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Review

A Comprehensive Review of Hierarchical Porous Carbon Synthesis from Rice Husk

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Abstract: Hierarchical porous carbon (HPC) materials exhibit superior performance profiles in various applications due to their well-developed multiscale interconnected pore structures. The synthesis of HPC from natural biomass precursors instead of fossil fuel-based precursors has gained considerable attention in recent decades. Rice husk, a globally abundant agricultural waste, offers a sustainable and cost-effective precursor for HPC production. The structural components and inherent silica content of rice husk act as a natural self-template for forming hierarchical pore structures with superior characteristics. In this review, recent studies on preparing rice husk-based HPC are summarized, and synthesis techniques are evaluated. In addition, recent advancements in activation methods and the effect of silica templates are reviewed while comparing these with traditional activated carbon production methods. Potential future directions for research and development activities are also discussed. Rice husk is a highly promising candidate for producing high-performance HPC materials.

Key words: hierarchical porous carbon; pyrolysis; rice husk silica template

Hierarchical porous carbon (HPC), a class of porous carbon with interconnected multiscale pores, has attracted much research interest in recent years due to its structural advantages over traditional porous carbon materials (Xiang et al, 2022; Romero et al, 2024). Conventionally, HPCs are fabricated using unsustainable fossil fuel-based precursors. However, the growing demand for eco-friendly and sustainable materials has shifted research toward renewable and low-cost biomass sources as alternative carbon precursors (He et al, 2023). Researchers have prepared HPC and porous carbon materials from different biomass sources with excellent characteristics. Some examples include coconut shells, açaí seeds, bagasse, kelp, walnut shells, and *Mikania micrantha* (Sun et al, 2019; de Souza et al, 2020; Liu H et al, 2020; Sanni et al, 2020; Mashhadimoslem et al, 2021; Zhao et al, 2023; Zou et al, 2023; Jalalah et al, 2024a). These materials have various applications in energy storage, water purification, gas adsorption, catalysts, and environmental remediation (Wang et al, 2020; Chakraborty et al, 2022; Jalalah et al, 2023a, 2024b). Although various biomass sources can be utilized to produce HPC, the availability, abundance, and supply need to be considered for sustainable manufacturing because the shortage of raw materials can impact the scalability and feasibility of production. Rice husk has emerged as a highly promising precursor for HPC synthesis due to its natural abundance and unique silica composition.

Rice (*Oryza sativa*) is a staple food for over half of the global population, with its demand steadily rising alongside population growth (Bandumula, 2018).

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According to the Food and Agriculture Organization data (FAO, 2023), global rice production reaches approximately 526 million tonnes annually (milled basis), resulting in the generation of millions of tons of rice husks each year. Open burning of rice husks is a common practice in many rice-producing countries, which emits harmful pollutants, greenhouse gases, and fine particulate matter, contributing to significant environmental and health issues (Junpen et al, 2018). Landfilling is another widespread method of rice husk disposal, but the slow decomposition rate of rice husks occupies valuable landfill space for extended periods (Zou and Yang, 2019). These challenges highlight the need for sustainable methods for rice husk waste management. Rice husks have several versatile and high-value applications in porous carbon materials, building materials, silica extraction, adsorbents, and soil amendments (Premaratne et al, 2014; Alam et al, 2020; Kordi et al, 2024).

HPCs derived from biomass are usually produced by pyrolysis-activation techniques (Zhou et al, 2021). However, rice husks are unlike the common lignocellulosic biomasses such as wood or straw, as they contain very high silica levels, typically ranging from 15%–20% by weight (Yerdauletov et al, 2023). Direct pyrolysis and activation of rice husk hardly produce well-developed hierarchical pore structures. Thus, additional processing steps such as silica removal or advanced activation methods are often required to achieve the desired hierarchical structure. The embedded silica nanoparticles in rice husks act as a natural template and play a crucial role in forming hierarchical pore structures (Rybarczyk et al, 2016). The pore size, distribution, and interconnectivity depend heavily on preparation techniques and associated process conditions. Understanding these factors is essential for optimizing HPC properties and enhancing their performance across diverse applications.

Comprehensive reviews have been published on the preparation and synthesis of HPCs from biomass sources (Cuong et al, 2021; Zhou et al, 2021). However, to the best of the authors' knowledge, no review has specifically examined the preparation of HPC from rice husks or the role of silica as a natural template. The current review focuses on synthesising HPC from rice husks, emphasizing the significance of silica as a natural template. Moreover, various synthesis methods and techniques, including pyrolysis, silica removal methods, and chemical and physical activation, are discussed. Finally, the applications and prospects of rice husk-derived HPCs are comprehensively summarized.

Hierarchical porous structure

According to the International Union of Pure and Applied Chemistry, pores are categorized into three types based on their size: micropores (< 2 nm), mesopores (2–50 nm), and macropores (> 50 nm). HPC contains interconnected multi-modal pore structures ranging from micropores to mesopores and macropores, as schematically depicted in Fig. 1-A. Generally, HPCs have a large specific surface area (SSA); however, a high SSA alone does not confirm the presence of a hierarchical porous structure, as sometimes conventional porous/activated carbon exhibits high SSA due to abundant micropores (Xiang et al, 2022; Yan et al, 2023). The defining feature of HPC compared with other porous carbon materials is the interconnectivity of these different pore sizes, forming a hierarchical architecture. Micropores provide a high SSA and numerous adsorption sites, while mesopores act as efficient pathways for ion transport, facilitating rapid diffusion to the interior surfaces where micropores are located (Zhang M X et al, 2021; Zhou et al, 2021). Additionally, macropores act as reactant reservoirs, minimizing diffusion distance and enhancing mass transfer kinetics (Kaur et al, 2019). Interconnected pores across various size ranges contribute synergistically to adsorption capacity, ion transport efficiency, and reactant accessibility, leading to superior performance in energy storage, catalysis, and adsorption applications (Cuong et al, 2021).

Many activated carbon materials have been produced from rice husks using conventional physical and chemical activation techniques without leveraging the natural silica templating effect. Carbon materials prepared by these methods often lack a well-developed hierarchical pore structure or high SSA and predominantly contain one type of pore, typically micropores. These micropores contribute significantly to the adsorption surface area; however, their small size restricts the movement of electrolyte ions, leading to slow diffusion and a sharp decline in capacitance at higher current densities in supercapacitors (Yan et al, 2023). Moreover, not all micropores are readily accessible to ions due to their size and tortuosity, reducing the effective surface area (Yin et al, 2020; Jiang et al, 2022). This restricts their applicability in advanced applications, and they are usually used as adsorbents. In contrast, rice husk-derived HPC is specifically designed to possess a hierarchical architecture. HPC preparation from rice husk often involves a silica template removal step after carbonization,

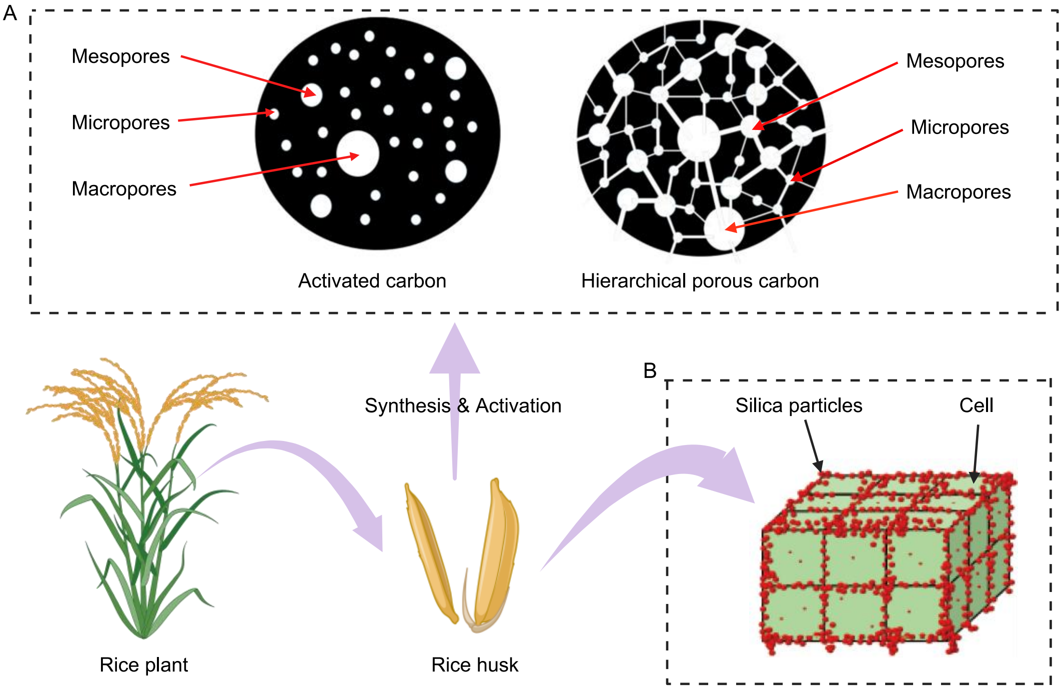

Fig. 1. Structural comparison of activated carbon and hierarchical porous carbon, and silica distribution in rice husk cells.
A, Schematic diagram of hierarchical porous and activated carbon derived from traditional physicochemical activation methods.
B, Schematic representation of silica distribution in rice husk cells.

leveraging the silica content as a natural template (Liu et al, 2015; Cuong et al, 2020).

Components of rice husk

Rice husks serve as the protective outer layer of rice grains, regulating moisture within the grain and preventing excessive water loss that could hinder proper development (Kordi et al, 2024). Rice husk consists of three main layers: the epidermis (the outermost layer, often featuring macro-hairs on its surface), the subhypodermis, and the hypodermis (Chen et al, 2018). Rice husk possesses vascular bundles that form natural microporous channels varying in size between 2 and 10 µm (Liu et al, 2015). These channels facilitate the transport of water and essential nutrients required for rice growth. The chemical composition of rice husk varies depending on the rice variety, growing region, and cultivation conditions (Alam et al, 2020). It primarily consists of cellulose (35%–40%), hemicellulose (15%–20%), lignin (20%–25%), and silica (15%–25%) (Gao et al, 2018; Yerdauletov et al, 2023). Cellulose, the main structural component, is a complex polysaccharide primarily composed of glucose monomers connected through β-1,4-glycosidic linkages. Hemicellulose is another carbohydrate component present in plant cell walls. Lignin, a complex organic polymer, enhances the strength and rigidity of plant tissues.

The primary inorganic component in rice husks is silica, which exists in an amorphous form. Rice roots absorb silica as silicic acid, which is transported to the outer epidermal walls and transformed into silica (Yamaji and Ma, 2009). Analysis using scanning electron microscopy and energy-dispersive X-ray spectroscopy revealed that silica is unevenly distributed within rice husks, with the highest concentration found on the outer layer, forming a protective coating (Chen Z M et al, 2020). Fig. 1-B shows a schematic diagram of silica distribution inside the rice husk. A significant fraction of silica particles ranges from 1 to 7 nm in size, while a smaller fraction consists of larger particles, ranging from 8 to 22 nm (Larichev et al, 2015). Silica forms a 3D nanostructured skeleton within the rice husk, acting as a natural template alongside organic components. Silica and organic components in rice husks mutually serve as structural templates (Chen Z M et al, 2020).

Preparation of HPC

HPC can be synthesized through various methods (Dutta et al, 2014; Kaur et al, 2019; Xiang et al, 2022; Yan et al, 2023), including template-based and template-free approaches. Template techniques are divided into hard-template and soft-template methods. Hard-template methods involve impregnating rigid templates (e.g., silica or metal oxides) with a carbon precursor. After carbonization, the template is removed via acid etching, leaving a carbon material with pores replicating the template's structure. The soft-template method employs self-assembling surfactants or block copolymers as templates to direct the organization of the carbon precursor. The dual-template approach integrates both hard and soft templates to create a well-defined, multi-scale pore structure. Template-free methods include pyrolysis activation, chemical vapor deposition, and other emerging techniques. Most biomass inherently contains naturally organized structures ranging from molecular to tissue levels. Pyrolysis, followed by activation, effectively utilizes these natural features to produce HPC, making it an attractive and sustainable approach for waste biomass conversion (Zhou et al, 2021).

Preparation of HPC from rice husk

Recent studies on preparing rice husk-based HPC are summarized in Table 1. The process generally involves carbonization of rice husk, removal of silica templates, and activation. The hierarchical porosity achieved depends on factors such as activation temperature, duration, and the rice husk precursor variety. There is a lack of comprehensive studies comparing the influence of rice husk variety, composition, and cultivation practices on the structural and functional characteristics of the synthesized HPC. To the best of the authors' knowledge, no study has specifically identified the optimal rice husk type for HPC production. Further research is needed to analyze how these rice husk compositional variations influence HPC synthesis.

The hierarchical porous structure of rice husk-derived HPC emerges through a sequence of structural transformations during carbonization, silica removal, and activation. Liu et al (2015) proposed a mechanism of hierarchical pore formation from rice husks. During carbonization, organic components in the rice husk decompose, releasing volatile matter, and leaving behind a carbon-silica composite. This stable porous structure preserves the vascular bundles of the rice husk. The

Table 1. Preparation methods of rice husk-based hierarchical porous carbon.

Carbonization process	Silica removal	Activation process	Impregnation ratio	SSA (m^2/g)	PV (cm^3/g)	Mesoporosity (%)	Reference
Pyrolysis at 800 °C for 1 h under N_2 atmosphere	HF leached	Steam activation at 900 °C for 3 h	–	1 340	1.20	–	Tabata et al, 2010
Pyrolyzed at 500 °C for 3 h under N_2 atmosphere	HF leached	KOH activation at 800 °C for 2 h	4:1	1 094	0.61	42.6	Wang et al, 2014
Pyrolysis at 500 °C for 1 h under N_2 atmosphere	NaOH leached	KOH activation at 700 °C for 1 h	4:1	2 804	1.79	–	Liu et al, 2015
Hydrothermal carbonization	NH_4HF_2 leached	Higher pyrolysis	–	525	0.48	76.4	Rybarczyk et al, 2016
Pyrolysis at 500 °C for 1 h under N_2 atmosphere	NaOH leached	KOH activation at 700 °C for 2 h	4:1	1 751	1.11	55.9	Yuan et al, 2016
Pyrolyzed under Ar atmosphere	HF leached	Higher pyrolysis	–	533	0.39	–	Fang et al, 2017
Pyrolysis at 700 °C for 3 h under N_2 atmosphere	HF leached	KOH activation at 700 °C for 3 h	3:1	2 242	1.33	–	Xiao et al, 2017
Pyrolysis at 500 °C for 1 h under N_2 atmosphere	Base leached	NaOH/KOH co-activation at 800 °C for 1 h	3:1	2 747	1.40	–	Wang C et al, 2018
Pyrolyzed at 500 °C for 1 h under N_2 atmosphere	NaOH leached	KOH/CO_2 physicochemical activation	3:1	2 330	1.32	81.0	Cuong et al, 2019
Pyrolysis at 900 °C for 1 h under N_2 atmosphere	HF leached	Steam activation at 850 °C	–	268	0.23	–	Kim et al, 2019
Pyrolysis at 450 °C for 3 h under N_2 atmosphere	–	KOH activation at 750 °C for 1 h	3:1	1 320	0.65	30.7	Shen et al, 2019
Pyrolysis at 600 °C for 1 h under N_2 atmosphere	NaOH leached	$CuCl_2$ activation at 800 °C	10:1	1 339	0.80	33.7	Tian et al, 2019
Pyrolysis at 600 °C for 1 h under N_2 atmosphere	NaOH leached	NaOH activation at 750 °C for 1 h	2.5:1.0	1 789	1.15	37.4	Chen Z M et al, 2020
Pyrolyzed at 800 °C for 2 h under N_2 atmosphere	NaOH leached	KOH activation at 800 °C	2:1	1 839	1.21	58.0	Cuong et al, 2020
Pyrolysis	NaOH leached	Higher pyrolysis	–	242	0.15	–	Nie et al, 2020
Hydrothermal carbonization	NaOH leached	–	–	445		–	Hou et al, 2021
Pyrolysis	Na_2CO_3 leached	NaOH activation at 800 °C	3:1	2 686	1.62	64.0	Zhang S P et al, 2021
Pyrolysis at 1000 °C for 2 h under N_2 atmosphere	NaOH leached	–	–	643	0.52	–	Qin et al, 2023

Impregnation ratio, Mass ratio of activating agent to rice husk; SSA, Specific surface area; PV, Total pore volume; Mesoporosity (%), Ratio of mesopore volume to PV; HF, Hydrofluoric acid. '–' indicates data not mentioned in the original paper.

composite initially has a small surface area. Base leaching removes the nano-silica, resulting in a templated carbon characterized by a 3D surface structure and parallel internal vascular channels. Chemical activation then gradually transforms this templated carbon into a hierarchical porous structure. At the start of activation, activators accumulate in the porous channels of the nano-structured carbon via capillary interaction. As activation proceeds, these channels widen into macropores and micropores, followed by the development of mesopores within them. Eventually, these pores become interconnected, forming a hierarchical porous framework.

The optimal HPC production method depends on the target application, cost, performance, and environmental impact. Overall, the most common approach combines pyrolysis, NaOH leaching for silica removal, and KOH/NaOH activation, which yields well-developed hierarchical porosity and superior performance.

Carbonization of rice husk

Carbonization is a process that involves the thermal decomposition of carbonaceous materials by heating under oxygen-depleted conditions. Generally, carbonization can be conducted through pyrolysis or hydrothermal carbonization. Optimizing carbonization parameters is important because this process significantly affects the properties of the resulting carbon materials.

Pyrolysis of rice husk

Pyrolysis of rice husks is the most widely used carbonization method for producing HPC due to its simplicity and scalability. The process involves heating rice husks in an inert environment (nitrogen or argon, N_2/Ar) and yields three main products: biochar, a solid carbon-rich material; bio-oil; and non-condensable gases (e.g., methane, hydrogen, carbon monoxide, and carbon dioxide) (Asadi et al, 2021). Jia et al (2020) studied rice husk pyrolysis characteristics using thermogravimetric analysis and identified four stages: Drying stage (20 ºC–110 ºC) releases moisture and breaks hydrogen bonds in macromolecular structures, causing approximately 10% mass loss. Preheating transition stage (110 ºC–220 ºC) involves hemicellulose depolymerization and recombination, resulting in a further mass loss of around 3%. Main pyrolysis stage (220 ºC–600 ºC) involves significant thermal decomposition of rice husks, breaking carbohydrate chains into simpler components. Carbonization stage (above 600 ºC) decomposes the remaining lignin slowly, resulting in a gradual mass loss. At temperatures above 750 ºC, silica facilitates carbon graphitization (Yerdauletov et al, 2023). Above 900 ºC, silica crystallinity increases, evidenced by sharper X-ray diffraction peaks (Barakat et al, 2024). Pyrolysis primarily forms micropores in rice husk-derived porous carbons, and the resulting carbon-silica composite exhibits a low surface area.

Hydrothermal carbonization (HTC)

HTC involves heating biomass in water (180 ºC–250 ºC) under pressure for several hours (Bushra and Remya, 2024). This energy-efficient method can process rice husks without pre-drying. During HTC, rice husks undergo dehydration, decarboxylation, and polymerization (Kaur et al, 2019), yielding hydrochar rich in oxygen-containing functional groups (Nizamuddin et al, 2018).

Removal of silica template

Chemical agents for silica removal

Silica removal is typically achieved through chemical etching, and the most used agents include hydrofluoric acid (HF) and sodium hydroxide (NaOH). HF effectively removes silica at room temperature (Tabata et al, 2010). Xiao et al (2017) reported near-zero silicon content after 25% HF treatment (confirmed by X-ray photoelectron spectroscopy). However, HF is toxic, corrosive, expensive, and hazardous in waste management (Sevilla et al, 2021). NaOH reacts with silica to form soluble sodium silicate (Na_2SiO_3) (Alam et al, 2020). Xue et al (2019) found that 5% NaOH at room temperature removes surface silica, but higher concentrations or elevated temperatures are needed for complete removal. Compared with HF leaching, NaOH treatment is less efficient and energy-intensive at high temperatures.

Ammonium bifluoride (NH_4HF_2) and sodium carbonate (Na_2CO_3) have been investigated as alternative agents for silica removal. NH_4HF_2 dissociates to release HF *in situ*, reducing direct HF handling but still poses hazards (Rybarczyk et al, 2016). Na_2CO_3 is less corrosive, posing fewer environmental hazards than HF and NaOH (Sevilla et al, 2021), which achieves desilication at 90 ºC (Zhang S P et al, 2021), though the process is energy-intensive.

Effect of silica removal

Silica removal increases SSA and pore volume (Chen W et al, 2020). Qin et al (2023) reported that a carbon-silica composite, before silica removal, exhibits an SSA of approximately 30.26 m^2/g along with a pore volume of 0.05 cm^3/g. After silica removal, the SSA exceeds 159.65 m^2/g, while the pore volume significantly improves to around 0.24 cm^3/g. Zhang S P et al (2021) reported that before desilication, the mesopore volume reaches 0.03 cm^3/g, representing 24.62% of the overall pore volume, and after silica removal, the mesopore volume rises to 0.14 cm^3/g, constituting 59.31% of the total pore volume. These enhancements are due to the size of silica nanoparticles within rice husk closely matching the diameters of mesopores and macropores (Park et al, 2003). Upon removal, it promotes the development of pores, particularly mesopores, leading to a hierarchical structure (Rybarczyk et al, 2016). Rice husk-derived HPCs, following the removal of the silica template, display a marked improvement in supercapacitor performance, achieving a maximum specific capacitance of 429 F/g at 0.5 A/g, along with excellent rate capability (Xiao et al, 2017). In comparison, HPCs without silica removal exhibit a significantly lower specific capacitance of 364 F/g at 0.5 A/g and poor rate capability. This enhancement is primarily due to the enhanced porous structure resulting from silica removal, which increases the available surface area for charge storage and enhances ion transport efficiency within the electrode material. Silica removal also alters the morphology of rice husk char. While the char retains the vascular bundle and ridge-like structure of the original rice husk, its surface becomes rougher due to the newly exposed pores (Rybarczyk et al, 2016).

Silica can react with some activating reagents, like NaOH and KOH, which can reduce the activation process efficiency (Wang Z F et al, 2018; Zhang S P et al, 2021). Liu et al (2019) investigated how silica influences the activation process, comparing rice husk samples, RHC1 (rice husk without silica removal) and RHC2 (rice husk with silica removal via NaOH leaching). Results showed that in RHC1, silica reacts preferentially with KOH, forming potassium silicate. The potassium silicate forms a ‘shell’ around the carbon, hindering KOH from reacting with it. This hinders the enlargement of pores in the carbon. This process leads to many micropores being formed instead of mesopores. As a result, the activated carbon obtained through this method exhibits a high SSA of 3 263 m^2/g, resulting in a low mesopore ratio (36.1%). This produces a high specific capacitance of 315 F/g and a high energy density. However, the low mesopore ratio leads to poor rate capability. Conversely, in RHC2, removing the silica from the rice husk before activation generates initial pores within the material. During the activation process, these pores enlarge into mesopores. The carbon material obtained through this process has a high mesopore ratio (73.5%), resulting in excellent rate performance, maintaining 77% capacitance retention as the current density rises from 0.5 to 20 A/g.

Activation of rice husk

Rice husk char can be activated through chemical, physical, and physicochemical methods. Activation temperatures, agents, time, and process parameters vary depending on the technique.

Physical activation

Physical activation involves treating rice husks with oxidizing gases, such as CO_2 or steam, at high temperatures. The gases react with the carbon atoms in the rice husk char structure, forming new pores as carbon atoms are removed. Simultaneously, pores formed during the carbonization stage are widened (Menya et al, 2018; Yin et al, 2020). The main chemical reactions involved are:

$$C\ (s) + 2H_2O\ (g) \rightarrow 2H_2\ (g) + CO_2\ (g)$$
$$C\ (s) + H_2O\ (g) \rightarrow H_2\ (g) + CO\ (g)$$
$$C\ (s) + CO_2\ (g) \rightarrow 2CO\ (g)$$
$$C\ (s) + O_2\ (g) \rightarrow CO_2\ (g)$$

Physical activation of rice husk char with CO_2 under different temperature conditions revealed that weight loss rises with higher activation temperatures, primarily due to carbon loss (Wazir et al, 2024). Physical activation is generally considered a greener option than chemical activation because it avoids the use of harsh and potentially hazardous chemicals. The main byproducts are gases that can be managed more easily than the wastewater generated during chemical activation. However, some drawbacks exist. Compared with chemical activation, physically activated carbon often has a lower SSA, limiting its effectiveness in applications requiring high adsorption capacity. It also requires elevated temperatures, contributing to significant energy consumption, which increases operational costs and offers less control over pore size distribution than chemical activation. This can be a disadvantage for applications that require specific pore characteristics for optimal performance.

Chemical activation

Chemical activation is a widely used method for producing porous carbon, employing chemical reagents as activating agents. This involves impregnating rice husks with chemical agents and heat treatment in an inert atmosphere. Commonly used reagents for chemical activation of rice husks include KOH, NaOH, $ZnCl_2$, H_3PO_4, and $CuCl_2$. Compared with other chemical agents, KOH is renowned for its superior performance, as it generally requires lower temperatures, produces higher biochar yield, and can result in high SSA porous carbon, reaching up to 3 000 m^2/g (Chen W et al, 2020). The KOH activation mechanism involves complex chemical reactions (Yin et al, 2020). The major reactions involved are:

$6KOH (s) + 2C (s) \rightarrow 2K (l) + 3H_2 (g) + 2K_2CO_3 (s)$

$K_2CO_3 (s) + 2C (s) \rightarrow 2K (l) + 3CO (g)$

$K_2CO_3 (s) \rightarrow K_2O (s) + CO_2 (g)$

$K_2O (s) + C (s) \rightarrow 2K (l) + CO (g)$

KOH reacts with carbon in the rice husk to form K_2CO_3, metallic potassium, hydrogen gas, and carbon monoxide. Gaseous intermediates generated, such as CO_2, H_2O, and H_2, escape from the biochar structure, creating a network of pores. CO_2 and H_2O also serve as activating agents during physical activation, further enhancing pore development. Metallic potassium can intercalate into the carbon lattice, expanding the structure and creating micropores (Xiang et al, 2022).

Several key factors, including the impregnation ratio, activation temperature, and activation time, influence the activation process for preparing HPC. Cuong et al (2020) prepared HPC from rice husks using a KOH impregnation ratio of 2:1 (KOH:rice husk) and an activation temperature of 800 ºC, demonstrating significant variations in structural properties based on activation time. When the activation time was extended from 0.5 to 2 h, the SSA of the HPC materials increased significantly from 1 520 to 1 839 m^2/g. However, further increasing the activation time to 3 h resulted in the SSA and pore volume reducing to 1 702 m^2/g and 1.14 cm^3/g, respectively. This decrease is attributed to the collapse of micropores, likely due to prolonged exposure to harsh activating conditions. A study reported that higher impregnation ratios, such as 3:1, enhance SSA and mesoporosity, with SSA increasing from 1 734 m^2/g at 2:1 to 2 330 m^2/g at 3:1 under identical conditions (Cuong et al, 2019). However, excessive ratios lead to pore collapse due to overreaction with carbon, reducing mesoporosity. Similarly, higher activation temperatures improve porosity and mesopore development, as seen with an increase in mesopore-to-total surface area ranging from 0.23 at 600 ºC to 0.73 at 800 ºC. Excessive temperatures, however, weaken the carbon framework, causing structural collapse. These findings highlight the need to optimize both parameters to balance porosity and structural integrity.

NaOH is another commonly used activating agent. The reactions involved during activation can be represented by the following equations (Heidarinejad et al, 2020):

$6NaOH (s) + 2C (s) \rightarrow 2Na (l) + 2Na_2CO_3 (s) + 3H_2 (g)$

$Na_2CO_3 (s) \rightarrow Na_2O (s) + CO_2 (g)$

$2Na (l) + CO_2 (g) \rightarrow Na_2O (s) + CO (g)$

Sodium can intercalate into the carbon structure, expanding the spaces between carbon atoms and contributing to pore development. Additionally, gases like CO, CO_2, and H_2 are released when NaOH reacts with carbon materials, creating pores within the carbon structure. However, variations in activation mechanisms result in porous carbons with lower surface areas when produced via NaOH activation compared with KOH activation.

Unlike traditional activators such as KOH and NaOH, $CuCl_2$ minimizes reactor corrosion and achieves an 80% recovery rate, enhancing sustainability (Tian et al, 2019). However, $CuCl_2$-activated porous carbon exhibits lower surface areas than KOH- and NaOH-activated HPCs.

Physicochemical activation

Physicochemical activation combines chemical and physical activation agents. According to Table 1, physicochemical activation achieves good performance metrics, including a high SSA of 2 330 m^2/g and significant mesoporosity, with a mesopore ratio of 81%. While physicochemical activation offers these advanced characteristics, it is more complex than single activation methods, requiring greater investment in equipment and reagents.

Other methods for preparation of rice husk-derived HPCs

Liang et al (2017) developed a method for synthesizing HPCs from rice husks that eliminates both activation and post-treatment steps by combining rice husks with polytetrafluoroethylene (PTFE) powder and carbonizing the mixture. During carbonization, PTFE decomposes and releases HF, which *in situ* etches away the silica present in rice husks. This single-route carbonization and silica removal process is environmentally friendly and cost-effective since it eliminates the need for additional steps and hazardous chemicals like HF. The resulting HPCs exhibit high SSA, reaching up to 2 051 m^2/g and a high specific capacitance of 317 F/g at a current density of 50 A/g. N_2 adsorption-desorption isotherms confirm the existence of a hierarchical pore structure consisting of micropores, mesopores, and macropores. Future research can investigate the scalability of this process for industrial applications.

Peng et al (2019) proposed an innovative strategy to fabricate HPCs using mixed biomass wastes, specifically crab shells and rice husks, as raw materials. In this method, a mixture of crab shells and rice husks was hydrothermally carbonized, and HCl was used to soak and remove $CaCO_3$, followed by KOH. The resultant HPCs showed a unique 3D interconnected pore architecture with a large surface area of 3 557 m^2/g, facilitating electrolyte ion transport and offering abundant energy storage capacity. Additionally, the HPCs exhibit a high specific capacitance of 474 F/g and excellent cyclic stability, retaining 95.6% of their capacitance over 20 000 charge/discharge cycles while employing 6 mol/L KOH as the electrolyte.

Molten salt carbonization is a one-step process that converts biomass into functional biochar using molten salt, which acts as a pyrolysis medium and a chemical activating agent for the biomass (Egun et al, 2022). This allows for carbonization and activation to occur simultaneously, streamlining the production of functional biochar. Liu et al (2023) reported the production of HPCs from rice husks via a single-step Na_2CO_3 and K_2CO_3 molten carbonization process. The salt bath acts as a chemical activating reagent, and the alkaline melt also dissolves the silica templates within the rice husks, eliminating the need for additional steps for silica template removal. The resultant HPC has an SSA of 1 012.8 m^2/g and a hierarchically porous structure, as demonstrated by isotherms and pore size distribution curves. Using eutectic mixtures of multiple salts can lower the melting point compared with single-component salts, and the pore properties of carbon materials can be tuned by altering the composition and ratios of the composite salts in the molten mixture (Yan et al, 2023). However, the corrosive nature of molten salts, especially at high temperatures, can challenge reactor materials and equipment. Selecting appropriate corrosion-resistant materials for reactors and handling systems is essential to ensure process longevity and safety.

Cheng et al (2020) fabricated HPCs by pre-treating rice husks with cellulose-degrading fungus. This treatment enhances carbonization and chemical activation efficiency by breaking down the rice husks' internal and external microstructures while altering the lignin, cellulose, and hemicellulose compositions. The resulting HPC material exhibits an SSA of 3 714 m^2/g and a hierarchical micro-mesoporous structure. It demonstrated remarkable adsorption properties, including a high toluene adsorption capacity of 708 mg/g at 100 mg/kg, outperforming porous carbons derived from untreated rice husks (538 mg/g).

Using a novel two-step KOH activation method, Li et al (2024) prepared HPCs from rice husks. The process involves carbonization of rice husks to biochar, followed by a pre-activation step at 400 ºC under nitrogen flow and a final activation at 800 ºC. The sample produced through this method is labelled RHAC-Tpre400-K3. For comparison, a control sample was prepared using a conventional single-step KOH activation method and labelled RHAC-K3. Compared with traditional single-step KOH activation, the optimized method (RHAC-Tpre400-K3) achieves a well-developed 3D hierarchical porous structure with superior specific capacitance performance relative to RHAC-K3. In pre-activation, additional activation sites are created by KOH reactions. These sites expand during the subsequent 800 ºC activation, facilitating the formation of interconnected pores. Heteroatom doping involves incorporating elements other than carbon into the HPC structure, which can alter its physicochemical and electronic characteristics, enhancing its applicability across various applications. Xue et al (2020) prepared nitrogen-doped rice husk-derived HPC by the pyrolysis of urea-impregnated rice husk and then leaching out the silica template using HF. The resulting materials display a high gravimetric capacitance of 242 F/g and volumetric capacitance of 306 F/cm^3 at 0.5 A/g in 1 mol/L H_2SO_4 electrolyte, surpassing the performance of activated carbon synthesized via KOH or $ZnCl_2$ activation.

Comparison with traditional method

Recent studies on preparing porous carbon/activated carbon from traditional physical and chemical activation are summarized in Table 2. Although traditional chemical activation can achieve high surface area and pore distribution, porous carbon lacks hierarchical architecture. The general procedure for preparing porous carbon/activated carbon typically includes pyrolysis and/or activation of rice husks. Sometimes, acidic or basic pretreatments are employed to reduce the metallic impurities and ash content in rice husks (Barakat et al, 2024).

Applications of rice husk-derived HPCs

Rice husk-derived HPCs perform better in diverse applications due to their high SSA, interconnected porous structure, and unique physicochemical properties. The most prominent area of rice husk-derived HPC application is energy storage, specifically supercapacitors. The supercapacitor functionality of HPC from rice husks depends on electrical double-layer capacitance (EDLC) and pseudo-capacitance mechanisms (Zhou et al, 2021). In EDLC, the charge is stored through the electrostatic adsorption and desorption of ions at the electrode-

Table 2. Recent studies on preparation of activated carbon through traditional method using rice husks.

Pre-treatment	Carbonization process	Activation process	Impregnation ratio	SSA (m^2/g)	PV (cm^3/g)	Reference
Base leached	406 °C for 1 h under N_2 atmosphere	Steam	–	1 016	0.58	Menya et al, 2018
Not leached	Pyrolyzed at 400 °C	KOH	1:3	887	0.48	Liu Z Y et al, 2020
Not leached	Pyrolyzed at 500 °C for 3 h under N_2 atmosphere	KOH	1:3	2 087	0.99	Lv et al, 2020
Not leached	Pyrolyzed at 400 °C for 4 h under N_2 atmosphere	NaOH	1:4	429	0.29	Saad et al, 2020
Not leached	Pyrolyzed at 500 °C for 1 h under N_2 atmosphere	KOH	1:2	890	0.44	He et al, 2021
Not leached	Pyrolyzed at 450 °C under N_2 atmosphere	NaOH	1:3	2 755	1.54	Silva et al, 2021
Not leached	Pyrolyzed at 600 °C for 2 h	KOH	1:3	755	0.39	Nandi et al, 2023
Not leached	Pyrolyzed at 750 °C under N_2 atmosphere	KOH	1:4	3 050	1.10	Yerdauletov et al, 2023
Not leached	Pyrolyzed at 400 °C under N_2 atmosphere	CO_2	–	502	0.12	Lesbayev et al, 2024
Not leached	Pyrolyzed at 900 °C for 1 h under N_2 atmosphere	CO_2	–	502	0.12	Wazir et al, 2024

Impregnation ratio, Mass ratio of activating agent to rice husk; SSA, Specific surface area; PV, Total pore volume. '–' indicates data not mentioned in the original paper.

electrolyte interface, thus creating an electric double layer without any faradaic reaction (Tian et al, 2019; Zhang S P et al, 2021). Their high SSA provides abundant sites for electrolyte ion accumulation, leading to high capacitance, and an interconnected network of pores facilitates rapid ion transport, enhancing rate capability and power density (Liu et al, 2015; Liang et al, 2017; Wang Z F et al, 2018). Oxygen-containing groups, such as hydroxyl, carboxyl, and carbonyl, are commonly found in HPCs and can enhance wettability, promoting electrolyte accessibility to the pore structure (Liu et al, 2015). Pseudo-capacitance arises from fast redox reactions at electroactive sites. Integrating metal oxides into HPC surfaces can enhance pseudo-capacitance through additional reversible redox reactions (Yuan et al, 2016). Nitrogen doping can introduce functional groups like pyridinic and quaternary nitrogen, enhancing electrical conductivity and contributing to pseudo-capacitance (Xue et al, 2020). The capacitance performances of rice husk-derived HPCs and other biomass-derived HPCs are shown in Table 3. Rice husk-derived HPCs has similar and sometimes higher performance than other biomass-derived HPCs.

Rice husk-derived HPC has been studied as an electrode material for lithium-ion, sodium-ion, and lithium-sulfur batteries. In lithium-ion batteries, the porous structure can provide a large SSA and accommodate the volume changes during lithium insertion and extraction, leading to high specific capacity and stable cycle performance (Hou et al, 2021). Similarly, in sodium-ion batteries, HPC demonstrates a low charge/discharge voltage platform and high SSA, promoting efficient sodium-ion diffusion and improved electrochemical stability (Nie et al, 2020). In lithium-sulfur batteries, HPC's interconnected mesoporous and microporous network functions as a conductive scaffold for sulfur loading, enabling better sulfur utilization and mitigating polysulfide shuttling (Rybarczyk et al, 2016).

Table 3. Electrochemical performance of rice husk-derived hierarchical porous carbon (HPC) and other biomass-derived HPCs in supercapacitor applications.

Biomass	Activation	SSA (m^2/g)	Test system/Electrolyte	Current density (A/g)	Specific capacitance (F/g)	Reference
Rice husk	KOH	2 804	6 mol/L KOH, Two-Electrode	0.5	278	Liu et al, 2015
Rice husk	PTFE	2 051	6 mol/L KOH, Two-Electrode	10	245	Liang et al, 2017
Rice husk	KOH	2 242	6 mol/L KOH, Two-Electrode	0.5	429	Xiao et al, 2017
Rice husk	NaOH-KOH	2 747	1 mol/L H_2SO_4, Two-Electrode	0.5	194.6	Wang C et al, 2018
Rice husk	$CuCl_2$	1 339.9	6 mol/L KOH	0.5	165.2	Tian et al, 2019
Rice husk	KOH	1 839	1 mol/L NaCl, Three-Electrode	0.1	120.5	Cuong et al, 2020
Rice husk	NaOH	1 789	6 mol/L KOH, Two-Electrode	0.5	256	Hou et al, 2021
Rice husk	NaOH-KOH	3 046	6 mol/L KOH, Two-Electrode	0.2	312	Zhang S P et al, 2021
Withered rose	KOH/KNO_3	1 911	6 mol/L KOH, Three-Electrode	0.5	208	Zhao et al, 2018
Acai seed	KOH	3 846	1 mol/L KOH, Three-Electrode	1.0	346	de Souza et al, 2020
Olive wood	KOH	1 352	1 mol/L H_2SO_4, Two-Electrode	0.125	231	Elmouwahidi et al, 2020
Hibiscus sabdariffa fruit	KOH	1 720.5	2 mol/L KOH, Three-Electrode	0.5	194.5	Hamouda et al, 2021
Foxtail grass seed (N and S co-doped)	$NaHCO_3/KHCO_3$	819	6 mol/L KOH	0.5	358	Liang et al, 2021
Bagasse	KOH	3 135	6 mol/L KOH, Two-Electrode	0.5	410.5	Tan et al, 2021
Enhydra fluctuant leaf	KOH	1 082	1 mol/L H_2SO_4, Three-Electrode	1.0	428	Jalalah et al, 2023b
Coconut shell	KOH	2 143.6	6 mol/L KOH, Three-Electrode	0.5	317	Zhao Y et al, 2023
Turmeric leaf	NH_4Cl/KOH	541	0.5 mol/L H_2SO_4, Three-Electrode	1.0	389	Chakraborty et al, 2024

Rice husk-derived HPC is a promising material for water purification and environmental remediation, particularly in the adsorption of heavy metals and organic compounds and capacitive deionization (CDI). HPCs' high surface area and functional groups facilitate the efficient adsorption of heavy metal ions from aqueous solutions. Cuong et al (2019) reported an HPC material exhibiting an SSA of 2 330 m^2/g and an 81% mesopore-to-total surface area ratio, with an adsorption capacity of 265 mg/g and efficient adsorption kinetics for copper ions. CDI is a process that uses a pair of electrodes with opposite charges to generate an electrostatic double layer, which captures and removes ions from water (Kim et al, 2019; Siriwardane et al, 2021). In CDI, researchers have reported that rice husk-derived HPC-based electrodes can remove inorganic ions such as NH^{4+}, Mg^{2+}, and Cu^{2+}, exhibiting high electro-sorption capacities and requiring low energy for water desalination (Cuong et al, 2020).

Tabata et al (2010) studied rice husk-derived HPC for *in vivo* applications. Rats with pneumonia-induced renal failure were fed 5% HPC for 3 d. The rats' plasma analysis revealed a remarkable decrease in uremic toxins after HPC administration. It demonstrated greater effectiveness in decreasing uric acid levels than activated carbon, a commonly used adsorbent. The study also concluded that HPC has low toxicity, as demonstrated by a separate study on normal rats. The rats showed no significant weight changes after consuming feed containing 5% HPC for 2 weeks.

The structural adaptability and physicochemical properties of husk-derived HPC also make them suitable for electromagnetic wave absorption applications (Fang et al, 2017). These materials exhibit excellent absorption performance, positioning them as promising candidates for electromagnetic shielding in electronics and communication devices.

Future perspectives and potential applications

Researchers have explored HPC from biomass sources other than rice husk for gas sensors, photocatalysis, and advanced biomedical applications (Sun et al, 2016; Zhou et al, 2021). Although such applications have not been extensively studied for rice husk-derived HPC, its hierarchical porosity, surface functionalization potential, and stability make it an excellent candidate for emerging applications. In addition, hybrid structures combining rice husk-derived HPC with nanomaterials can enhance overall material performance. HPC-based polymer composites may improve mechanical strength and chemical resistance, enabling their use in membranes, coatings, and hydrogen storage applications. Functionalized rice husk-based-HPC can also support CO_2 capture and conversion. While significant progress has been made, synthesizing HPC from rice husks presents opportunities for scientific and technological advancements. Future research on synthesizing HPC from rice husks should prioritize addressing current gaps and expanding its applications. A major focus area is replacing hazardous chemicals like HF in silica removal processes with safer and greener alternatives, such as sodium carbonate. This is a bottleneck in many HPC preparation studies because hazardous chemicals cause environmental damage. Further detailed studies on activation conditions, such as temperature, time, and activating agents, are necessary to better control pore size distribution and surface area. Innovations in physicochemical activation and hybrid approaches, including green activation methods like biological methods, should be explored to enhance the efficiency of HPC production. Many studies are limited to laboratory-scale experiments; therefore, pilot-scale and industrial-scale investigations are essential to evaluate the feasibility of rice husk-derived HPC production. Developing cost-effective and energy-efficient systems for large-scale synthesis is critical for commercialization. Rice husk-derived HPC has primarily been researched for energy storage applications and should be explored in a broader range of applications, such as catalysts and dye molecule adsorption. Carbon-based materials have been successfully incorporated to enhance textile properties (Gunasekera et al, 2015). Future research could focus on integrating HPC into textile materials for advanced multifunctional applications. Combining HPC production with other rice husk utilization pathways, such as silica extraction, can enhance resource efficiency and sustainability. By addressing these research directions, the synthesis of HPC from rice husks can advance toward achieving sustainable and high-performance material solutions for a wide range of global challenges.

Conclusions

Rice husk is a good candidate for the synthesis of HPC because of its low-cost availability and sustainability. The unique self-templating effect of silica in rice husks facilitates the formation of hierarchical porosity with a high SSA, well-interconnected pores, and tunable physicochemical characteristics suitable for the desired application. The general method of HPC preparation involves carbonization, silica removal, and subsequent physical or chemical activation. However, its commercialization faces several challenges, such as the environmental impact of silica removal, optimizing synthesis conditions, scalability, and significant energy consumption in synthesis processes. Novel emerging approaches have shown promise to address these challenges. With continued advancements in synthesis and functionalization innovations, rice husk-derived HPC could provide a sustainable and high-performance alternative to fossil fuel-derived carbons, addressing critical global challenges in the energy and environmental sectors.

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