



Transforming sugarcane bagasse into zeolitic material: a sustainable approach to wastewater treatment

Nuhu AA ^{1*}, Garba ZN ¹; Ibrahim H ¹; Abdulrazak S ^{1,2}

¹ Department of Chemistry, Faculty of Physical Science, Ahmadu Bello University, Zaria, Nigeria

² Department of Veterinary Physiology, Faculty of Veterinary Medicine, Ahmadu Bello University, Zaria, Nigeria

*Corresponding author E-mail: aanuhu@yahoo.com

Abstract

Sugarcane bagasse, an abundant agricultural byproduct rich in silicates and cellulose, continues to be underutilized, making a significant contribution to the ever-growing global solid waste predicament. This study delves into the intricate process of producing and enhancing zeolite material derived from economically viable sugarcane bagasse by employing hydrothermal treatment. It meticulously explores four pivotal process variables: particle size (90-200 μm), reagent (0.5 M NaOH+1.5 M NaCl) ratio (0.5-1), contact time (40-72 hr), and temperature (70-100°C), by utilizing 2⁴ full factorial design to optimize synthesis conditions. The investigation carefully delineates the nuanced impacts of these variables on the resulting zeolite porosity. After 16 experimental runs, the study identified the optimal synthesis conditions as follows: a particle size of 90 μm , a reagent ratio of 1, a contact time of 72 hr, and a temperature of 100°C. The fit statistics that signified the adequacy and significance of the developed model are $R^2 = 0.9965$, Adjusted $R^2 = 0.9827$, Predicted $R^2 = 0.9018$; Adeq Precision = 26.6195; Std. Dev. = 1.69 and C.V = 2.72%. Furthermore, the synthesized zeolite exhibited potentially a heightened adsorption capability due to its amplified porosity. This opens up promising avenues for wastewater treatment, offering effective solutions to a myriad of environmental concerns. This approach not only addresses the pressing issue of waste management but also underscores the potential of transforming waste into valuable resources for sustainable development.

Keywords: Wastewater; Hydrothermal Method; Porosity; Sugarcane Bagasse; Optimization.

1. Introduction

Zeolites, along with their structural variations, are part of an extensive category of porous materials known as ceramic molecular sieves and are exceptionally stable hydrothermally [1]. These materials possess inherent sieve-like properties that find widespread application in catalysis, ion-exchange, water purification, membrane separation processes, solar cell technology, drug delivery, and antimicrobial activities [2], [3].

Conventional zeolites are microporous, hydrated aluminosilicates composed of alkali or alkaline earth metals, exhibiting a well-defined porous structure. Their frameworks consist of $[\text{SiO}_4]^{4-}$ and $[\text{AlO}_4]^{5-}$ tetrahedra [4], which are interconnected to form cages linked by pore openings of precise dimensions. They are typically represented by the general formula $\text{M}_x/n[\text{AlO}_2]_x(\text{SiO}_2)_y \cdot z\text{H}_2\text{O}$, where M denotes the compensating cation, usually from groups I or II, with a valence of n. The properties of zeolites are significantly influenced by the Si/Al ratio of the framework, while the quantity of cations regulates surface properties and affects adsorption, catalytic, and ion-exchange characteristics [5]. Extensive research has been conducted on zeolites, revealing that the Si/Al ratio plays a crucial role in determining the type and final crystal structure of the zeolites produced [6-11].

Sugarcane bagasse encompass residual byproducts from the production and processing of sugarcane, often containing materials potentially beneficial to human activities but frequently considered economically unviable due to the high costs associated with their collection, transportation and processing for beneficial applications; it is widely acknowledged as a significant component of total waste globally [12]. Sugarcane bagasse, categorized as an agricultural waste [13], holds promise for various applications. With its considerable cellulose and silicate content, it shows potential as an adsorbent material [14]. Furthermore, its fibrous nature makes it suitable for applications in the textile and civil engineering sectors, albeit requiring specific treatments before utilization. Additionally, bagasse can strengthen composite materials, enabling the development of innovative products [15].

A full factorial design facilitated the modeling and optimization of process conditions in this study. This methodology serves as a robust tool for optimizing zeolite synthesis via hydrothermal method and predicting the adsorptive capacity of the optimized zeolite [16]. Such an approach holds promise for enhancing the efficiency and cost-effectiveness of zeolite production from inexpensive sugarcane bagasse as an adsorbent, thereby making significant advancements in water treatment and environmental remediation. The objective of this study was to synthesize and optimize zeolitic material derived from low-cost sugarcane bagasse via the hydrothermal method, employing four process variables, at two levels.



2. Materials and methods

2.1. Sampling and pre-treatment process

Sugarcane bagasse was acquired from local markets situated in Kaduna State, located in the North West region of Nigeria. A washing procedure employing distilled water was conducted to remove any extraneous debris, floating residues, and soluble impurities. Following this, the bagasse underwent drying at 100°C, followed by a two-stage grinding process, ensuring thorough homogenization. Subsequent to grinding, sieving was carried out to remove larger particles, with the sieved material being earmarked for zeolite production.

2.2 Hydrothermal method

In Hydrothermal method, as delineated by Shah et al. [14], 200 µm mesh size sieved material sourced from Sugarcane Bagasse (SB) was suspended in a solution comprising 0.5 M NaOH and 1.5 M NaCl (at a ratio of 1:10, w/v) in a round bottom flask. Subsequently, hydrothermal treatment was conducted by refluxing the resultant mixture with intermittent stirring at 100°C for a duration of 72 hr. Following cooling of the reaction vessel to room temperature, the suspension underwent filtration. The residual material was then subjected to repeated washing with distilled water to eliminate excess sodium hydroxide and sodium chloride, followed by drying at 100°C in an oven for 6 hr. The produced zeolite was stored in clean, airtight plastic container until further use.

2.3. Optimization of zeolite production

An exhaustive approach employing a full factorial design, incorporating four factors at two levels, was employed to optimize conditions for zeolite production via hydrothermal method. These factors include particle size (90-200 µm), reagent ratio (0.5-1), contact time (40-72 hr), and temperature (70-100°C). Porosity was selected as a crucial indicator of adsorption efficacy based on its significance, with its determination executed using the stochastic method elucidated by Matko [17].

2.4. Statistical design of experiment

The optimization process was facilitated through a full factorial design incorporating four factors and two levels. Thorough analysis of all experimental data was conducted utilizing diverse statistical metrics such as ANOVA, R², and standard deviation. Subsequently, the developed mathematical model was utilized to construct response surface plots, facilitating the prediction of relationships between independent and dependent parameters. Importantly, all statistical analyses were performed using the Stat-Ease Design Expert 13.0 software package.

3. Results and discussion

The concept of optimization involves utilizing specific methodologies to ascertain the most economically viable and efficient solution for a given problem or the design of a particular process. The importance of achieving optimal operating conditions cannot be overstated as it is crucial in preventing the wastage of raw materials, energy, and time. Attaining optimum conditions often necessitates the thorough examination of multiple factors across various potential scenarios.

In the present study, the optimization process was facilitated through the utilization of Design Expert version 13.0. A 2⁴ full factorial design methodology was employed to enhance the synthesis of zeolites derived from low-cost sugarcane bagasse using the hydrothermal method. Sixteen experimental runs were conducted, and the porosity (%) was assessed using a stochastic method, as detailed by Matko [17], which served as the response (Table 1).

Table 1: Experimental Design and Response for Zeolite Produced from Sugarcane Bagasse Using Hydrothermal Method

| Std | Run | Factor 1 A:PARTICLE SIZE µm | Factor 2 B:REAGENT RATIO | Factor 3 C:TEMPERATURE °C | Factor 4 D:CONTACT TIME Hr | Response 1 POROSITY % |
|-----|-----|-----------------------------------|-----------------------------|---------------------------------|----------------------------------|-----------------------------|
| 13 | 1 | 90 | 0.5 | 100 | 72 | 57.9 |
| 9 | 2 | 90 | 0.5 | 70 | 72 | 39.3 |
| 15 | 3 | 90 | 1 | 100 | 72 | 79.8 |
| 10 | 4 | 200 | 0.5 | 70 | 72 | 62.4 |
| 8 | 5 | 200 | 1 | 100 | 40 | 77.6 |
| 1 | 6 | 90 | 0.5 | 70 | 40 | 80 |
| 5 | 7 | 90 | 0.5 | 100 | 40 | 43.3 |
| 14 | 8 | 200 | 0.5 | 100 | 72 | 55.3 |
| 3 | 9 | 90 | 1 | 70 | 40 | 70.5 |
| 16 | 10 | 200 | 1 | 100 | 72 | 55.7 |
| 11 | 11 | 90 | 1 | 70 | 72 | 80.5 |
| 6 | 12 | 200 | 0.5 | 100 | 40 | 59.6 |
| 4 | 13 | 200 | 1 | 70 | 40 | 55.9 |
| 7 | 14 | 90 | 1 | 100 | 40 | 49.3 |
| 2 | 15 | 200 | 0.5 | 70 | 40 | 63.2 |
| 12 | 16 | 200 | 1 | 70 | 72 | 64 |

3.1. Model suitability assessment

The efficacy of the developed model was evaluated through the creation of multiple analytical plots. A plot comparing predicted data against actual data for the hydrothermal method was generated to elucidate the correlation between predicted and observed values, thereby affirming the validity of the model (Fig. 1).

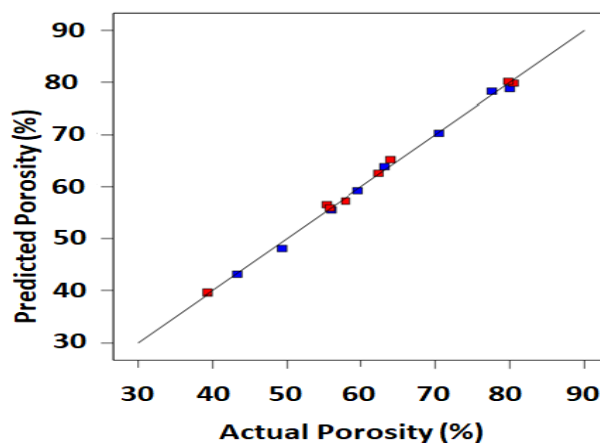


Fig. 1: Actual Value vs. Predicted Value Plot of Response for Zeolite Produced from Sugarcane Bagasse Using Hydrothermal Method.

The linear model, derived from the full factorial design, was adjudged as optimal for delineating the influences of particle size, reagent ratio, contact time, and temperature on the hydrothermal synthesis of zeolites. The formulated equation, expressed in terms of coded factors pertinent to the hydrothermal approach utilized in this investigation, is presented (Equation 1).

Equation 1: Final Equation in Terms of Coded Factors for Zeolite Produced from sugarcane Bagasse using Hydrothermal Method

$$Y = 62.1437 + 4.51875 * B - 2.33125 * C - 2.93125 * AB + 2.66875 * AC - 2.08125 * AD + 1.26875 * BC + 3.61875 * BD + 2.64375 * CD + 1.74375 * ABC - 4.70625 * ABD - 6.83125 * ACD - 3.83125 * BCD.$$

Where Y = Response (% Porosity), A, B, C and D are the Particle size (μm), Reagent ratio, Temperature ($^{\circ}\text{C}$) and Contact time (hr) respectively.

3.2. Model fit summary and evaluation

Table 2 elaborates on the fit summary, encompassing the sequential model sum of squares and model summary statistics pertinent to the hydrothermal method employed for zeolite synthesis. Upon meticulous examination of the sequential model sum of squares, the term was deemed significant, thus affirming the appropriateness of the full factorial model for the hydrothermal method utilized in this study. The significance and adequacy of the developed models were assessed through analysis of variance.

The results unveiled that both the interactive (2FI) and linear models exhibited R^2 values approaching unity (0.9965), indicative of a profound correlation. Additionally, the adjusted R^2 value (0.9827) and predicted R^2 values (0.9018) demonstrated satisfactory agreement, with a minimal disparity of less than 0.2. Moreover, notably lower p-values (below 0.05) underscored the significance of the model.

Table 2: ANOVA for Factorial Model of Zeolite Produced from Sugarcane Shaft Using Hydrothermal Method

| Source | Sum of Squares | Df | Mean Square | F-value | p-value | |
|-----------------|----------------|----|-------------|---------|---------|-------------|
| Model | 2466.05 | 12 | 205.50 | 72.13 | 0.0024 | Significant |
| B-REAGENT RATIO | 326.71 | 1 | 326.71 | 114.68 | 0.0017 | |
| C-TEMPERATURE | 86.96 | 1 | 86.96 | 30.52 | 0.0117 | |
| AB | 137.48 | 1 | 137.48 | 48.25 | 0.0061 | |
| AC | 113.96 | 1 | 113.96 | 40.00 | 0.0080 | |
| AD | 69.31 | 1 | 69.31 | 24.33 | 0.0160 | |
| BC | 25.76 | 1 | 25.76 | 9.04 | 0.0574 | |
| BD | 209.53 | 1 | 209.53 | 73.54 | 0.0033 | |
| CD | 111.83 | 1 | 111.83 | 39.25 | 0.0082 | |
| ABC | 48.65 | 1 | 48.65 | 17.08 | 0.0257 | |
| ABD | 354.38 | 1 | 354.38 | 124.39 | 0.0015 | |
| ACD | 746.66 | 1 | 746.66 | 262.08 | 0.0005 | |
| BCD | 234.86 | 1 | 234.86 | 82.44 | 0.0028 | |
| Residual | 8.55 | 3 | 2.85 | | | |
| Cor Total | 2474.60 | 15 | | | | |

* $R^2 = 0.9965$; Adjusted $R^2 = 0.9827$; Predicted $R^2 = 0.9018$; Adeq Precision = 26.6195; Std. Dev. = 1.69; Mean = 62.14; C.V. % = 2.72.

The Model F-value of 72.13 underscored the adequacy of the model. This F-value indicates that the factors effectively elucidated the variation in data concerning its mean, thereby substantiating the credibility of potential factor effects. Furthermore, the conditions requisite for achieving optimal response in this study are presented in Table 3.

Table 3: Conditions for Obtaining Optimum Response for the Selected Agricultural Waste Using Hydrothermal Method

| Agricultural Wastes | Particle Size (μm) | Reagent Ratio | Contact Time (hr) | Temperature ($^{\circ}\text{C}$) | Porosity (%) | Desirability | |
|---------------------|---------------------------------|---------------|-------------------|------------------------------------|--------------|--------------|----------|
| Sugarcane Bagasse | 90 | 1.0 | 72 | 100 | 80.169 | 1.000 | Selected |

4. Conclusion

The production and optimization of zeolite material from low-cost sugarcane bagasse was analytically undertaken via hydrothermal technique. This entailed the manipulation of four variables at dual levels, guided by an extensive full factorial design. Importantly, the pinnacle of zeolite production was realized through precise parameter configurations: a particle size of 90 μm , a reagent ratio maintained at 1, a contact time spanning 72 hr and a temperature of 100 $^{\circ}\text{C}$.

Noteworthy is the revelation from this study that the produced zeolite had notable porosity, underscoring its remarkable adsorption capability, which opens up promising avenues for wastewater treatment, offering effective solutions to a myriad of environmental concerns. This approach not only addresses the pressing issue of agricultural waste management but also underscores the potential of transforming waste into valuable resources for sustainable development.

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References

- [1] F. Chen, Y. Li, A. Huang, Hydrophilicity reversal by post-modification: hydrophobic zeolite FAU and LTA coatings on stainless-steel-net for oil/water separation, *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, vol. 601, (2020). <https://doi.org/10.1016/j.colsurfa.2020.124936>.
- [2] E. Nyankson, J.K. Efavi, A. Yaya, G. Manu, K. Asare, J. Daafuor, R.Y. Abrokwah, Synthesis and characterisation of zeolite-A and Zn-exchanged zeolite-A based on natural aluminosilicates and their potential applications, *Cogent Engineering*, 5:1440480 (2018) 1-23. <https://doi.org/10.1080/23311916.2018.1440480>.
- [3] L. Mazzeo, T. Boscarino, M.B. Falasconi, S. Polvi, V. Piemonte, L. Pecchia, Zeolite Synthesis from Waste Materials for the Medical Field of Oxygen Concentrators: Focus on the African Scenario. *Chemical Engineering Transactions*, vol. 101 (2023) 163-168
- [4] N. Jiang, R. Shang, S.G.J. Heijman, L.C. Rietveld, Adsorption of triclosan, trichlorophenol, and phenol by high-silica zeolites: adsorption efficiencies and mechanisms, *Separation and Purification Technology*, vol. 235, (2020). <https://doi.org/10.1016/j.seppur.2019.116152>.
- [5] C.G. Flores, H. Schneider, S.J. Dornelles, B. Gomes, R. Marcilio, J.P. Melo, Synthesis of potassium zeolite from rice husk ash as a silicon source. *Cleaner Engineering and Technology*, vol 4:100201 (2021) 1-7. <https://doi.org/10.1016/j.clet.2021.100201>.
- [6] E.B.G. Johnson, S.E. Arshad, Hydrothermally synthesized zeolites based on kaolinite: a review. *Applied Clay Science*, vol. 97-98 (2007) 215–221. <https://doi.org/10.1016/j.clay.2014.06.005>.
- [7] A.A.B. Maia, R.N. Dias, R.S. Angélica, R.F. Neves, Influence of an aging step on the synthesis of zeolite NaA from Brazilian Amazon kaolin waste. *Journal of Materials, Research and Technology*, vol. 8, (2019) 2924–2929. <https://doi.org/10.1016/j.jmrt.2019.02.021>.
- [8] P. Krongkrachang, P. Thungngern, P. Asawaworarit, N. Hounkhang, A. Eiad-Ua, Synthesis of zeolite Y from kaolin via hydrothermal method,” *Materials Today: Proceedings*, vol. 17, (2019) 1431–1436. <https://doi.org/10.1016/j.matpr.2019.06.164>.
- [9] I.V. Joseph, L. Tosheva, A.M. Doyle, Simultaneous removal of Cd(II), Co(II), Cu(II), Pb(II), and Zn(II) ions from aqueous solutions via adsorption on FAU-type zeolites prepared from coal fly ash, *Journal of Environmental Chemical Engineering*, vol. 8:4 (2020) 103895. <https://doi.org/10.1016/j.jece.2020.103895>.
- [10] Z. Ma, X. Zhang, G. Lu, Y. Guo, H. Song, F. Cheng, Hydrothermal synthesis of zeolitic material from circulating fluidized bed combustion fly ash for the highly efficient removal of lead from aqueous solution. *Chinese Journal of Chemical Engineering*, vol. 47 (2022). 193–205. <https://doi.org/10.1016/j.cjche.2021.05.043>.
- [11] L.F. Magalhães, G.R. Silva, A.E.C. Peres, Zeolite Application in Wastewater Treatment. *Adsorption Science & Technology*, vol. 2022: 4544104 (2022) 1-26. <https://doi.org/10.1155/2022/4544104>.
- [12] M.A. Mahmud, F.R. Anannya, Sugarcane bagasse - A source of cellulosic fiber for diverse applications. *Heliyon*, vol. 7: 07771(2021) 1-14. <https://doi.org/10.1016/j.heliyon.2021.e07771>.
- [13] D. Michel, B. Bachelier, J.Y. Drean, O. Harzallah, Preparation of cellulosic fibers from sugarcane for textile use, in: *Conference Papers in Materials Science*, Guimaraes (2013). <https://doi.org/10.1155/2013/651787>.
- [14] B. Shah, P. Darshini, P. Hiren, A. Amare, S. Ajay, Zeolitic composites from agricultural detritus for pollution remedy: A Review. *J Crit Rev*, vol. 3:3 (2016). 41-49.
- [15] Y.R. Loh, D. Sujana, M.E. Rahman, C.A. Das, Sugarcane bagasse—the future composite material: a literature review. *Res. Cons. Recycl*, vol. 75: (2013) 14–22. <https://doi.org/10.1016/j.resconrec.2013.03.002>.
- [16] B.A. Shah, D.D. Pandya, H.A. Shah, Impounding of ortho-chlorophenol by zeolitic materials adapted from bagasse fly ash: four factor three level Box-Behnken design modelling and optimization. *Arabian Journal of Science and Engineering*, vol 42: (2016) 241–260. <https://doi.org/10.1007/s13369-016-2294-0>.
- [17] V. Matko, Porosity determination by using two stochastic signals. *Sensors and Actuators*, vol. 112 (2004) 320-327. <https://doi.org/10.1016/j.sna.2003.10.065>.