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Modelling and Simulation of Benzene Production from Biogas using Zeolite Catalyst

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Abstract. Biogas has emerged as a promising sustainable resource due to its abundant methane content, which can be converted into valuable chemicals. This study investigates benzene production from biogas using a Mo/HZSM-5 catalyst in a packed bed reactor, aiming to provide an alternative feedstock for the petrochemical industry. A process model was developed using DWSim software to simulate and optimize the benzene production process. The simulation demonstrated that with a biogas feed of 7.5 kiloton/year, benzene production reached 1.2 kiloton/year, yielding 0.16% with a reactor conversion efficiency of 43.89%. Key process parameters were analyzed to ensure feasibility and sustainability, including energy consumption (0.6 MW heating, 0.8 MW cooling, and 0.2 MW electricity). The separation of CO_2 and light gases was achieved using a mixed-matrix membrane, followed by benzene purification through absorption and distillation, achieving a final purity of over 98%-mol. Compared to conventional benzene production methods from shale gas or LPG, this process offers lower energy requirements and improved yield, making it a viable approach for industrial implementation. This study not only presents a detailed simulation but also highlights the potential of biogas as a renewable feedstock for benzene synthesis, contributing to sustainable chemical production. Further techno-economic analysis and experimental validation are recommended to assess scalability and commercial feasibility.

Keywords: Benzene, Biogas, DWSim, Simulation, Zeolite

INTRODUCTION

The production of benzene is of great importance in the chemical industry due to its versatile applications in producing various chemicals and materials. The traditional benzene production method involves using petroleum-based feedstocks through catalytic reforming or steam cracking. However, the increasing demand for sustainable and renewable alternatives has led to the exploration of new routes for producing benzene.

Biogas, a renewable energy source produced from organic waste materials, has been identified as a promising feedstock for benzene production. Mainly, biogas consists of light gases such as methane (Calbry-Muzyka *et al*., 2022) and has a high calorific value (Bharathiraja *et al*., 2018). Recent

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advancements in biogas conversion pathways highlight its feasibility for synthesizing high-value chemicals such as benzene (Deng, *et al*., 2024). Nonoxidative aromatization of biogas is a promising route for benzene production, where a nonoxidative agent converts methane into higher hydrocarbons, which are subsequently converted to benzene. Although this reaction is thermodynamically unfavorable and could produce unfavorable byproducts (Spivey and Hutchings, 2014), the use of specific catalysts, such as Mo/HZSM-5 catalyst, could increase the selectivity for the aromatic products (Xu *et al*., 1994).

While several studies have explored benzene production using petroleum-based feedstocks or other raw materials such as natural gas and methanol, limited attention has been given to sustainable alternatives like biogas. For instance, benzene production from natural gas uses Aspen Plus (Chanenchuk and Evans, 2015). Another study proposed methanol as a reaction agent (Niziolek et al., 2016). Regulatory frameworks worldwide continue to evolve to support biogas utilization for industrial-scale applications (Abanades *et al*., 2022). However, most prior research has focused on reaction mechanisms and catalysts without developing comprehensive simulation studies to model and optimize the production process. This study aims to bridge these gaps by utilizing DWSim software to develop a detailed simulation model for benzene production from biogas, specifically in a Mo/HZSM-5 zeolite catalyst system (Hu *et al*., 2024). Such simulations are critical for accurately predicting process performance, optimizing energy consumption, and ensuring the feasibility of scaling up to industrial applications. Minimizing energy consumption and maximizing material recovery are crucial aspects of sustainable chemical production. Gasification and methane aromatization have emerged as promising conversion routes, ensuring environmental viability while maintaining process efficiency (Lackner *et al*., 2024).

MATERIALS AND METHODS

Process Overview

This scientific study aimed to develop a process capable of converting biogas into 1 kiloton of benzene annually. Furthermore, particular emphasis was placed on ensuring the plant's design and operations adhered to environmentally friendly practices, with strict compliance with local regulations. To align with these objectives, minimizing energy consumption and maximizing material recovery and recycling were essential without compromising the process's economic viability—finally, the plant design needed to prioritize controllability and safety during its operations.

The gas phase dehydrocyclization reactions in this process are shown in Eq. (1) and Eq. (2).

$$6CH_4 \Leftrightarrow C_6H_6 + 9H_2 \quad (1)$$

$$CH_4 \Leftrightarrow C + 2H_2 \quad (2)$$

The dehydrocyclization reaction is unfavorable thermodynamically (ΔG_r^o = +433 kJ mol^{-1}), so benzene formation is significant at higher temperatures (Spivey and Hutchings, 2014). This could be achieved with an H-ZSM-5 catalyst to speed the reaction up to 0.01 WHSV (Van Der Mynsbrugge *et al*., 2012).

The process was divided into two main parts: dehydrocyclization, which converts biogas into benzene, and separation of benzene with light gases. Dehydrocyclization

converts methane from biogas into mainly benzene products using a zeolite catalyst. Then, the light gases (methane, nitrogen, hydrogen, and ethylene) are separated with benzene using absorption, and 99.9% of the light gases are recycled and mixed with the feed biogas. The flow diagram of this process is shown in Figure 1.

Reaction Kinetics Model

The catalyst used in this study's dehydrocyclization is Mo/HZSM-5 catalyst. This catalyst reportedly has a high mass of benzene formed and converted methane (Shaw *et al.*, 2006). The kinetics, properties, and operating conditions are inputted into the simulation based on a study reported on the Mo/HZSM-5 catalyst (Iliuta *et al.*, 2003; Yao *et al.*, 2008). The dehydrocyclization kinetic models are expressed in Eq. (3) and Eq. (4).

$$r_1 = \frac{k_1 . f_{CH_4}^6 . (1-\beta_1)}{\left(1+K_{CH_4} . f_{CH_4}+K_{H_2} . f_{H_2}+K_{C_6H_6} . f_{C_6H_6}\right)^6} \quad (3)$$

$$r_2 = \frac{k_2 . f_{CH_4} . (1-\beta_2)}{1+K_{CH_4} . f_{CH_4}+K_{H_2} . f_{H_2}+K_{C_6H_6} . f_{C_6H_6}} \quad (4)$$

Where k_1 and k_2 are the rate constant of reaction (1) and (2), respectively, f_i is the fugacity of component i, K_i is the adsorption equilibrium constant of component i, and β_i is the departure amount of the balance. All kinetic parameters followed by the data given by respective papers are

$$k_1 = 8.1283 \times 10^{-3} . \exp\left(-\frac{2.0909 \times 10^5}{R}\left(\frac{1}{T} - \frac{1}{943.15}\right)\right)$$

$$k_2 = 2.3252 \times 10^{-3} . \exp\left(-\frac{1.2096 \times 10^5}{R}\left(\frac{1}{T} - \frac{1}{943.15}\right)\right)$$

$$K_{CH_4} = \exp\left(-1.1963 - 1.3209 \times 10^2\left(\frac{1}{T} - \frac{1}{943.15}\right)\right)$$

$$K_{H_2} = \exp\left(1.6736 + 1.5796 \times 10^3\left(\frac{1}{T} - \frac{1}{943.15}\right)\right)$$

$$K_{C_6H_6} = \exp\left(-9.09 + 1.1770 \times 10^5\left(\frac{1}{T} - \frac{1}{943.15}\right)\right)$$

Separation Process

Purification of Biogas

The biogas is known to have high CO_2 content. In the dehydrocyclization process, CO_2 impacted a higher yield of benzene formed (Shaw *et al.*, 2006). However, it can reduce the purity of benzene in the absorption process. The CO_2 must be separated before light gases and benzene

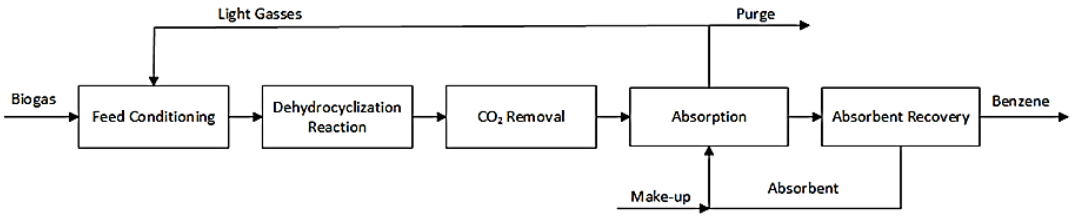

Fig 1: Process Block Flow Diagram of Benzene Production from Biogas

are separated to improve benzene purity.

The separation of CO_2 from biogas can be done with a membrane. A mixed-matrix membrane (MMM) comprising polymeric and inorganic membranes was reported to incorporate both the advantages of polymeric and inorganic membranes (Chawla *et al*., 2020). The MMM used in this study is Pebax 1657 with 5-30% 4-A Zeolite, which gives high permeability for CO_2 (Surya Murali *et al*., 2014). The separated CO_2 can be injected into a well or utilized for other purposes.

Separation of Benzene from Light Gasses

In this study, the benzene separation process is modeled using an absorption tower, where mass transfer is simulated through the equilibrium stage model embedded in DWSim. The absorbent, 2,2,3,3-tetramethyl hexane, is introduced at the top of the column to selectively absorb benzene from the vapor stream, ensuring effective separation. The equilibrium stage model assumes ideal stage behavior, accounting for mass transfer and phase equilibria between each stage's liquid and vapor phases.

The simulation includes energy balances within the column for heat transport, maintaining stable operation through controlled temperature profiles. The condenser at the top and the reboiler at the bottom ensure heat exchange to sustain the required separation efficiency and purity. Parameters such as the absorbent flow rate (optimized to 35 kg/h based on simulation studies) and reflux ratio (set at 1.5 in the distillation column) are determined to achieve a benzene purity exceeding 98%-mol. These values were validated through sensitivity analysis to optimize separation efficiency while minimizing energy and material costs.

DWSim

DWSim is an open-source chemical process simulator that has gained increasing traction in academic research and industrial applications. Its popularity is reflected in the growing number of published papers utilizing DWSim for petrochemical and process simulations (Andreasen, 2022; Cubides-Román *et al*., 2018). The flexibility of DWSim in incorporating user-defined unit operations and Python scripting has contributed to its widespread adoption in modeling complex chemical processes.

The reported discrepancy of less than 5% between DWSim and other commercial process simulators is based on comparative studies (Tangsriwong *et al*., 2020). This comparison generally applies to thermodynamic property estimations and process modeling accuracy, though specific variations may exist depending on the complexity of unit operations. In this study, the accuracy of DWSim was validated against published benzene production simulation data to ensure reliable predictions.

One of the advantages of using DWSim is that the user is able to program unit operations using Python script. Python language is easy to understand, open-source, and has recently been widely used in academia and industry. This makes DWSim versatile for solving complex process problems.

RESULTS AND DISCUSSION

This benzene production process model is modeled and simulated in DWSim version 8.5.1. The flowsheet of this process is shown in Figure 2. Notable process streams are shown in Table 1 and Table 2.

Biogas feed, which consists of methane, CO_2, oxygen, and a little bit of H_2S (<10 ppm),

is compressed to 1.5 bar and then mixed with the recycled methane (Chozhavendhan *et al.*, 2020). The mixed biogas is heated to 700°C, which activates the zeolite catalyst. Heated feed is fed into a 24 m^3 heterogenous packed bed reactor isothermally, which converts methane into benzene with an overall conversion of 43.89%. The kinetic parameters used in the simulation follow experimentally verified data reported by Iliuta *et al.* (2003) and Yao *et al.* (2008). These references provide a strong foundation for the accuracy of the reaction kinetics implemented in DWSim.

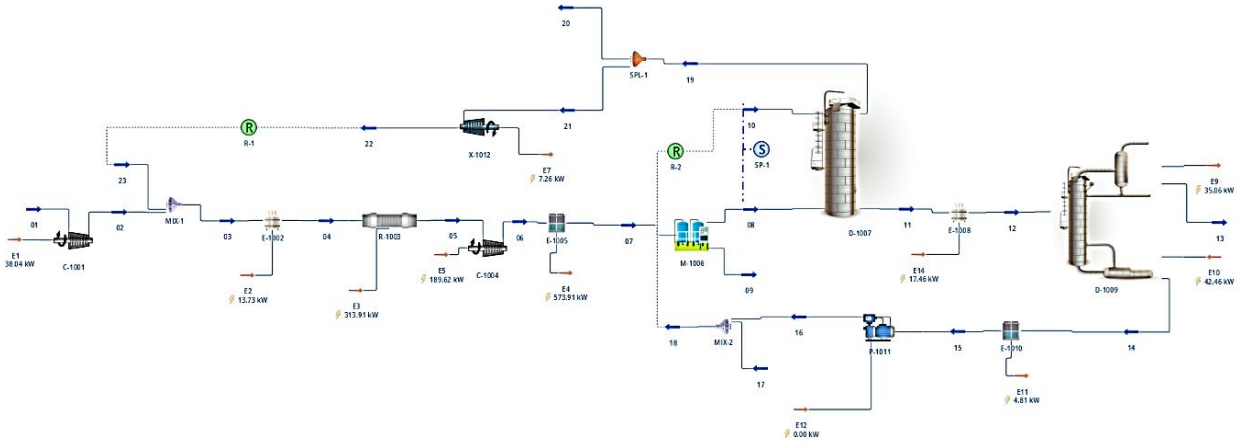

Fig 2: DWSIM Flowsheet of Benzene Production from Biogas

Table 1. List of streams and their properties (1)

Object (unit)	01	04	05	07	08	09	10
Temperature (C)	711.75	700.00	700.00	43.51	43.51	43.51	24.09
Pressure (bar)	1.01	1.40	1.37	5.00	5.00	5.00	5.15
Mass Flow (kg/h)	870.63	970.63	970.63	970.63	352.58	618.05	35.26
Molar Flow (kmol/h)	31.16	36.07	43.64	43.64	13.33	30.30	0.25
Vapor Fraction	1	1	1	1	0.88728	1	0
Molar Fraction							
Benzene	0.00000	0.00768	0.04967	0.04967	0.16241	0.00007	0.00009
CO_2	0.42425	0.36655	0.30303	0.30303	0.00099	0.43591	0.00000
Methane	0.57369	0.62312	0.25516	0.25516	0.83426	0.00037	0.00000
Nitrogen	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000
Hydrogen	0.00000	0.00000	0.38995	0.38995	0.00000	0.56152	0.00000
2,2,3,3-Tetramethylhe-xane	0.00000	0.00086	0.00071	0.00071	0.00233	0.00000	0.99991
Carbon	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000
Oxygen	0.00201	0.00173	0.00143	0.00143	0.00000	0.00206	0.00000
H_2S	0.00006	0.00005	0.00004	0.00004	0.00000	0.00006	0.00000

Table 2. List of streams and their properties (2)

Object (unit)	11	13	14	17	19	20	22
Temperature (C)	45.03	139.75	236.00	25.00	56.98	56.99	7.04
Pressure (bar)	5.05	4.80	5.00	5.15	5.00	5.00	1.50
Mass Flow (kg/h)	172.66	140.29	32.38	2.88	215.18	2.15	213.03
Molar Flow (kmol/h)	2.05	1.82	0.23	0.02	11.53	0.12	11.42
Vapor Fraction	0	0.99947	0.00195	0	1	1	0.98893
Molar Fraction							
Benzene	0.87405	0.98345	0.00010	0.00000	0.03261	0.03261	0.03261
CO_2	0.00008	0.00010	0.00000	0.00000	0.00113	0.00113	0.00113
Methane	0.01461	0.01644	0.00000	0.00000	0.96182	0.96182	0.96182
Nitrogen	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000
Hydrogen	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000
2,2,3,3-Tetramethylhe-xane	0.11125	0.00010	0.99990	1.00000	0.00443	0.00443	0.00443
Carbon	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000
Oxygen	0.00000	0.00000	0.00000	0.00000	0.00001	0.00001	0.00001
H_2S	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000

Reactor products are compressed to 5.1 bar to accommodate the membrane operating pressure. The MMM used in this study, Pebax 1657 with 5-30% 4-A Zeolite, is modeled in the simulation using the compound separator unit model in DWSim. This unit model replicates the membrane's high permeability for CO_2 by setting the separation efficiency parameters based on experimental data reported by Surya Murali *et al.* (2014). The compound separator ensures that 99.9% of CO2 is removed from the biogas, minimizing its impact on the yield and purity of benzene. This modeling approach accurately predicts the membrane's performance within the process simulation. This is modeled using the compound separator unit model in DWSim. CO_2 and light gases retentate are injected into the well.

Permeate is then fed into a 6-stage absorption column. The absorbent for separating benzene is modeled as 2,2,3,3-tetramethyl hexane in DWSim, fed at the top of the column. The mixture of absorbent and benzene can be obtained in the liquid product, while light gases can be obtained in the vapor product. 1% of the absorber vapor product is converted into CO_2 by combustion and then injected into the well. The rest is conditioned by releasing the stream pressure before being recycled and mixed with the biogas feed.

The absorbent flow rate is studied to know the effect of the flow rate on the benzene flow rate obtained in the liquid flow rate. The graph is shown in Figure 3.

Based on Figure 3, the absorbent mass flow rate above 40 kg/h would become less

significant to the absorbed benzene. This study chose the mass flow of 35 kg/h absorbent to reduce the cost of absorbent make-up due to the usage of the absorbent.

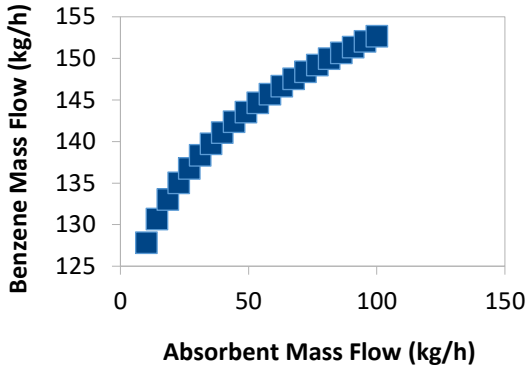

Fig 3: Effect of absorbent flow on the absorbed benzene mass flow

The liquid absorber product is then fed to the distillation column to separate benzene from other components, including absorbent. Benzene purification is set to more than 98%-mol benzene to accommodate market specifications. The distillation column is first modeled using a shortcut column to obtain the initial specification of the column. The specification of the distillation column obtained from the shortcut column is shown in Table 3.

Table 3. Absorbent recovery column specification (D-1009)

Parameters	Value
Number of stages	15.00
Feed stage	7.00
External reflux ratio	1.50
Condenser pressure (bar)	4.80
Condenser temperature (C)	139.75
Reboiler pressure (bar)	5.00
Reboiler temperature (C)	236.00

Based on the obtained simulation, to produce 1.2 kiloton/year benzene, the biogas feed needed is 7.5 kiloton/year. The yield of this production is 0.16 because, besides methane, CO_2 is the second most abundant component in biogas (more than 40%). So, instead of methane conversion to benzene, CO_2 effluent is the biggest flowrate in this flowsheet.

The results obtained were compared with previous studies on benzene production, like shale gas or liquefied petroleum gas; producing benzene from biogas gives a better overall yield to ensure the reliability of the simulation. The comparison of benzene production simulation from various sources is shown in Table 4.

Table 4. Benzene production from various sources

CH_4 sources	Energy required (kW)	Feed molar flow (kmol/hr)	Reaction	Benzene yield	References
Biogas	0.6 MW (heating), 0.8 MW (cooling), 0.2 MW (electricity)	31.16	Methane dehydrocyclization	0.16	This study
Shale gas	31.08 MW (heating) and 11.73 MW (cooling)	1000	Direct methane aromatization	0.10	Pérez- Uresti *et al.* (2017)
Natural gas	385.98 MW	9400	Methane dehydrocyclization	0.23	Consoli *et al.* (2013)
Liquefied petroleum gas	3.78 MW (heating) and 3.28 MW (cooling)	100	Low-chain alkanes aromatization	0.04	Ubam *et al.* (2019)
Hydro-processed Micro-algae	0.98 MW (Heating) and 1.28 MW (Cooling)	100	Low-chain alkanes Aromatization	0.11	Corbetta *et al.* (2014)

Benzene production from biogas shows a promising result. Both studies on benzene production from biogas, Corbetta *et al.* (2014) and this study showed higher benzene yield with lower energy requirements. Compared to Ubam *et al.* (2019), the energy required is 5 times higher with raw material, which is only 3 times higher than this study. A similar result of higher energy requirement is shown by Perez-uresti *et al.* (2017), with more than 50 times the energy required. The higher yield of this study results from a more direct reaction route than aromatization. However, while using the same reaction of Methane dehydrocyclization, natural gas showed a better yield than biogas. This phenomenon results from higher CO_2 in biogas that can inhibit the reaction. CO_2 removal by any means needs to be done to increase yield and improve the overall quality of biogas. The removal process can be integrated into biogas production or installed as a pretreatment process in benzene production.

Compared to conventional benzene production methods from natural gas, this simulation shows comparable energy efficiency while utilizing biogas as a sustainable feedstock. This suggests potential for industrial implementation pending further experimental validation. A techno-economic analysis needs to be performed to evaluate the economics of this plant. Comprehensive sensitivity analysis is recommended as biogas prices vary depending on the biomass sources, biomass conversion process and technology, and the methane yield from the conversion process.

CONCLUSIONS

This study simulated the benzene production with biogas as its raw material through methane dehydrocyclization. The reaction converts methane in biogas to benzene with the help of Mo/HZSM-5 catalyst, a common catalyst used in benzene production. The process takes place in a heterogenous packed bed reactor. The simulation with DWSim showed a promising result of 0.16% benzene yield. From the simulation data, 1.2 kilotons/year of benzene was needed to produce 7.5 kilotons/year of biogas. The energy requirement of this process is 0.6 MW heating duty, 0.8 MW cooling duty, and 0.2 MW of electricity. The energy requirement is considered low compared to other studies. The result obtained from this research is limited to the simulation data of DWSim; other calculations or comparisons using other types of process simulation might be needed.

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NOMENCLATURE

ΔG_r°	:	Reaction Gibbs energy
k_n	:	Reaction rate constant
f_i	:	Fugacity
K_i	:	Adsorption constant
β_i	:	Departure amount
r	:	Reaction Rate
R	:	Gas constant
T	:	Temperature

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