

UTILIZATION OF ORANGE PEEL-DERIVED BIOCHAR FOR AMMONIA ADSORPTION: ISOTHERM ANALYSIS AND HYDROGEN STORAGE PROSPECTIVE FOR SUPPORTING SUSTAINABLE DEVELOPMENT GOALS (SDGS)

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Abstract

This study investigated the potential of orange peel-derived biochar as an environmentally friendly and sustainable adsorbent for ammonia adsorption under isothermal conditions, with implications for hydrogen storage applications. The preparation process involved drying orange peels in an oven until carbonized, followed by grinding and sieving to obtain particle sizes of 500, 250, and 105 μm . A 0.05-g sample of each particle size was added to 500 mL of demineralized water, containing ammonia solutions at concentrations of 10, 20, 40, 60, and 80 ppm. The adsorption process was carried out under isothermal conditions, and the remaining ammonia concentrations were analysed to evaluate the biochar's adsorption capacity. The results showed that smaller particle sizes (105 μm) exhibited higher ammonia adsorption efficiency, demonstrating the influence of surface area and active site availability. Adsorption efficiency increased with higher ammonia concentrations, indicating the suitability of orange peel biochar for high-pollutant environments. This study discussed the role of particle size and concentration in adsorption mechanisms and highlighted the potential for ammonia adsorption. Furthermore, orange peel biochar demonstrates promise for ammonia-based hydrogen storage due to its high adsorption capacity, offering a sustainable pathway for renewable energy applications. This research supports the development of cost-effective, biomass-based materials for environmental remediation and energy storage technologies. This study also supports current issues in sustainable development goals (SDGs).

Keywords: Chemistry, Competence, Creative, Educational cluster, Methodology.

1. Introduction

Ammonia (NH_3) has garnered significant attention as a potential hydrogen carrier due to its high hydrogen content, ease of storage, and transportability. It is increasingly recognized as a key player in the development of sustainable hydrogen storage systems. However, ammonia's inherent properties, including its corrosive nature, pose challenges in handling and utilization. Efficient management is crucial to prevent material degradation and environmental hazards while enabling its effective use in hydrogen storage applications [1].

One promising solution is the use of adsorbents, which offer the ability to capture and contain ammonia safely. Many reports suggested the use of biomass for adsorbent (see Table 1). The table summarizes various studies on ammonia adsorption using activated carbon derived from orange peel and other biomass materials. Orange peel-derived activated carbon consistently demonstrates significant ammonia adsorption capacity, with enhancements achieved through chemical activation, surface functionalization, and microwave-assisted processes. Adsorption efficiency increases with smaller particle sizes and acidic conditions, as highlighted in multiple studies.

Table 1. Summary of research on orange peel and biomass-derived activated carbon for ammonia adsorption.

No.	Results	Ref.
1	Ammonia adsorption capacity significantly increased through chemical activation.	[2]
2	Higher ammonia removal efficiency was observed with coconut shells and orange peels.	[3]
3	Adsorption follows both Langmuir and Freundlich isotherms.	[3]
4	Surface functionalization enhances ammonia uptake capacity.	[4]
5	Ammonia removal is more effective in acidic conditions but declines in neutral/alkaline pH.	[5]
6	Smaller particle sizes exhibit higher adsorption capacities.	[6]
7	Microwave-assisted activation results in more efficient adsorbents.	[7]
8	Orange peel-derived activated carbon shows the highest ammonia adsorption capacity.	[8]
9	Adsorption driven by electrostatic forces and bonding on the carbon surface.	[9]
10	Cost-effective and environmentally sustainable, but regeneration is limited.	[10]

The adsorption process is influenced by electrostatic forces, bonding on the carbon surface, and the structural properties of the activated carbon, such as pore size and surface area. The Langmuir and Freundlich isotherms are commonly used to describe the adsorption mechanisms. Orange peel-based activated carbon is recognized as a cost-effective and environmentally sustainable solution, with high ammonia removal efficiency. However, some studies note limitations, such as reduced efficiency under neutral or alkaline conditions and challenges in regeneration. These findings emphasize its potential for industrial applications, particularly in waste gas treatment and hydrogen storage systems.

Although activated carbon has been extensively studied (as shown in Table 1), the use of orange peel-derived carbon remains underexplored [11]. Some researchers [12, 13] highlighted its potential for ammonia adsorption, yet critical factors such as particle size variation and simpler carbonization methods require further investigation. Additionally, its adsorption behaviour under neutral conditions, which are highly relevant to hydrogen storage and industrial applications, has not been comprehensively examined.

In our previous studies, we reported a method for producing carbon biochar from various raw materials: banana stem [14], pumpkin seeds [15], eggshell waste [16, 17], natural zeolite [18], rice husk [19], tamarind [20], date palm seeds [21], mangosteen [22], water hyacinth [23], olive industry solid waste [24], and others. These materials were primarily utilized as adsorbents in various environmental applications, focusing on wastewater treatment, dye removal, and pollution control. Many of them were derived from agricultural, food, and industrial waste, highlighting the studies' emphasis on sustainability and waste utilization. This body of work contributes significantly to advancing sustainable technologies for environmental protection, promoting the use of agricultural and industrial waste for adsorption, and improving understanding of adsorption mechanisms through experimental and computational approaches. The research is valuable for academic, industrial, and educational contexts, driving innovation in wastewater treatment, pollution control, and sustainable material development.

This study explored the potential of orange peel-derived biochar for ammonia adsorption, focusing on the effect of particle size on adsorption capacity and applying various isotherm models to describe the adsorption process. Biomass-derived adsorbents, particularly orange peel biochar, present an eco-friendly and cost-effective approach to ammonia adsorption. Orange peel, often regarded as waste, contains cellulose and functional groups that, when carbonized, produce activated carbon with excellent adsorption properties. Its high surface area and porous structure make it a suitable candidate for ammonia adsorption, addressing both its corrosive nature and its role as a hydrogen carrier.

This study explores the potential of orange peel-derived carbon as a sustainable adsorbent for ammonia adsorption under isothermal conditions. It specifically examines the effect of particle size variation (105, 250, and 500 μm) on the adsorption capacity of orange peel carbon, combining principles of physical chemistry with biomass waste management. The research began with the carbonization of orange peels through oven drying, resulting in black, charcoal-like material. This carbon was ground and sieved using mesh sizes of 34, 60, and 150 to evaluate the influence of particle size on adsorption efficiency. A 0.05-g sample from each particle fraction was added to 500 mL of demineralized water containing ammonia at concentrations of 10, 20, 40, 60, and 80 ppm. The adsorption process was conducted under controlled isothermal conditions, followed by filtration and measurement of residual ammonia concentration in parts per million (ppm).

To understand the adsorption mechanism, ten adsorption isotherm models were used. Adsorption isotherms, such as Langmuir, Freundlich, Dubinin-Radushkevich, and Halsey models, provide insights into the interaction mechanisms between ammonia and adsorbents. The Langmuir model describes monolayer adsorption on uniform surfaces, while the Freundlich model addresses adsorption on heterogeneous surfaces. Advanced models like Dubinin-Radushkevich and Halsey explain energy distribution and multilayer adsorption.

However, these models are often applied individually, limiting a holistic understanding of adsorption dynamics.

Although prior research has demonstrated the effectiveness of orange peel-derived carbon for ammonia adsorption, the role of particle size variation in enhancing adsorption efficiency has remained largely unexplored. Many studies prioritize energy-intensive chemical activation or surface functionalization over simpler carbonization techniques. Furthermore, while activated carbons have shown effectiveness in acidic or basic conditions, limited attention has been given to their performance in neutral environments, which are more relevant for industrial applications. This study aims to address these gaps by investigating the feasibility of using orange peel carbon as an economical, eco-friendly adsorbent in neutral, isothermal settings.

The experimental results confirmed that orange peel carbon effectively adsorbs ammonia, with higher concentrations leading to increased adsorption capacity. Smaller particle sizes (150 mesh) demonstrated superior performance, highlighting the significance of surface area and active site availability. The study also analysed the adsorption mechanisms using ten isotherm models, including Langmuir, Freundlich, and Dubinin-Radushkevich, to provide an in-depth understanding of the physicochemical interactions between orange peel carbon and ammonia molecules. Thermodynamic analyses further elucidated the factors driving the adsorption process.

This study aims to bridge these gaps by evaluating orange peel-derived biochar for ammonia adsorption, emphasizing particle size variations, and incorporating multiple isotherm models to characterize adsorption mechanisms. Furthermore, the dual importance of ammonia-as a corrosive chemical requiring effective handling and as a hydrogen carrier-highlights the relevance of this research. By optimizing adsorption conditions and advancing the understanding of ammonia interaction with biomass-based adsorbents, this work supports the development of efficient hydrogen storage systems while promoting the utilization of sustainable and eco-friendly materials.

This research contributes to the development of sustainable biomass-based adsorbent materials, offering a cost-effective solution for ammonia gas treatment, particularly in industrial applications. Orange peel-derived carbon, produced through straightforward carbonization without chemical activation, is a viable alternative for mitigating ammonia emissions. Moreover, the study emphasizes the potential of turning orange peel waste, often considered valueless, into a high-performance material for adsorption technology. By bridging the gap between fundamental scientific principles and practical applications, this research opens new avenues for the utilization of biomass-derived materials in air pollution control and hydrogen storage technologies. This study also supports the current issue of sustainable development goals (SDGs), as reported elsewhere [25-29].

2. Method

The materials used in this study include orange peel waste obtained from local markets in Sumedang and Tasikmalaya, Indonesia. Analytical-grade ammonium chloride (NH_4Cl) was purchased from Merck, while Nessler's Reagent (Hanna) and distilled water (Amidis, Indonesia) were used for preparing ammonia solutions and conducting adsorption tests.

A total of 72 g of orange peel waste was first sun-dried for 5 hours to remove surface moisture. The dried samples were then carbonized/pyrolyzed in an electrical oven at 250°C for 6 hours to ensure complete conversion into charcoal. The resulting carbon was ground to produce smaller particles and subsequently sieved through meshes with sizes of 500, 250, and 105 μm to obtain fractions for testing.

The carbon particles were then put into the adsorption process, following our previous studies [30-32]. A stock ammonia solution of 100 ppm was prepared by dissolving 0.297 g of NH_4Cl into 1 liter of distilled water. From this stock solution, ammonia solutions with varying concentrations (10, 20, 40, 60, and 80 ppm) were prepared. The concentration of NH_4^+ ions was analysed using the colorimetric method with a Hanna HI733 instrument. For adsorption testing, 0.05 g of orange peel-derived carbon from each particle size (500, 250, and 105 μm) was used. The adsorption process was conducted with all ammonia solution concentrations. The ammonia-carbon mixtures were stirred at 5,000 rpm using a magnetic stirrer for contact times of 0, 2, 15, and 30 minutes. After stirring, the samples were filtered, and the remaining ammonia concentration was measured using the ammonia HR instrument.

The physical structure of the orange peel-derived carbon was analysed using a Digital Microscope (BXAW-AX-BC, China) to visualize particle shapes and sizes. This methodology ensures a comprehensive understanding of the adsorption behaviour of orange peel carbon and evaluates the influence of particle size and contact time on ammonia removal efficiency.

3. Results and Discussion

Figures 1(a)-1(c) provide detailed visualizations of the surface morphology and distribution of orange peel-derived adsorbent particles. The images reveal a range of particle sizes, highlighting the distinct dimensions and characteristics of each. Figure 1(a) depicts the smallest particles, measuring 105 μm , with a finer structure compared to the larger particles shown in Figs. 1(b) and 1(c), which measure 250 and 500 μm , respectively. Across all three sizes, the particles exhibit irregular geometries and heterogeneous surface characteristics, emphasizing the non-uniform nature of the adsorbent material. These irregularities likely contribute to the differences in adsorption efficiency, as they influence the surface area and the availability of active sites for ammonia interaction. Figure 1(d) presents a histogram illustrating the relationship between particle size and frequency. The data indicate that the particle size with the highest frequency corresponds to 250 μm , while the lowest frequency is associated with 60 μm . This distribution highlights the dominance of certain particle sizes within the processed material and underscores the variability inherent in the preparation process. Understanding this relationship is crucial for optimizing the particle size to enhance adsorption efficiency, as surface area and particle distribution significantly impact the performance of the adsorbent. These results also confirmed that pyrolysis is effective in converting biomass into carbon [33-37].

The equilibrium data for ammonia adsorption using orange peel-derived adsorbents were analysed with ten isotherm models, including Freundlich, Temkin, Dubinin-Radushkevich, Jovanovic, Halsey, Harkin-Jura, Flory-Huggins, Fowler-Guggenheim, and Hill-DeBoer. Parameter values and statistical correlations are summarized in Table 2.

The correlation coefficients (R^2) for ammonia adsorption using orange peel-derived adsorbents of varying particle sizes were analysed to determine the most suitable adsorption isotherm models for each dimension. The resulting rankings of isotherm models based on their compatibility with the experimental data are as follows:

- (i) 500 μm : Langmuir, Freundlich, Halsey > Temkin, Jovanovic > Dubinin-Radushkevich > Hill-DeBoer > Harkin-Jura > Fowler-Guggenheim > Flory-Huggins.
- (ii) 250 μm : Langmuir, Freundlich, Halsey > Temkin, Jovanovic > Dubinin-Radushkevich > Harkin-Jura > Hill-DeBoer > Flory-Huggins > Fowler-Guggenheim.
- (iii) 105 μm : Langmuir, Freundlich, Halsey > Temkin, Jovanovic > Hill-DeBoer > Dubinin-Radushkevich > Harkin-Jura > Fowler-Guggenheim > Flory-Huggins.

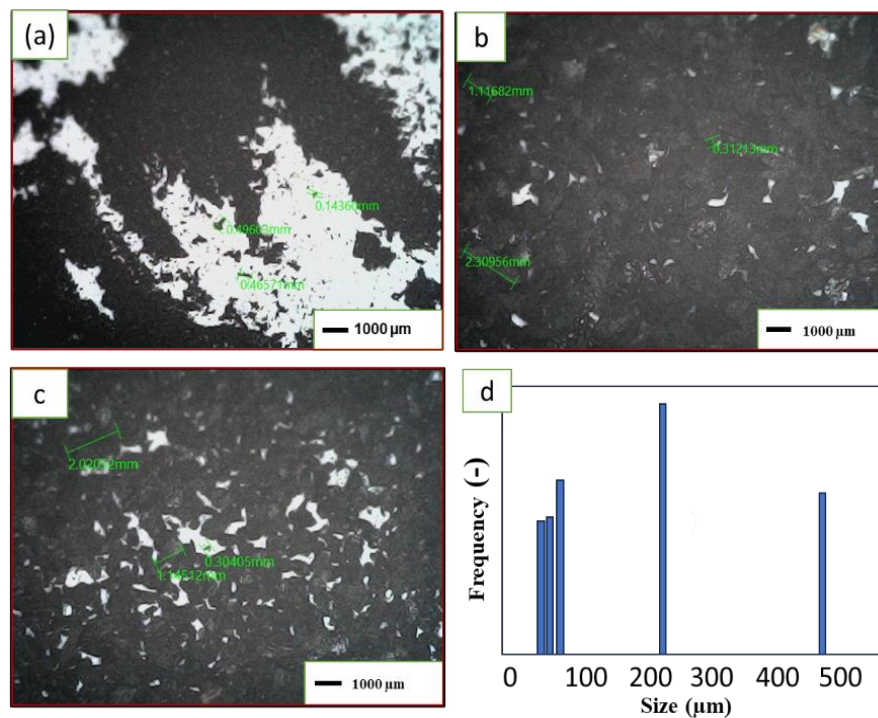


Fig. 1. Digital microscope camera image of adsorbent with their size distribution (a) 105, (b) 250, and (c) 500 μm , and (d) chart analysis.

Table 2. Isotherm models and parameters for the adsorption.

Model	Parameter	Particle Size (μm)			Note
		500	250	105	
Langmuir	R^2	1	1	1	Monolayer ($R^2 > 0.70$)

Model	Parameter	Particle Size (μm)			Note
		500	250	105	
Freundlich	Q_{max}	1.442	1.442	1.442	Adsorbent maximum capacity (mg/g)
	R_L	0.262	0.557	0.511	$0 < R_L < 1$ favourable
	K_L	0.693	0.693	0.693	Langmuir model
	R^2	1	1	1	Multilayer ($R^2 > 0.70$)
	K_f	2.728	2.718	2.718	Freundlich constant
	nF	1	1	1	Linear adsorption ($n = 1$)
	$1/n$	1	1	1	Non-cooperative adsorption ($1/n = 1$)
Temkin	R^2	0.949	0.939	0.946	Monolayer ($R^2 > 0.70$)
	A_T (L/g)	7.417	0.236	0.624	Coefficient of binding
	β_T (J/mol)	1.651	5.086	2.752	Physical adsorption ($\beta_T < 8$ kJ)
Dubinin-Raduskhevich	R^2	0.902	0.931	0.857	Microspore ($R^2 > 0.7$)
	β (mol^2/kJ^2)	0.033	0.005	0.01	Dubinin-Raduskhevich constant
	E (kJ/mol)	3.882	9.36	6.813	If $E > 8$ kJ, chemical adsorption, If $E < 8$ kJ, physical adsorption
Jovanovic	R^2	0.949	0.939	0.946	Monolayer ($R^2 > 0.70$)
	Q_{max}	4.894	1.669	2.574	Adsorbent's maximum capacity (mg/g)
	K_j	0.13	0.397	0.216	Jovanovic constant
Halsey	R^2	1	1	1	Multilayer ($R^2 > 0.70$)
	N	1	1	1	Halsey constant
	K_H	2	2	2	Halsey constant
Harkin-	R^2	0.885	0.907	0.794	Multilayer

Model	Parameter	Particle Size (μm)			Note
		500	250	105	
Jura					($R^2 > 0.70$)
Flory-Huggins	A_{HJ}	- 29.086	-3.143	-7.631	Harkin-Jura constant
	B_{HJ}	0.037	0.192	0.117	Associated with the adsorbent's surface area
	R^2	0.326	0.673	0.253	Monolayer ($R^2 < 0.70$)
	n_{FH}	0.628	0.516	0.462	$n_{FH} < 1$, active adsorbent zone
	K_{FH} (L/mg)	2.141	0.509	0.545	Flory-Huggins constant
Fowler-Guggenheim	ΔG^o (kJ/mol)	18.298	- 16.203	- 14.569	spontaneous for $\Delta G^o < 0$ not spontaneous for $\Delta G^o > 0$
	R^2	0.691	0.293	0.655	Monolayer ($R^2 < 0.70$)
	K_{FG} (L/mg)	0.002	0.002	1.117	Fowler-Guggenheim constant
Hill-DeBoer	W (kJ/mol)	-6.919	-8.072	- 19.291	Repulsive interaction between adsorbed molecules ($W < 0$)
	R^2	0.891	0.84	0.897	Multilayer ($R^2 > 0.70$)
	K_1 (L/mg)	1.2	2.14	2.88	Hill-DeBoer constant
	K_2 (L/mg)	-1391	-5305	-7722	Attraction between adsorbed species

These classifications indicate that the Langmuir, Freundlich, and Halsey models consistently provided the best fit for all particle sizes, suggesting their suitability in describing both monolayer and multilayer adsorption on heterogeneous surfaces. Variations in the ranking of other models reflect differences in adsorption mechanisms influenced by particle size and surface characteristics.

Based on the parameter fit analysis presented in Table 2, the general characteristics of ammonia adsorption phenomena for the three particle sizes are illustrated in Fig. 2, which depicts the mechanism of ammonia adsorption using orange peel-derived adsorbents.

The experimental results highlight several key aspects of the ammonia adsorption process with orange peel biochar. Using data derived from ten adsorption isotherm models, the findings indicate that, across all particle sizes, ammonia adsorption predominantly occurs as a monolayer. Additionally, the presence of pores facilitates the ammonia adsorption process, which occurs in a non-cooperative and favourable manner.

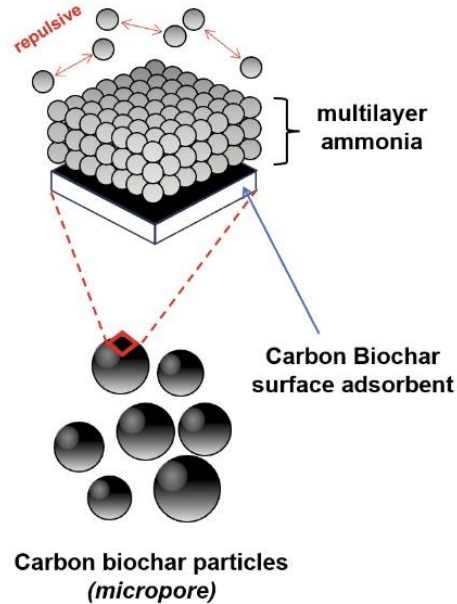


Fig. 2. Illustration of ammonia adsorption mechanism using orange peel-derived adsorbents.

Despite these shared characteristics, the adsorption behaviour varies depending on particle size. For 250 and 105 μm particles, the process occurs spontaneously, while for 500 μm particles, the adsorption is non-spontaneous. Furthermore, at a particle size of 250 μm , adsorption is chemically driven, whereas at 500 and 105 μm , adsorption is dominated by physical interactions. These differences underscore the influence of particle size on the underlying adsorption mechanisms and efficiency.

Orange peel-derived carbon demonstrates significant potential as a material for ammonia hydrogen storage due to its adsorption efficiency, sustainability, and cost-effectiveness. Ammonia (NH_3) is increasingly recognized as a promising hydrogen carrier, offering a high hydrogen content (17.6% by weight) and ease of storage and transport under moderate conditions. The ability of orange peel biochar to effectively adsorb ammonia positions it as a viable candidate for applications in hydrogen storage systems.

- (i) **High Adsorption Capacity and Surface Area:** Orange peel biochar exhibits excellent ammonia adsorption capacity due to its porous structure and high surface area. These properties enhance the storage of ammonia in solid adsorbents, facilitating controlled adsorption and desorption processes necessary for hydrogen release.

- (ii) **Chemical and Physical Versatility:** The results of adsorption studies highlight the dual ability of orange peel carbon to engage in both physical and chemical adsorption processes, depending on particle size. This adaptability allows for optimization of adsorption conditions, enhancing its efficiency in storing and releasing ammonia for hydrogen extraction.
- (iii) **Eco-Friendly and Sustainable:** Derived from agricultural waste, orange peel biochar represents an environmentally friendly alternative to conventional adsorbents. Its use aligns with circular economy principles, reducing waste while supporting sustainable energy systems.
- (iv) **Cost-Effective Production:** The straightforward carbonization process used to produce orange peel-derived carbon eliminates the need for chemical activation or costly functionalization methods. This makes it a cost-effective solution for large-scale applications in hydrogen storage.
- (v) **Scalability and Industrial Applications:** Orange peel biochar's versatility and performance under various conditions, including neutral environments, make it suitable for integration into industrial-scale ammonia hydrogen storage systems. It offers a low-cost, renewable option for industries seeking to adopt sustainable energy solutions.
- (vi) **Thermal Stability and Reusability:** Preliminary findings suggest that orange peel-derived carbon exhibits good thermal stability, an essential property for the repeated adsorption and desorption cycles required in hydrogen storage technologies. Further research into its regeneration capabilities can enhance its practicality for long-term applications.

To fully realize the potential of orange peel-derived carbon in ammonia hydrogen storage, further studies should focus on:

- (i) Advanced characterization techniques (e.g. surface analysis, electron microscope) to optimize pore structure and surface area for enhanced performance.
- (ii) Investigation of thermal and chemical stability under repeated adsorption-desorption cycles.
- (iii) Development of hybrid systems combining orange peel biochar with other materials to improve storage efficiency and hydrogen release rates.
- (iv) Assessment of scalability and cost-efficiency in industrial applications.

Orange peel-derived carbon represents a promising step toward sustainable and efficient hydrogen storage systems, contributing to the transition toward renewable energy and reduced reliance on fossil fuels. Finally, this study adds new information and ideas regarding the use of carbon for the adsorption process, as reported elsewhere [38-42]. This study also supports the current issue of SDGs from using biomass and the potential use of biomass for supporting clean energy.

4. Conclusion

This study demonstrates the significant potential of orange peel-derived carbon as an eco-friendly and cost-effective material for ammonia adsorption, with promising implications for ammonia-based hydrogen storage. The adsorption process was found to vary with particle size, with smaller particles (105 μm) exhibiting higher adsorption efficiency due to their increased surface area and availability of active sites. Across all particle sizes (500, 250, and 105 μm), the adsorption process was predominantly monolayer, facilitated by the porous structure of the carbon and

occurring under favourable conditions. The ability of orange peel carbon to engage in both physical and chemical adsorption, depending on particle size, highlights its versatility. For instance, 250 μm particles demonstrated chemically driven adsorption, while 500 and 105 μm particles exhibited physical adsorption. This adaptability enhances its suitability for applications requiring controlled adsorption and desorption, critical for hydrogen storage technologies. Orange peel-derived carbon offers several advantages, including a straightforward production process without chemical activation, cost-effectiveness, and alignment with sustainability goals by repurposing agricultural waste. Its potential extends beyond ammonia removal to include applications in renewable energy systems, particularly as a material for ammonia hydrogen storage. Future research should focus on enhancing its thermal stability, regeneration capabilities, and scalability, ensuring its effectiveness in industrial and energy applications.

Acknowledgment

This paper is supported by DRPTM Dikti, Indonesia.

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