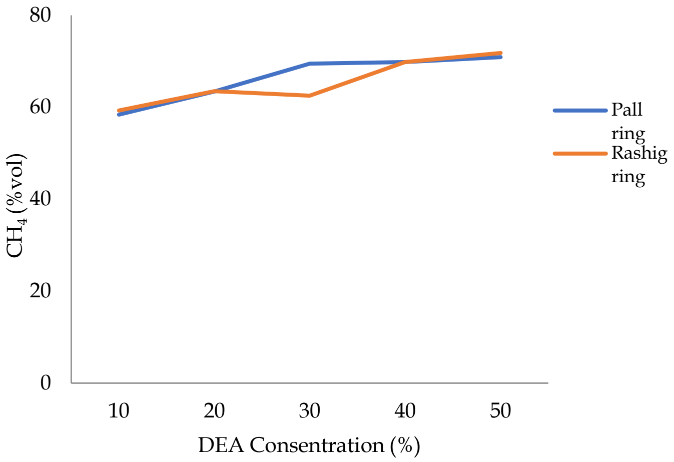


Figure 4. Effect of DEA Concentration and Packing Type on Methane Content in Upgraded Biogas

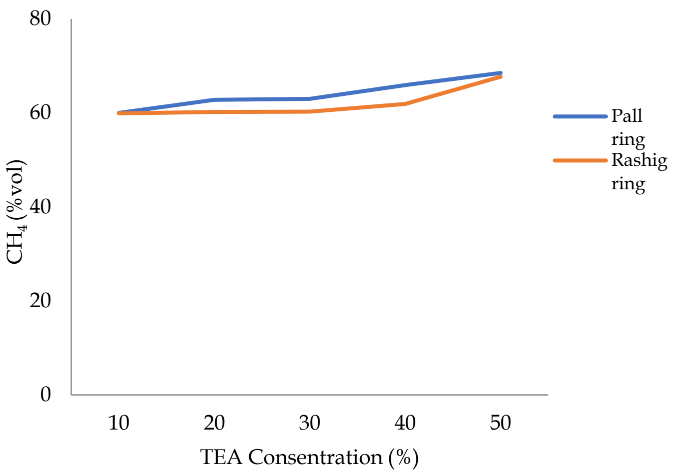


Figure 5. Effect of TEA Concentration and Packing Type on Methane Content in Upgraded Biogas

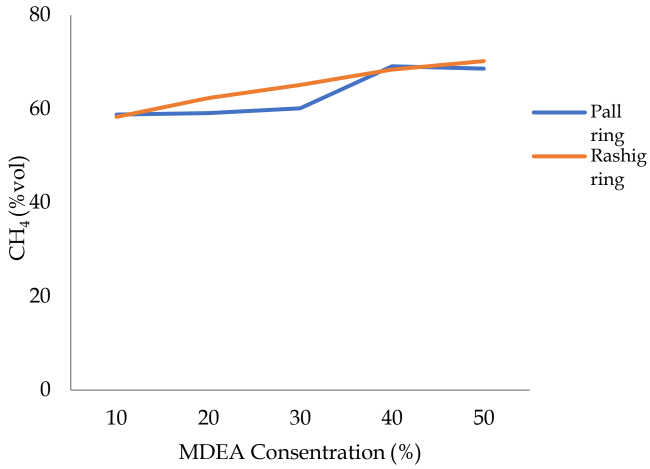


Figure 6. Effect of MDEA Concentration and Packing Type on Methane Content in Upgraded Biogas

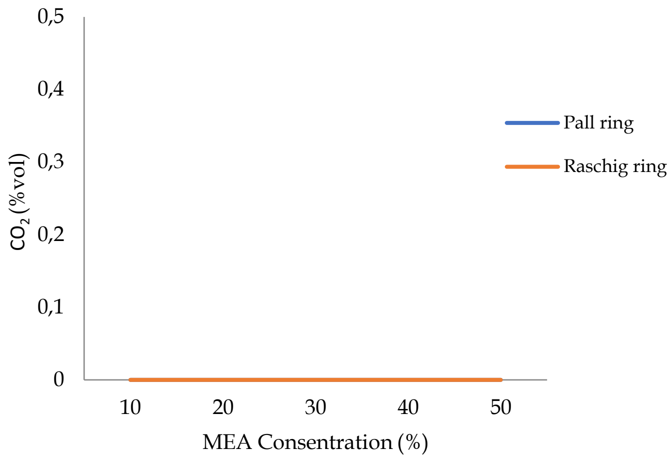


Figure 7. Effect of MEA Concentration and Packing Type on Carbon dioxide Content in Upgraded Biogas

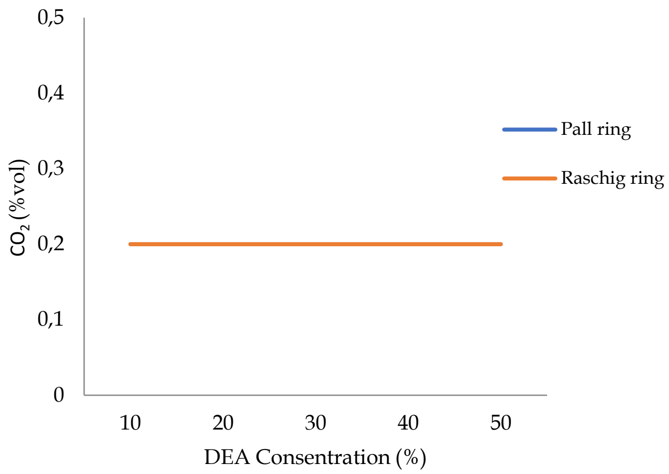


Figure 8. Effect of DEA Concentration and Packing Type on Carbon dioxide Content in Upgraded Biogas

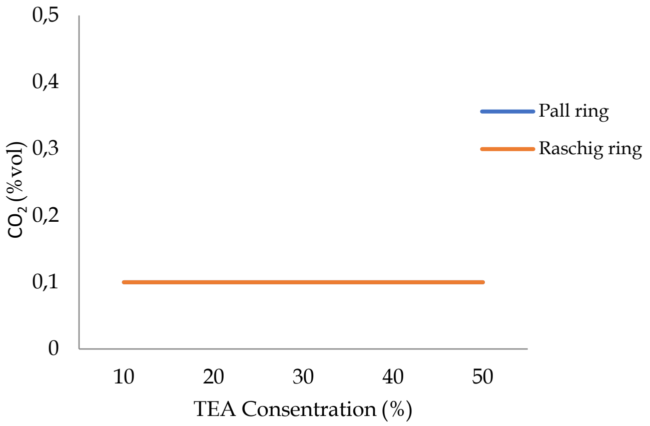


Figure 9. Effect of TEA Concentration and Packing Type on Carbon dioxide Content in Upgraded Biogas

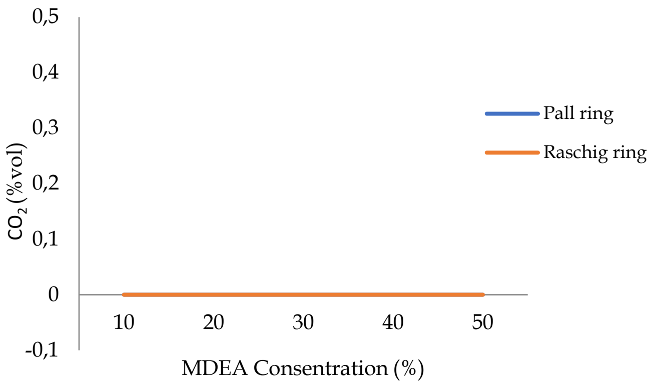


Figure 10. Effect of MDEA Concentration and Packing Type on Carbon dioxide Content in Upgraded Biogas

Biogas Purification Using a Packed Column

In this study, biogas upgrading was carried out using a vertically oriented absorber column, which was fitted with two different types of packing materials and tested with several amine solutions at varying concentrations. The main aim was to investigate how both the type of amine and its concentration affected the composition of the purified biogas, focusing primarily on methane content and carbon dioxide removal. Although more advanced techniques, such as microtube contactors or membrane-based systems, can achieve higher selectivity and greater efficiency, packed columns remain a practical and reliable choice for comparative experiments, particularly in small-scale or laboratory setups where simplicity, reproducibility, and ease of maintenance are important (Kalsum et al., 2022).

The column was operated in a countercurrent mode, with biogas entering from the bottom while the absorbent flowed downward from the top. This arrangement maximized the contact time between the gas and liquid phases, allowing the reactive sites on amine molecules to interact effectively with CO_2 and H_2S. It also helped maintain a stable liquid film on the packing surfaces, which is essential for efficient mass transfer. Raschig rings and Pall rings were selected as packing materials due to their different geometrical properties, which influence liquid distribution, turbulence generation, and overall absorption efficiency. By testing both types under identical conditions, the experiment could clearly distinguish the effects of packing geometry from those of absorbent chemistry.

To identify the most suitable time for purification, raw biogas was collected and analyzed every five days throughout the digestion cycle using a Perkin Elmer Clarus 680 Gas Chromatograph. This sampling strategy made it possible to pinpoint the day with the highest methane content. The analysis revealed that day 25 contained the peak methane concentration of 58.3%, and this batch of biogas was then used in all subsequent purification experiments to ensure consistency and representativeness. These results are broadly in line with (Tetteh et al., 2018), who reported that methane concentration typically peaks around day 20 of anaerobic digestion.

The choice of packing material had a noticeable impact on gas–liquid interactions and overall absorption performance. Pall rings, with their open structure and internal cross-supports, promoted more uniform liquid distribution, reduced pressure drop, and increased turbulence compared to Raschig rings. These features improved wetting of the gas–liquid interface and facilitated more efficient mass transfer of CO_2 and H_2S. Comparative results showed that Pall rings consistently produced higher methane content across most types of absorbents and concentrations. For instance, MEA at 50% concentration achieved 78.2% CH_4 with Pall rings, while the same conditions using Raschig rings reached only 75.5%. The enhanced geometry of Pall rings likely increased residence time and the effective contact area, leading to more efficient chemical absorption (Kadarjono et al., 2017; Ramesh & Moorthi, 2017a).

Effect of Amine Type on Biogas Purification

The type of amine used had a noticeable impact on both methane enrichment and CO_2 removal during the purification process. Among the four absorbents tested, monoethanolamine (MEA) consistently achieved the highest methane concentrations, while also removing nearly all CO_2 from the gas (Maile et al., 2017). For example, when MEA was applied at 50% concentration using Pall rings, methane reached 78.2%, whereas with Raschig rings it was slightly lower at 75.5%. The effectiveness of MEA can be attributed to its chemical nature as a primary amine, which allows it to react rapidly with CO_2, forming stable carbamate intermediates. This rapid reaction explains why MEA performs well even in systems with relatively short contact times, such as the packed column used in this study. Interestingly, across multiple trials, MEA showed consistent performance regardless of packing type, indicating its robustness for small-scale experimental setups.

Diethanolamine (DEA), a secondary amine, provided moderate results. At 50% concentration with Pall rings, methane content reached 71.8%, though a small fraction of CO_2 (0.2%) remained. The slightly lower efficiency is likely caused by the steric hindrance of the secondary amino group, which slows the reaction with CO_2 compared to MEA. Nonetheless, DEA has practical advantages, including better stability and lower volatility, making it suitable for longer-duration operations where solvent degradation could otherwise reduce effectiveness. Some minor fluctuations were observed between repeated trials, suggesting that operational factors such as flow distribution can slightly affect DEA performance.

Methyldiethanolamine (MDEA), representing the tertiary amine group, showed slightly lower performance than DEA but remained more effective than TEA. At 50% concentration, MDEA achieved 70.2% methane and complete CO_2 removal. The slower absorption rate is linked to steric hindrance around the nitrogen atom, which reduces the availability of reactive sites. While MDEA requires longer contact time to achieve full purification, its lower energy requirement for regeneration and greater chemical stability make it advantageous for larger-scale or continuous processes.

Triethanolamine (TEA) produced the lowest methane enrichment among the four amines. At 50% concentration, methane reached only 67.7%, and partial CO_2 remained at lower concentrations. The bulky molecular structure and weak basicity of TEA slow its reaction with CO_2, which limits efficiency in short-contact systems like the one used in this study. Observations also indicated that TEA's performance was more sensitive to variations in liquid distribution, suggesting its use may be more appropriate in columns with enhanced residence time or alternative packing arrangements.

Finally, the interaction between the amine and packing material contributed significantly to overall performance. Pall rings improved liquid distribution and turbulence, enhancing contact between the amine and CO_2, which likely explains why MEA and DEA showed better results with Pall rings than with Raschig rings. These findings align with previous reports that primary amines generally outperform secondary and tertiary amines in short-contact packed columns due to faster reaction kinetics and higher chemical reactivity (Maile et al., 2017; Xiao et al., 2018).

In conclusion, the results demonstrate that amine chemical structure, reaction kinetics, and column packing geometry collectively determine purification efficiency. Primary amines such as MEA are most suitable for short-contact systems, while secondary and tertiary amines may require longer residence times or optimized column designs to achieve comparable performance.

Effect of Amine Concentration on Biogas Purification

The concentration of the amine solutions had a notable effect on the efficiency of biogas purification, particularly in terms of methane enrichment and CO_2 removal (Kalsum & Hasan, 2025). In general, increasing the concentration of the absorbent led to improved purification outcomes across all four amine types tested. For example, a 10% MEA solution resulted in biogas with 61.2% methane, while increasing the concentration to 50% raised the methane content to 75.5%. Similar trends were observed for DEA, TEA, and MDEA, indicating that a higher availability of reactive amine groups allows more extensive reactions with CO_2.

This pattern can be explained by the chemical reactivity of the amines and the reaction kinetics between CO_2 and the functional groups in the molecules. Primary amines such as MEA react rapidly with CO_2 to form carbamate intermediates. By increasing the concentration of MEA, more reactive sites become available, providing a higher probability for CO_2 molecules to encounter and react with the absorbent. The countercurrent flow in the packed column, where the gas rises while the liquid flows downward, maximizes the contact time between phases, further enhancing absorption efficiency.

However, higher amine concentrations also introduce practical challenges. More concentrated solutions exhibit higher viscosity, which can hinder uniform flow and reduce effective wetting of the packing surfaces. This effect may generate micro-zones where CO_2 absorption is less efficient, particularly for tertiary amines like MDEA and TEA, which already react more slowly due to steric hindrance around the nitrogen atom. In this study, these factors likely contributed to the slightly lower performance observed for TEA and MDEA compared to MEA, even at the same concentration.

The type of packing material also played a key role in modulating the effect of concentration. Pall rings, with their open structure and internal supports, promoted better liquid distribution and turbulence, enhancing gas–liquid interaction and allowing higher CO_2 capture efficiency. As a result, even moderate concentrations of MEA or DEA produced effective purification. In contrast, Raschig rings, which offer less turbulence and less uniform liquid coverage, limited the benefits of increasing amine concentration, particularly for secondary and tertiary amines. This finding aligns with previous studies showing that the geometry and surface characteristics of packing materials can significantly influence mass transfer in packed columns (Kadarjono et al., 2017; Ramesh & Moorthi, 2017b).

Another factor affecting performance is the solubility limit of CO_2 in the amine solution. At lower concentrations, the number of available amine molecules may be insufficient to capture all CO_2, resulting in residual gas in the purified biogas. As the concentration increases, the solution capacity improves, and residual CO_2 decreases until other operational constraints, such as solution viscosity and flow limitations, become the limiting factors.

In summary, the efficiency of biogas purification in this study was determined by a complex interplay between amine concentration, chemical reactivity, packing design, and flow dynamics. For primary amines like MEA, moderate concentrations were sufficient due to fast reaction kinetics, whereas secondary and tertiary amines required higher concentrations to achieve optimal CO_2 removal. These results emphasize the importance of balancing chemical effectiveness with practical operational considerations, especially in small-scale or experimental packed column systems, and are consistent with previous reports on amine-based biogas upgrading.

The heating value of the upgraded biogas, expressed as both Gross Heating Value (GHV) and Net Heating Value (NHV), showed a strong correlation with methane concentration. As observed in Tables 1 and 2, methane enrichment following amine absorption led to a marked increase in GHV and NHV. For example, raw biogas on day 25 had a GHV of 589 kJ/mol, whereas purification with 50% MEA and Pall ring packing enhanced the GHV to 790 kJ/mol. This trend is consistent with the fundamental role of methane as the primary energy carrier in biogas, since a higher methane fraction directly translates into higher calorific value (Mekonen et al., 2023). From an application perspective, achieving GHV values above 700 kJ/mol suggests that the upgraded biogas approaches the fuel quality of bio-CNG, making it suitable for direct use in energy systems.

Another critical parameter is the total sulfur (TS) content, primarily in the form of hydrogen sulfide (H_2S). High sulfur concentrations in raw biogas can cause severe corrosion in engines and turbines, as well as contribute to SO_2 emissions during combustion (Konkol et al., 2022). In this study, the packed column absorption system reduced TS values significantly, with MEA and MDEA showing the most effective sulfur capture. For instance, TS values decreased from 1.477% in raw biogas to 0.595% after treatment with 50% MEA. These findings align with the results (Sihlangu et al., 2024), who reported substantial reductions of H_2S during amine-based upgrading processes. The removal of sulfur compounds thus improves both the environmental performance and the durability of energy conversion equipment.

Relative density (RD) of the gas mixture also served as an indicator of purification quality. Because carbon dioxide has a higher molecular weight than methane, a reduction in CO_2 content correspondingly decreases the RD of biogas. As shown in the results, RD values declined from 0.968 in raw biogas to as low as 0.652 following treatment with MEA at 50%. A lower RD reflects a lighter gas composition dominated by methane, which more closely resembles the density characteristics of natural gas (Duan et al., 2019)This parameter is particularly important for storage and transportation, as lower-density biogas can be compressed and distributed more efficiently.

Overall, the combined improvements in heating value, sulfur reduction, and relative density highlight the effectiveness of the packed column absorption system in producing upgraded biogas with fuel properties that meet international standards. These results emphasize that purification strategies should not only focus on methane enrichment but also ensure the removal of harmful sulfur compounds and optimize gas density for downstream applications.

In addition to the observed improvements, the relationship between methane enrichment and calorific value has been widely confirmed in previous research. Studies by (Kalsum et al., 2023) and (Sengur et al., 2024) demonstrated that the calorific value of upgraded biogas increases almost linearly with methane content. This supports the notion that effective CO_2 separation not only enhances fuel quality but also improves combustion efficiency and energy recovery. Furthermore, maintaining higher methane purity (>75%) is essential for achieving standards comparable to natural gas, particularly when biogas is intended for vehicle fuel or electricity generation (Francisco López et al., 2024).

Regarding sulfur removal, the reduction in total sulfur concentration in the present study aligns with global findings that emphasize the efficiency of alkanolamines in removing hydrogen sulfide. Research by (Huertas et al., n.d.) highlighted that amine-based purification can achieve more than 95% H_2S removal efficiency when operated at optimal liquid-to-gas ratios. This high performance not only minimizes corrosion risk in downstream equipment but also ensures compliance with emission regulations for SO_2. Moreover, the effectiveness of MEA and MDEA in sulfur reduction indicates their dual advantage in capturing both acidic gases (CO_2 and H_2S) due to their favorable thermodynamic selectivity.

In terms of gas density, the decrease in relative density (RD) observed after purification further confirms the efficiency of the upgrading process. Lower RD values indicate lighter gas composition with reduced CO_2 and higher CH_4 content, which facilitates gas compression and transport. Reported similar reductions in RD, showing that upgraded biogas can reach density values close to 0.65, comparable to compressed natural gas (CNG). This property enhances storage efficiency and reduces transportation costs, providing practical advantages for decentralized biogas facilities.

Overall, these findings strengthen the conclusion that the combined improvements in heating value, sulfur reduction, and relative density demonstrate the reliability of the packed column absorption method. The process not only enhances energy quality but also ensures environmental compliance and operational stability. Future optimization should focus on dynamic absorber modeling, cost reduction for solvent regeneration, and hybrid systems integrating chemical and physical absorption to achieve sustainable large-scale biogas upgrading.

Beyond the current findings, several recent studies emphasize the integration of hybrid putification system to further enchance gas quality and process sustainability. Optimization of amine-based absorption, particularly through the use of blended solvents such as MEA-MDEA of MDEA-PZ, has been reported to improved CO2 removal efficiency and reduce energy consumption during solvent regeneration (Muntaha et al., 2022). Such hybrid systems also allow continuous operation with lower energy demand for solvent regeneration. Furthermore, integrating amine solvents with ionic liquids offers a promising approach to enhance CO_2 capturre efficiency and reduce regeneration energy, providing more stable operation than single-solvent system (Yang et al., 2014). In addition, techno-economic evaluations indicate that optimization of absorber packing geometry, gas residence time, and solvent circulation rate can substantially reduce operational costs while maintaining high CO_2 and H_2S removal efficiency (Kotamreddy et al., 2020; Schellevis et al., 2021). These advancements collectively underline that the continuous refinement of amine-based technologies remains crucial for achieving both environmental compliance and commercial viability in biogas upgrading processes.

Conclusion

This study shows that the efficiency of biogas purification in a packed column depends strongly on the type of amine, its concentration, and the packing material. Among the tested absorbents, MEA combined with Pall rings produced the highest methane enrichment (up to 78.2%) while also achieving significant CO_2 and sulfur removal. DEA and MDEA showed reasonable effectiveness, though with slower kinetics and potential issues at higher concentrations, while TEA proved least effective due to steric hindrance. Although higher amine concentrations improved purification, they also introduced challenges such as increased viscosity, solvent degradation, and potential corrosion. For practical applications, concentrations between 10–30% appear to offer the best balance between efficiency and cost. In addition, the choice of packing material was shown to play a critical role, with Pall rings outperforming Raschig rings by enhancing gas–liquid contact and resulting in higher methane yields. These findings highlight MEA with Pall rings as a robust option for small- to medium-scale biogas upgrading. Future studies should focus on long-term system performance, continuous operation, solvent regeneration, and the use of blended or hybrid absorbents to further improve efficiency and reduce overall costs, ensuring both economic feasibility and environmental sustainability.

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Author Contributions
The authors in this study have made maximum contributions in their respective roles according to their areas of expertise.

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Conflicts of Interest
The authors declare that they have no conflict of interest.

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