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THE EFFECT OF CHEMICAL ACTIVATION AGENTS AND ACTIVATION TEMPERATURE ON THE PORE STRUCTURE OF RICE HUSK-DERIVED ACTIVATED CARBON

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Abstract: This study investigates the optimization of production parameters for rice husk-derived activated carbon, targeting its effective application in direct air capture (DAC) technology. Various chemical activation agents—potassium hydroxide (KOH), urea, and their combination—and activation temperatures (600°C, 700°C, and 800°C) were explored using pyrolysis. The morphology of the resulting activated carbon was analyzed via scanning electron microscopy (SEM) and ImageJ. Results demonstrate that the choice of activation agent and temperature significantly influence pore diameter size and quantity. Higher temperatures led to smaller pore diameters and an increased number of pores. The combination of KOH and urea at 800°C produced the most favorable particle size (0.811 µm), suitable for applications requiring a well-defined pore structure. This combination also exhibited the most even pore distribution and highest pore density. These findings provide valuable insights for optimizing the production of rice husk-derived activated carbon, contributing to the development of sustainable and effective sorbents for CO_2 capture in DAC technology. Additionally, they offer potential for broader applications of husk-activated carbon in various industrial and environmental fields.

Keywords: Direct air capture, activated carbon, rice husk, chemical activation agents, activation temperature

1. Introduction

The escalating atmospheric CO_2 burden necessitates immediate intervention. Deploying efficient and sustainable CO_2 capture technologies is critical to mitigate its adverse impacts. Current options include pre-combustion, post-combustion, and oxyfuel capture methods, serving as platforms for further advancement (Cannone et al., 2021). However, the spatiotemporal variability of atmospheric CO_2 concentrations presents a significant challenge for the widespread implementation of capture technologies (Deng et al., 2021). Consequently, direct air capture (DAC) technology has emerged as a promising approach, focusing on directly removing CO_2 from ambient air.

DAC technology represents a negative emissions technique that can directly reduce CO_2 concentrations in the atmosphere. One of its key advantages is independence from concentrated emission sources, such as flue gases from cement plants, steam power plants, and steel processing facilities, making it a promising option for achieving significant CO_2 reduction targets. However, several challenges must be addressed to successfully develop and deploy DAC technology. These challenges include the low partial pressure of CO_2 in the air, which reduces the sorbent's efficiency, as well as the low thermal and chemical stability of the sorbent material and the high energy requirements for sorbent regeneration (Lee et al., 2021). The search for environmentally friendly sorbent candidates has become crucial to advance DAC technology within the framework of a green economy. This study aims to identify sorbents that possess the desired characteristics for efficient CO_2 capture. Ideal criteria for sorbent materials include high specific surface area, high thermal conductivity, high CO_2 adsorption capacity, high selectivity for CO_2 over N_2, high porosity, ease of recyclability, and low energy consumption for regeneration (Dissanayake et al., 2020; Shi et al., 2020; Wang et al., 2023).

Several adsorbents such as porous carbon (Dissanayake et al., 2020), activated carbon (Rinawati et al., 2019), zeolite (Zhang et al., 2019), metal-organic frameworks (Elhenawy et al., 2020), and nanomaterials (Segneri et al., 2023) have been studied for CO_2 capture applications. These materials demonstrate excellent CO_2 adsorption performance. However, the difficulty of finding raw materials and the limited availability of materials pose challenges in terms of cost-effectiveness and commercial feasibility for industrial applications (Goembira et al., 2021). To overcome this problem, the synthesis of activated carbon from rice husks has received much attention. Activated carbon derived from rice husks is known for its extraordinary adsorption capabilities, making it a versatile and popular adsorbent material (Rinawati et al., 2019; Yaumi et al., 2018; Hussin et al., 2021). Utilizing agricultural waste as a precursor for activated carbon synthesis aligns with the increasing environmental concerns and the demand for sustainable practices. Rice husks, an abundant byproduct of rice milling, offer significant potential as a cost-efficient and sustainable source of activated carbon, particularly in regions such as West Sumatera, Indonesia, where rice cultivation is prevalent. However, the properties and functionality of activated carbon are significantly influenced by various factors,

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such as the selection of chemical activating agents and the activation temperature (Jouhara et al., 2018).

Therefore, it is important to know the optimal combination of chemical activating agents and activation temperature to optimize the production process and improve the performance of activated carbon derived from rice husks. This research aims to determine the most suitable parameters to produce activated carbon with the desired characteristics, especially a clear pore structure. Observations of pore structure focus on the size, shape, distribution, and density. The results of these observations can not only be used as a reference for determining treatments and materials as CO_2 adsorbents but can also be applied to various industrial and environmental applications. Industrial and environmental applications of activated carbon include supercapacitors, water filters, oil and gas purifiers, poison absorbers, pharmaceutical product cleaners, and aroma and taste absorbers.

Research to determine the microstructural parameters of a material has been carried out, but the material observed is often nanoporous. Such research is necessary to use statistical analysis methods to process large arrays of nanosized pores, develop methodologies and algorithms to study morphological and nanostructural features, and automate the measurement process (Lushpa et al., 2018). In this research, the observations focused on activated carbon material from rice husk charcoal with a different end goal. The observed characterization only focuses on morphological observations using Scanning Electron Microscopy (SEM) which will be used as quantitative data with the help of the ImageJ application.

The size, shape, distribution, and density of rice husk-activated carbon treated with physical and chemical activation variations were observed. To achieve this objective, a systematic experimental approach was employed. Various chemical activating agents, namely potassium hydroxide (KOH), urea, and a combination of KOH and urea, were selected for the activation process. Different activation temperatures within a predetermined range of 600°C, 700°C, and 800°C were considered. The activation process was carried out using the pyrolysis method. Meanwhile, the rice husk-derived activated carbon morphology was investigated using scanning electron microscopy (SEM) and the ImageJ application. The experimental results provided insights into the impact of different activating agents and activation temperatures on the pore diameter size and pore quantity of the activated carbon derived from rice husk. By understanding the correlation between the synthesis variables and the properties of activated carbon, this research seeks to contribute to the development of effective treatments to produce activated carbon that can be applied in industry and the environment.

2. Materials and methods

2.1 Materials

Rice husk (Oryza sativa L.), obtained from a rice mill in Padang, West Sumatera, Indonesia, was selected as the raw material. The rice husks were cleaned with distilled water to remove adhering dust and inorganic impurities and then dried at 105°C for 12 hours. Potassium hydroxide (KOH ≤ 100%) was purchased from PT Smart Lab Indonesia, and urea ($CH4N2O \leq 99\%$) was obtained from Pudak Scientific. Before testing, the samples were cleaned and neutralized with distilled water and hydrochloric acid (HCl 15%) purchased from Rofa Laboratory Centre.

2.2 Preparation of sample

Activated carbon (AC) was prepared using the pyrolysis method, as shown in the flow diagram in Figure 1. Initially, the cleaned rice husks were placed in a container for carbonization. Prior to the carbonization process, nitrogen was introduced into the furnace for 5 minutes at a flow rate of 80 ml/min to displace oxygen and prevent combustion reactions. The carbonization process was conducted at 500°C with a heating rate of 10°C/minute for 2 hours, while nitrogen continued to flow at a rate of 80 ml/minute. After cooling to room temperature, the resulting biochar was removed from the furnace for the activation process.

In the activation process, the biochar was impregnated with activating agents (KOH, urea, and a combination of KOH and urea) to produce porous carbon, with a mass ratio of 1:2 (mass of adsorbent: activating agents). The mixture was stirred and left for 12 hours at 60°C. Subsequently, the samples were pyrolyzed in the furnace for 1 hour at various activation temperatures of 600°C, 700°C, and 800°C, with a nitrogen flow of 50 ml/minute. After cooling to room temperature, the samples were taken out of the furnace and immersed in a 15% HCl solution, followed by repeated washing with distilled water until the washing solution became neutral. The samples were then dried for 24 hours in an oven at 80°C. Finally, the activated carbon samples were stored in sample bags.

2.3 Characterization

2.3.1 Morphology investigation using field emission Scanning Electron Microscopy (SEM)

The morphology of the activated carbon samples was studied using field emission scanning electron microscopy (SEM). The samples were placed on SEM stubs and plated with carbon, followed by gold for two minutes using an argon plasma metallizer (sputter coater K575X) (Edwards Limited, Crawley, United Kingdom) to reduce electron charge through the sputtering process. Field Emission Scanning Electron Microscopy (FEI Nova Nano SEM 230) was utilized with a 10 kV voltage to optimize the observation of sample surface morphology at 5000x magnification.

2.3.2 Measurement pore diameter dan pore number using ImageJ application

The ImageJ application was chosen for measuring the pore diameter and pore number due to its simplicity and user-friendly

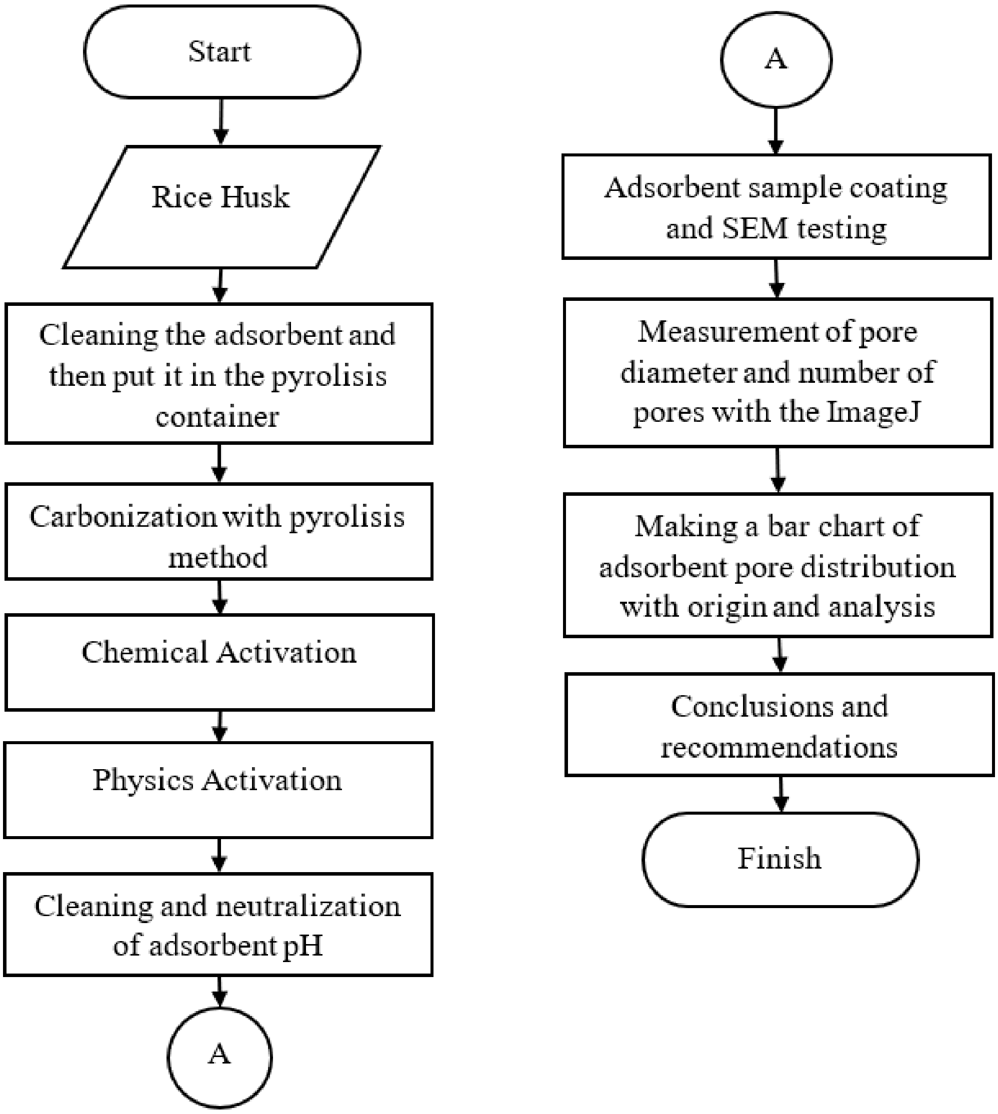

Figure 1. Sample Preparation Flow Diagram

interface. ImageJ is widely used open-source software that provides various image analysis tools, making it suitable for analyzing SEM images and extracting quantitative data. To begin the measurement process, the SEM images were imported into the ImageJ software. The images were calibrated to ensure accurate measurements by aligning the scale and line tool units with the SEM ruler. This calibration step allowed for precise conversion of pixel dimensions into real-world measurements.

The SEM images were divided into four sections for more manageable and systematic measurements. This division helped avoid confusion and ensure consistency in the measurements across the image. Using the line tool in ImageJ, manual measurements of pore diameter and pore number were conducted on each section of the SEM images. The line tool allowed for the selection of individual pores, and their diameters were measured directly from the images. Additionally, the number of pores present in each section was counted manually.

Once the pore diameter and pore number data were obtained, further analysis was conducted by grouping the data based on the same activation temperature and activating agent. This grouping allowed for a comprehensive examination of the influence of different activating agents and activation temperatures on the pore characteristics of the rice husk-activated carbon. To visualize the distribution of pore diameters, the data were processed using OriginPro 2019 software. The software facilitated the creation of a bar chart, where the pore sizes were plotted along the x-axis, and the corresponding frequencies or counts were represented on the y-axis. This graphical representation provided a clear understanding of the pore size distribution in the rice husk-activated carbon samples.

The analysis of the bar chart was crucial for assessing the characteristics of the adsorbent material. By comparing the pore diameter distributions resulting from different activating agents and activation temperatures, valuable insights could be gained

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regarding the impact of these parameters on the surface conditions and adsorption properties of the rice husk-activated carbon.

3. Results and discussions

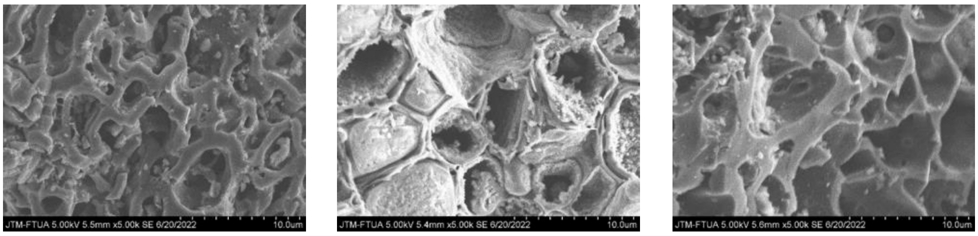
3.1 Effect of chemical activation agents and activation temperatures on the morphology of adsorbent

Figure 2 (a - i) visually represents the morphology resulting from different chemical activation agents (KOH, urea, KOH+urea) at various activation temperatures on the pore formation process in rice husk-derived activated carbon. Specifically, Figure 2 (a-c) illustrates the formation of pores in activated carbon using KOH, urea, and a combination of KOH and urea at an activation temperature of 600°C. The presence of visible boundaries between pores is evident in these samples. This phenomenon is also observed with the activation agent urea and the combination of KOH and urea.

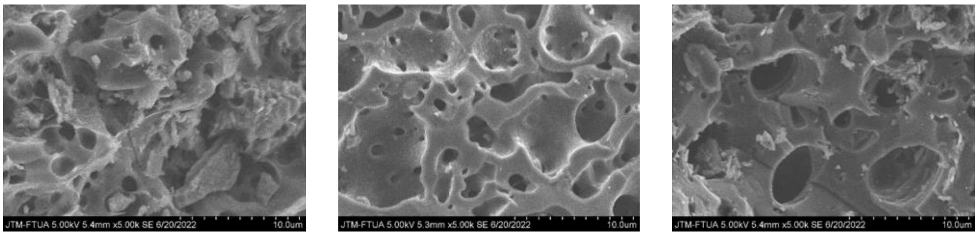
Furthermore, when the activation temperature was increased from 700°C to 800°C, there was a noticeable enhancement in pore formation, as shown in Figure 2 (d-i). This result indicates that the activation process becomes more effective and efficient at higher temperatures. The higher temperature provides the necessary energy for enhanced chemical reactions and for developing a more extensive pore structure in the rice husk-activated carbon. As a result, the visible boundaries between pores observed at 600°C start to diminish, suggesting a more complete and optimized activation process at 800°C.

This phenomenon can be explained by the fact that potassium hydroxide (KOH), being a strong alkali, exhibits corrosive properties towards carbonaceous materials at high temperatures. Consequently, it has the ability to permeate through the material, creating a significant number of pores. Additionally, the reaction between KOH and carbon during the activation process generates gas. The effusion of this gas contributes to the expansion and enlargement of the pore structure. Thus, the combined effects of chemical reactions and gas effusion play a crucial role in altering the particle size of rice husk-activated carbon when KOH is used as the activating agent.

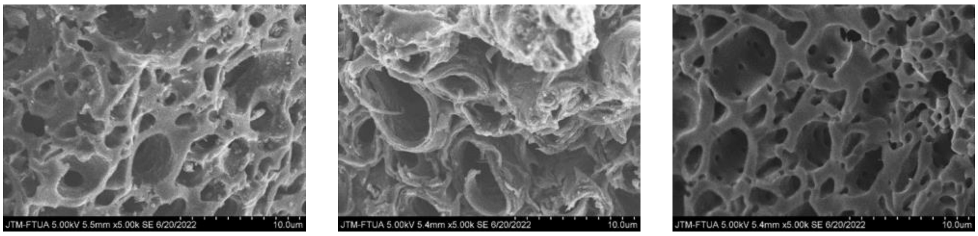
Furthermore, urea plays a significant role in the pore formation process of rice husk-activated carbon. When urea is used as an activation agent, it undergoes thermal decomposition during the activation process. This decomposition releases gases, such as ammonia (NH_3) and carbon dioxide (CO_2), which create pressure within the carbon matrix. The pressure generated by the gas release helps to create pores in the rice husk-activated carbon. The gases diffuse through the carbonaceous material, causing the formation of voids and channels. These voids and channels contribute to the development of a porous structure in the activated carbon. The decomposition of urea and the subsequent gas release also facilitate the removal of volatile



KOH, 600°C
Urea, 600°C
KOH and Urea, 600°C



KOH, 700°C
Urea, 700°C
KOH and Urea, 700°C



KOH, 800°C
Urea, 800°C
KOH and urea, 800°C

Figure 2. Morphology view of rice husk activated carbon at various chemical activating agents and activation temperatures

components and impurities from the rice husk, resulting in a purer and more highly porous carbon material.

Meanwhile, the combination of KOH and urea as activation agents for rice husk-activated carbon at high temperatures leads to several significant effects. Firstly, the interaction between KOH and urea initiates chemical reactions with the carbon precursor (rice husk), resulting in the decomposition and transformation of the carbonaceous material. This process releases gases and creates pores. Additionally, the combination of KOH and urea exhibits a synergistic effect on the activation process. The presence of both agents together enhances pore formation compared to using them individually, possibly due to a more efficient activation process or the generation of unique pore structures.

Moreover, the combined activation process promotes the release of gases, such as ammonia (NH_3) and carbon dioxide (CO_2), which contribute to the expansion and enlargement of the pore structure in the rice husk-activated carbon. The effusion of these gases facilitates the creation of interconnected pores, resulting in an overall increase in the material's porosity. Thus, the combined use of KOH and urea at high temperatures effectively induces chemical reactions, exhibits synergistic effects, and promotes gas release, all of which play crucial roles in pore formation and the development of a highly porous activated carbon material.

3.2 Effect of chemical activation agents and activation temperatures on the size and number of pores

The effects of chemical activation agents (KOH, urea, KOH + urea) and activation temperatures on the size and number of pores in rice husk-activated carbon were investigated. SEM images were analyzed using the ImageJ application, providing quantitative data on pore characteristics. The number and diameter of pores under various chemical activation agents at temperatures of 600°C, 700°C, and 800°C are presented in Figure 3 (a-c). Table 1 shows the average pore diameter of rice husk-activated carbon formed under different conditions. The KOH + urea activating agents produced a smaller average pore size (0.92 µm) than urea (1.17 µm) and KOH (1.2 µm). An activation temperature of 700°C resulted in a smaller average pore size (1.00 µm) compared to activation temperatures of 800°C (1.01 µm) and 600°C (1.28 µm).

During the activation process, the chemical activation agents (KOH, urea, and their combination) primarily act as catalysts and react with the carbonaceous components in the rice husk. However, the structural properties of the precursor material seem to play a dominant role in determining pore size rather than the specific activation agents used. The natural composition and arrangement of the carbonaceous components within the rice husk result in a relatively consistent range of pore diameters. Despite the use of different activation agents, the resulting pore diameter in the rice husk-activated carbon remains relatively unchanged. This suggests that the unique characteristics of the rice husk material itself strongly influence the resulting pore size, overriding the individual effects of the activation agents.

Table 1. Average diameter of activated carbon with various variations

		Chemical Activation Agents		
		KOH (µm)	**Urea (µm)**	**KOH+Urea µm)**
Activation Temperatures	**600°C**	1.40	1.37	1.06
	700°C	1.11	1.00	0.90
	800°C	1.10	1.13	0.81

The increase in activation temperature correlating with an increase in the number of pores in the rice husk-activated carbon can be attributed to enhanced reactivity between the chemical activation agents and the material at higher temperatures. Higher activation temperatures increase thermal energy for chemical reactions, promoting more extensive and efficient interactions between the activation agents (KOH, urea, KOH + urea) and the carbonaceous components in the rice husk. The chemical activation agents act as catalysts, facilitating the removal of volatile matter and the development of porosity in the rice husk material. Higher temperatures provide the necessary energy for these reactions to occur at a faster rate and to a greater extent, leading to the formation of additional pores.

Among the various combinations tested, the combination of KOH and urea as chemical activation agents at an activation temperature of 800°C yielded the most favorable results in terms of particle size, as shown in Figure 3 (c). This specific combination and temperature likely provided an optimal environment for the reactions to occur, resulting in a well-defined pore structure. The average diameter of the porous particles obtained under these conditions was measured to be 0.811 µm. This particle size is particularly suitable for applications requiring a well-defined pore structure, such as adsorption, gas separation, or catalysis. The presence of well-defined pores allows for efficient adsorption and separation processes, maximizing the material's performance in these applications.

In summary, the activation temperature increase in the number of pores in the rice husk-activated carbon can be attributed to higher temperatures facilitating more extensive reactions between the chemical activation agents and the material. The use of KOH and urea as activation agents at an activation temperature of 800°C resulted in the most favorable particle size.

4. Conclusion

The experimental results and discussions elucidate the effects of chemical activation agents and activation temperatures on the morphology, size, and number of pores in rice husk-activated carbon. SEM images clearly show the impact of different activation agents (KOH, urea, KOH + urea) on pore formation at various activation temperatures. The results indicate that an increase in activation temperature correlates with increased pore formation in the rice husk-activated carbon. This can be attributed to the higher thermal energy available at elevated temperatures, which facilitates more extensive reactions between the chemical activation agents and the carbonaceous material. Higher temperatures promote enhanced pore development, resulting in a more complete and optimized activation process.

Among the various combinations tested, that of KOH and urea as chemical activation agents at an activation temperature of 800°C yielded the most favorable results in terms of particle size. The resulting activated carbon exhibited an average pore diameter of 0.811 µm. This particle size is well-suited for applications requiring a well-defined pore structure, e.g., adsorption, gas separation, or catalysis.

The experimental findings demonstrate that the specific activation agents (KOH, urea, KOH + urea) had a limited impact on the pore diameter of the rice husk activated carbon. Alternatively, the rice husk material's structural properties tended to dominate in determining the pore size. The natural composition and

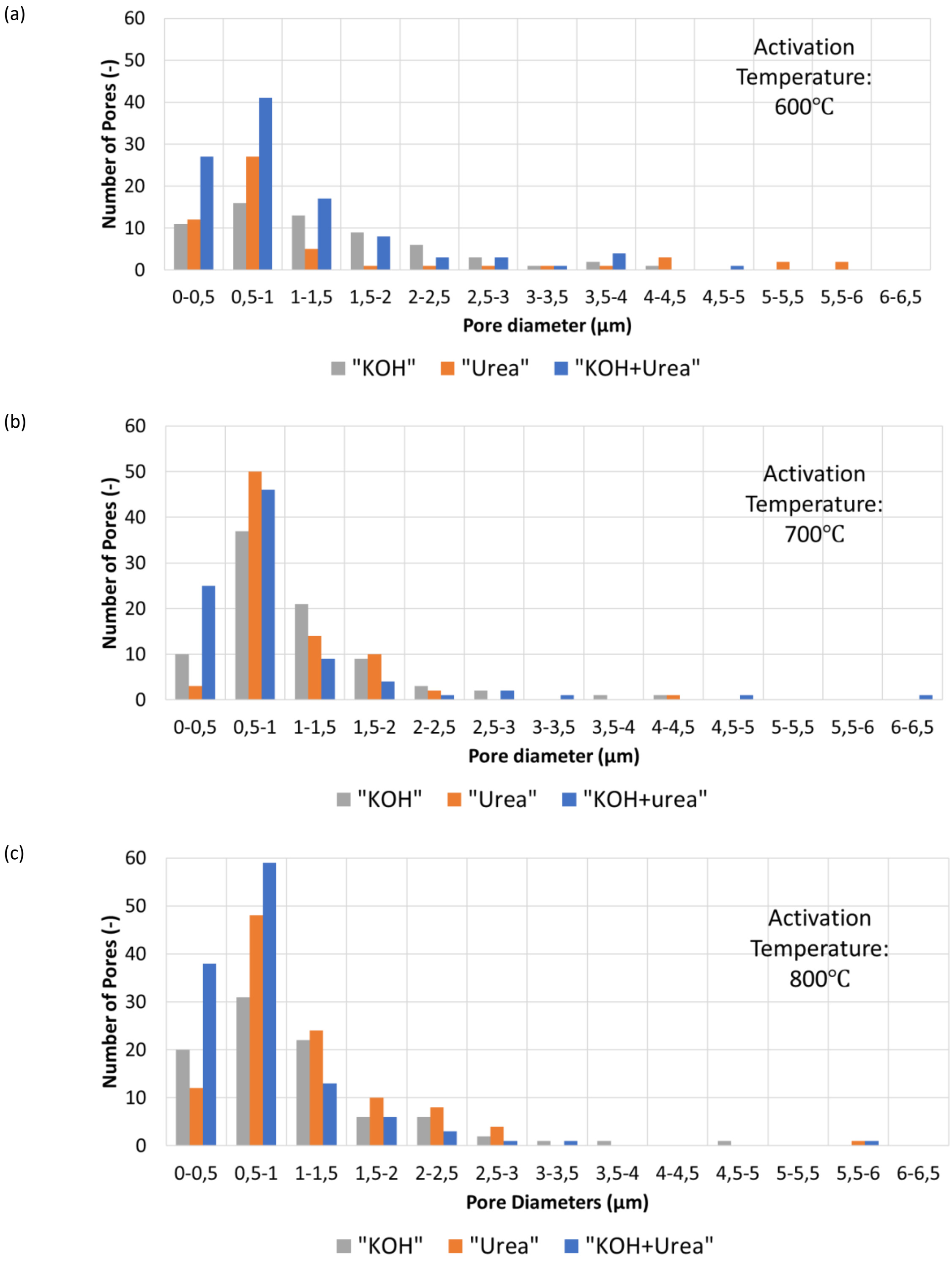


Figure 3. Effect of the chemical activating agent and activation temperatures on the pore number and size

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arrangement of the rice husk's carbonaceous components strongly influenced the resulting pore size, regardless of the activation agents employed. In summary, the research demonstrates the significance of activation temperature in pore development, with higher temperatures leading to increased pore formation. The combination of KOH and urea as activation agents at 800°C yielded a desirable particle size for applications requiring a well-defined pore structure. These results show that variations in the combination of KOH and urea produce a more even pore distribution and higher pore density compared to other variations. The study highlights the importance of considering both the activation temperature and the inherent properties of the precursor material when optimizing pore characteristics in rice husk-activated carbon.

5. Acknowledgement

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