

Review

Exploring the versatile production techniques and applications of nitrogen-doped activated carbon

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Abstract: Carbon based materials are really an integral component of our lives and widespread research regarding their properties was conducted along this process. The addition of dopants to carbon materials, either during the production process or later on, has been actively investigated by researchers all over the world who are looking into how doping can enhance the performance of materials and how to overcome the current difficulties. This study explores synthesis methods for nitrogen-doped carbon materials, focusing on advancements in adsorption of different pollutants like CO₂ from air and organic, inorganic and ions pollutants from water, energy conversion, and storage, offering novel solutions to environmental and energy challenges. It addresses current issues with nitrogen-doped carbon materials, aiming to contribute to sustainable solutions in environmental and energy sciences. Alongside precursor types and synthesis methods, a significant relationship exists between nitrogen content percentage and adsorption capacity in nitrogen-doped activated carbon. Nitrogen content ranges from 0.64% to 11.23%, correlating with adsorption capacities from 0.05 mmol/g to 7.9 mmol/g. Moreover, an electrochemical correlation is observed between nitrogen atom increase and specific capacity in nitrogen-doped activated carbon electrodes. Higher nitrogen percentage corresponds to increased specific capacity and capacity retention. This comprehensive analysis sheds light on the potential of nitrogen-doped carbon materials and highlights their significance in addressing critical environmental and energy challenges.

Keywords: CO₂ capture; doping agent; energy conversion; energy storage; raw material; synthesis methods

1. Introduction

Nitrogen-doped activated carbon, a sophisticated and innovative material, has emerged as a key player in the realm of environmental science and technology, particularly in the context of carbon dioxide (CO₂) adsorption [1]. This unique variant of activated carbon holds significant importance in addressing the escalating concerns related to global climate change and greenhouse gas emissions [2]. The integration of nitrogen into the carbon matrix enhances its adsorption capabilities, making it an invaluable tool in the pursuit of sustainable solutions. One of the pivotal roles of nitrogen-doped activated carbon lies in its exceptional capacity for CO₂ adsorption [3]. Carbon dioxide, a major greenhouse gas, contributes substantially to the warming of the Earth's atmosphere. The urgency to mitigate the adverse effects of climate change has intensified the search for efficient methods to capture and sequester CO₂ [4].

Nitrogen-doped activated carbon, with its tailored surface chemistry and increased porosity, provides an ideal substrate for the adsorption of CO₂ molecules [5]. The introduction of nitrogen functionalities enhances the affinity of the material for CO₂, making it a promising candidate for carbon capture applications [6]. The production methods employed in the synthesis of nitrogen-doped activated carbon play a crucial role in determining its structural properties and adsorption performance [7]. Typically derived from precursor materials such as biomass or synthetic polymers, the activation process involves subjecting the carbonaceous material to high temperatures in the presence of a nitrogen-containing gas [8]. This not only creates an activated carbon framework with a high surface area but also introduces nitrogen atoms into the carbon lattice [9]. The resulting material exhibits improved textural properties and enhanced surface functionality, contributing to its heightened CO₂ adsorption capacity [10]. Understanding the necessity of CO₂ capture becomes imperative in the context of climate change and its far-reaching consequences. The Earth's climate is intricately linked to the delicate balance of greenhouse gases in the atmosphere [11]. The alarming increase in CO₂ concentrations, the human activities such as the combustion of fossil fuels and the clearing of forests, are count as the main cause of the global temperature increases [12]. By capturing and sequestering CO₂, nitrogen-doped activated carbon offers a tangible and effective means to mitigate the impacts of climate change [12]. At all, nitrogen-doped activated carbon stands at the forefront of sustainable technologies, particularly in the vital realm of CO₂ adsorption [13]. Its tailored production methods and inherent structural properties make it a powerful tool in the quest for mitigating the impacts of climate change [14]. As the global community grapples with the imperative to reduce CO₂ emissions, the development and application of advanced materials like nitrogen-doped activated carbon underscore the potential for innovation to contribute to a more sustainable and environmentally conscious future [15].

2. Production of N-doped activated carbon

Nitrogen doped activated carbon (NACs) is already produced by scientists from different types of plant waste by various activation methods and they obtained N-doped AC with high surface area, suitable structure and pore size and huge adsorption capacity for uptake CO₂ [16]. The general synthesis principle of Nitrogen doped AC are classified in Chemical or direct activation also this method is known as in-situ synthesis [17], Physical activation [18], and hydrothermal method [13].

Chemical Activation with Nitrogen-Containing Compounds: In this method, the precursor material is impregnated with a nitrogen-containing compound, like urea [19], melamine [20], before carbonization and activation. The activation process is carried out at high temperatures, typically between 700–900 °C and adding some activation agent like KOH [21], ZnCl [22], ... This method introduces nitrogen atoms to the activated carbon structure [23].

Physical Methods: Physical activation is a widely used method for producing activated carbon. The procedure includes converting a precursor material into carbon through carbonization, followed by activation using a physical agent like steam or carbon dioxide [16]. Nitrogen doping can be achieved during the physical activation

process by introducing nitrogen-containing gases, such as ammonia [24] or other gases that contain nitrogen atoms [9]. The synthesis of nitrogen-doped activated carbons (N-ACs) through physical methods involves processes that modify the carbon structure without the use of chemical agents [10]. Various physical methods and associated nitrogen types include:

High-Temperature Pyrolysis: N-doped activated carbons are created through the pyrolysis of carbon precursors at elevated temperatures, allowing for the incorporation of nitrogen into the carbon lattice [25]. Nitrogen functionalities introduced may include pyridinic and pyrrolic nitrogen.

Gas Activation: In this method, carbon precursors are exposed to activating gases, such as steam or carbon dioxide, at high temperatures [26]. Physical activation results in tailored pore structures and the introduction of nitrogen functionalities like pyridinic and pyrrolic nitrogen [27].

Template Method: A template, often a sacrificial substance, is utilized to create porous structures in the carbon material. Nitrogen-doping is achieved through the introduction of nitrogen-containing precursors during the templating process [28].

These physical synthesis methods yield nitrogen-doped activated carbons with specific characteristics, offering versatility in applications such as CO₂ capture.

Hydrothermal method: this is a promising technique for obtaining nitrogen-doped activated carbon due to its ability to incorporate nitrogen into the carbon structure and control the pore size distribution [29]. In this method according to **Figure 1** the precursor is mixed with a nitrogen-containing compound, such as urea or ammonia, and placed in furnace tube for several hours and 400-8000C [11], according to the type of precursor. And after passing hydrothermal treatment, applied carbonization and activation steps the same as physical method on threated material [27].



Figure 1. Schematic exhibition of hydrothermal synthesis process for N-doped activated carbon materials derived from a GO/MR composite. Reproduced by permission from Ref [30].

However, the selection of raw materials with excellent carbon content and no

inorganic components, suitable production method and high nitrogen content doping agents are all important to achieve N-doped activated carbon with high quality (high surface area, high adsorption capacity, and appropriate pore size and pore structure [31]). The production of N-doped activated carbons (N-ACs) in all mentioned methods involve introducing nitrogen functionalities into the carbon matrix, to increase CO₂ adsorption capacity of product [32]. There are three types of nitrogen functionalities that were introduced during production (see **Figure 2**).

Pyridinic Nitrogen: Embedded in the carbon lattice, pyridinic nitrogen provides active sites for CO₂ adsorption [33].

Pyrrolic Nitrogen: Present in the form of five-membered rings, pyrrolic nitrogen contributes to enhanced surface reactivity [34].

Quaternary Nitrogen: This nitrogen form improves basicity and can act as a potential site for CO₂ adsorption [34].

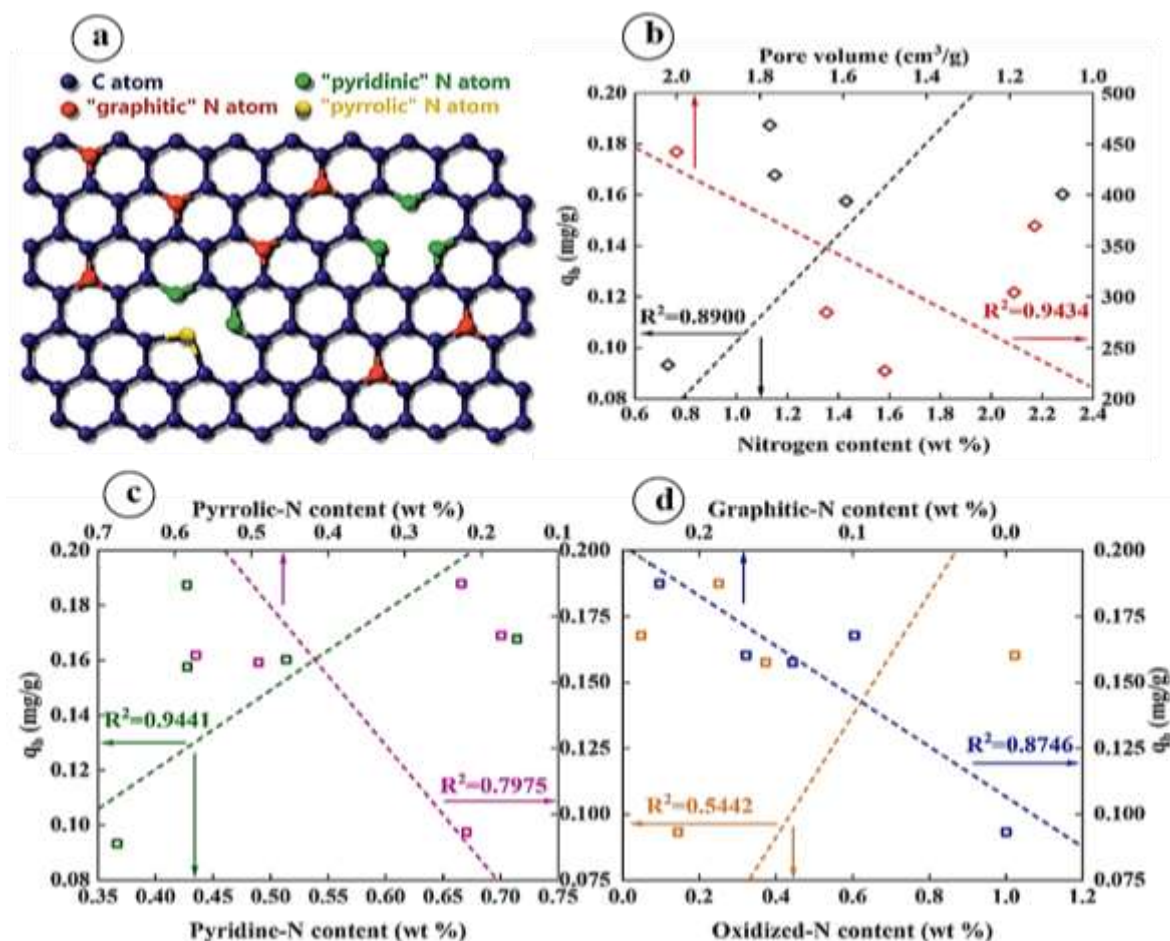


Figure 2. (a) Schematic Representation of Various Nitrogen Atom Types; (b) Nitrogen with graphitic, pyridinic, and pyrrolic structures, contributes to the composition of N-doped AC. Reprinted by permission from Ref [33]; (b) Correlations between adsorption capacity and lattice volume (normalized by BET surface area), as well as with the nitrogen content of the polymer species (q_b values); (c, d) Affinity relationship between nitrogen content and q_b. Reproduced by permission from Ref [35].

3. Conducted methods and synthesized N-doped activated carbon

Numerous methods for producing N-doped materials have been documented in

the literature (see **Table 1**), each utilizing different nitrogen sources, obtaining conditions, and equipment. The synthesis of N-doped carbon materials can occur either in situ or as a separate post-treatment process following the preparation of carbon materials. The addition of nitrogen atoms into carbon network can be achieved during CVD by use of nitrogen compounds together with carbon source or through the use of nitrogen containing carbon based compounds [33], This method is particularly beneficial for carbon nanotubes and graphene production [6]. However, other synthesis techniques such as solvothermal and arc-discharge methods can give the doping characteristics as well [36]. Other materials such as biomass have been used as a precursor. Pyrolysis acts as an in situ doping technique that is done at 800 °C in mostly cases [37]. Consideration of MOFs with nitrogen and carbon atoms has been done as precursors [38]. for N-doped carbon materials. These types of materials are usually formed by carbonization of MOFs at temperatures between 500 °C and 800 °C. But the temperatures over 800 °C may cause the issue of structural damages and degradation [39]. On some other occasions mechanochemistry has been applied to bring in nitrogen species to the carbon matrix without requiring high temperatures [13].

Another approach is comprising of the electrochemical exfoliation of a carbon material to form N-doped graphene as electrode catalyst. This led to an amelioration of the graphene capacity in comparison to the pristine graphene [40]. By contrast, the second type of technique-post-treatment technique-is to add additional nitrogen atoms to the existing sites such as edge of a substrate and defects. For example, post-treatment techniques include annealing (300–500 °C), hydrothermal processes [41], plasma treatment [13], and ion implantation [33].

Of all the doping techniques, chemical vapor deposition (CVD) is noted to be especially useful for nitrogen doping of carbon materials because it is highly efficient and also it is adaptable [34]. However, it requires significant temperatures. Implementation of ions produces more doped materials with few impurities, allows for precise control over doping levels and guarantees consistency of doping, whereas their disadvantages are the high cost and energy demands [42]. Leveraging the phenomenon of self-doping biomass as a potential source of precursors appears to be an attractive solution due to its abundant availability and eco-friendly nature. Nevertheless, using activation agents requires serious consideration, given that some chemicals like KOH can be harmful to the environment [13]. Consequently, reflection should become a mandatory step when choosing a more sustainable option, especially in the case of biomass application [43].

Table 1. The summary of some literature that is used for this review.

N-doped AC from	Production method	N-(%), surface area (m ² /g)	Application	Performance	Ref
Rice husk	Hydrothermal, KOH-activated	- (1495.52)	CO ₂ adsorption	5.83 mmol/g at 273 K, 1 bar	[1]
Rice husk	Pre-deashed, pyrolysis, ammonia activated	N-0.38%,	CO ₂ adsorption	High accordingly	[44]
FAU zeolite	Pyrolysis, Ca ²⁺ ions catalyzed	N-5%, (2400)	CO ₂ adsorption	3.25 mmol/g, 1 bar, (30 °C)	[27]
Ethylenediamine + carbon tetrachloride	Pyrolysis, KOH activated	and N-3.3%, (1463)	supercapacitor	363 F/g at 0.1 A/g in 1 M H ₂ SO ₄	[45]

Table 1. (Continued).

N-doped AC from	Production method	N-(%), surface area (m ² /g)	Application	Performance	Ref
Samanea saman leaves	Direct-activated by NaOH	N-4.6%, (2930)	supercapacitor	in organic electrolyte 179 F/g and 88 F/cm ³	[23]
Soybean	Both (chemical used ZnCl ₂ and followed by physical used CO ₂)	N-0.64%, (811)	CO ₂ adsorption	23 mg/g at 120 C	[46]
Waste leaves of citrus aurantium	Hydrothermal carbonization, ZnCl ₂ -activated	-	CO ₂ adsorption	8.43 mmol/g, 1 bar, (25 °C)	[47]
Cross-linked triazine polymer	Pyrolysis, KOH activated	N (7.89 atomic %), P (0.74 atomic %). (1332)	CO ₂ adsorption, Supercapacitor	1.52 and 5.68 mmol/g at 1 and 5 bar, 172.7 F/g at a current density of 1 A/g.	[48]
Fe ₃ O ₄ +N-doped AC	Hydrothermal	-	Li- ion battery	480 mAh/g) at 20 A/g-1000 cycles.	[49]
N-doped rGO	Pyrolysis	-	Supercapacitor, Na-ion batteries	18.7 F/cm ³ at a current density of 1 A/cm ³ .	[50]
Coconut shells	Hydrothermal, KOH activated	nitrogen and oxygen contents (11.23%, 13.53%)	CO ₂ adsorption	(7.9 mmol/g, 100 kPa and 273 K	[51]
Yangchangwan coal	Hydrothermal, KOH activated	N-3.3%, (1920.7-3078.9)	CO ₂ adsorption	5.96mmol/g at 25 °C and 3.92 mmol/g at 0 °C	[19]
CNs grafted on ACFs	Chemical vapor deposition (CVD)	-	CO ₂ adsorption	1.53–1.92 mmol/g at 1bar, 25 °C	[34]
Hollow Ga ₂ O ₃ @N-CQD nanospheres	Hydrothermal	-	Li-ion battery	at 0.5 A/g, 700.5 mAh/g-500 cycle	[52]
Chitosan gel	Sol-gel, KNO ₃ activated	N-8.6%, O-13.5%, (678.8)	Supercapacitor	329.2 F/g in 6M KOH at 0.5A/g	[53]
PACP hollow nanosphere	Co-polymerization strategy-NH ₃ -assisted carbonization, KOH activated.	N-6.25 atom%, (1237)	Energy Storage	789 mAh g ⁻¹ at 0.5 C after 200 cycles	[54]
Melamine resorcinol formaldehyde resin-graphene Oxide (MR-GO)	Pyrolysis, KOH activated	N-6.92%, (1264)	CO ₂ adsorption	5.21 mmol/g at 25 °C and 500 kPa	[30]

Table 1: Overview of N-doped carbon materials outlined.

4. Application of N-doped activated carbon

The application of nitrogen-doped activated carbon represents a pivotal stride in materials science, presenting a myriad of opportunities across diverse sectors. The advantageous properties imbued by nitrogen doping have propelled this material to the forefront of technological innovation, with implications for energy storage, catalysis, and environmental remediation [13]. One of the most noteworthy applications is in the realm of energy storage [55]. Nitrogen-doped activated carbon has exhibited exceptional performance as an electrode material, especially in the evolution of supercapacitor and batteries [56]. Its enhanced electrochemical properties, stemming from the introduction of nitrogen functionalities, contribute to improved energy density, charge-discharge efficiency, and overall stability [57]. These attributes are critical for addressing the growing need for efficient and sustainable energy storage solutions in the face of escalating global energy needs [58]. Catalysis is another domain where nitrogen-doped activated carbon has demonstrated considerable prowess [59]. The introduction of nitrogen species into the carbon matrix serves as a

catalyst, influencing various chemical reactions [60]. This has far-reaching implications for industrial processes, including the production of fine chemicals and environmental remediation strategies. The catalytic activity, combined with the material's stability, makes it a compelling choice for advancing green and sustainable chemical synthesis. Moreover, the environmental applications of nitrogen-doped activated carbon are noteworthy [12]. Its adsorption capacity, influenced by the nitrogen functionalities, proves effective in capturing and removing pollutants from air and water [11]. This capability aligns with the growing emphasis on developing materials for environmental remediation, addressing issues such as water purification and air quality management [61]. As research on nitrogen-doped activated carbon continues to unfold, its versatility and adaptability become increasingly apparent. The exploration of novel synthesis methods, coupled with a deeper understanding of its structural and chemical characteristics, opens avenues for even more tailored applications [10]. Ultimately, the multifaceted nature of nitrogen-doped activated carbon positions it as a cornerstone material, contributing significantly to advancements in sustainable technologies and environmental conservation [18].

4.1. CO₂ adsorption

As the greenhouse gas CO₂ is being widely recognized as the major contributor to global warming, scientists have been researching the potential of nitrogen-doped materials in environmental and sustainable solutions [62]. The CO₂ adsorption process utilizes solid adsorbents having different adsorption mechanisms [31]. A range of porous solid materials, including carbon materials [7], and metal-organic frameworks (MOFs) [63], and their composites, have been researched and tested as CO₂ adsorbents [2]. Carbonaceous materials are among the most affordable options and have excellent CO₂ adsorption effectiveness (see **Figure 3a**), structural stability, and chemical stability. They can be used in many applications [64]. For instance, the formation of N-doped activated carbon (N-doped AC) by physical activation with carbon dioxide and nitrogen gas and has been reported. The material was carbonized at 800 °C and then activated with carbon dioxide and nitrogen gas at 800 °C for 2 h. The nitrogen doped AC thus produced had elevated nitrogen content and well defined pore size distribution and hence was suitable for catalysis [65]. The study further investigated the effect of activation temperature on the structures and properties of N-doped AC resulting from steam and ammonia physical activation. Beforehand, the precursor was carbonized at 900 °C and it was activated with steam and ammonia at 750 to 950 °C for 2 h. Our research showed that the activation temperature was the major factor governing the pore size distribution and nitrogen content of N-doped AC [66]. Moreover, peanut shell activated carbon was produced through the hydrothermal method by using urea solution as nitrogen source and KOH as a chemical activator. The synthesized activated carbon had a high surface area of 2700 m²/g and a nitrogen content of 4.9%. The adsorption isotherm showed that the activated carbon had a high CO₂ adsorption capacity of 7.6 mmol/g at 298 K and 1 bar [67]. doping nitrogen in activated carbon enhances CO₂ adsorption capacity through several mechanisms: as instance, increasing the surface area [68], provide the context of chemisorption [7], improved pore structure, enhanced wettability, and increase surface basicity [8]. these

combined effects make N-doped activated carbon a promising material for CO₂ capture (**Figure 3b**), and sequestration applications.

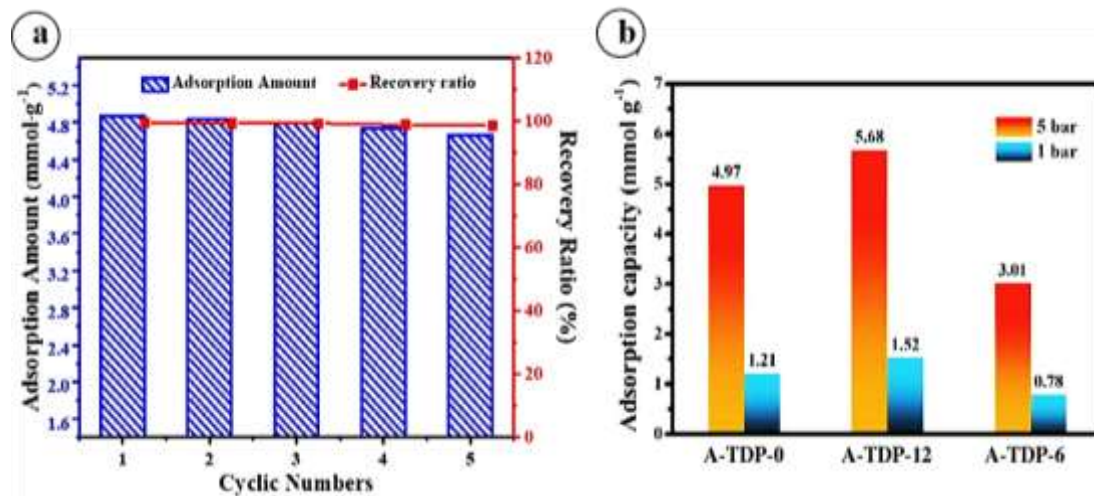


Figure 3. Cycling of CO₂ adsorption by GO (0.25)/MR-500 at 298.15 K and 500 kPa. **(a)** Amount of CO₂ adsorbed by MR/GO material at RT and 5 bar in 5 cycles. Reprinted by permission from Ref [30]. **(b)** The adsorption capabilities of diverse N/P co-doped carbon materials under varying pressures. Reprinted by permission from Ref [48].

4.2. Up taking Mechanisms of CO₂ by N-doped AC

The adsorption of carbon dioxide (CO₂) by nitrogen-doped activated carbon involves several key mechanisms that contribute to the overall process. Activated carbon is a porous material with a high surface area, and the introduction of nitrogen functionalities enhances its ability to adsorb CO₂ [19]. Here's an overview of the mechanisms involved:

4.2.1. Pore filling and surface adsorption

Nitrogen-doped activated carbons play a vital role in CO₂ adsorption by employing a dual mechanism of pore filling and surface adsorption (**Figure 4a**). These materials, crafted through diverse methods like urea modification, KOH activation, and physical activation, demonstrate remarkable efficacy in capturing CO₂ [7]. Nitrogen doping significantly enhances CO₂ adsorption capacity by introducing active sites for interaction with CO₂ molecules [8]. Pores within the activated carbon structure facilitate pore filling, enabling the physical entry of CO₂ molecules [69]. Furthermore, the nitrogen-doped activated carbon surface provides specific sites for chemical adsorption, fostering interactions with CO₂ molecules [1]. Research underscores the substantial contribution of N-doping to CO₂ adsorption, surpassing the impact of exceedingly high surface area values [70]. The synthesis of N-rich porous carbon serves as a tangible demonstration of the favorable traits that enhance CO₂ uptake [31].

4.2.2. Chemisorption with nitrogen functionalities

Nitrogen doping introduces nitrogen-type functional groups on to the activated carbonaceous surface. The most common nitrogen precursors include pyridinic, pyrrolic and graphitic nitrogen. When such nitrogen groups chemically bind with CO₂ with the help of chemisorption, these bonds become stronger and result in inherently resilient chemical bonds. As such, pyridinic nitrogen is the most active nitrogen found

in graphene as it can bond with CO₂ through Lewis acid-base action, (**Figure 4b**).

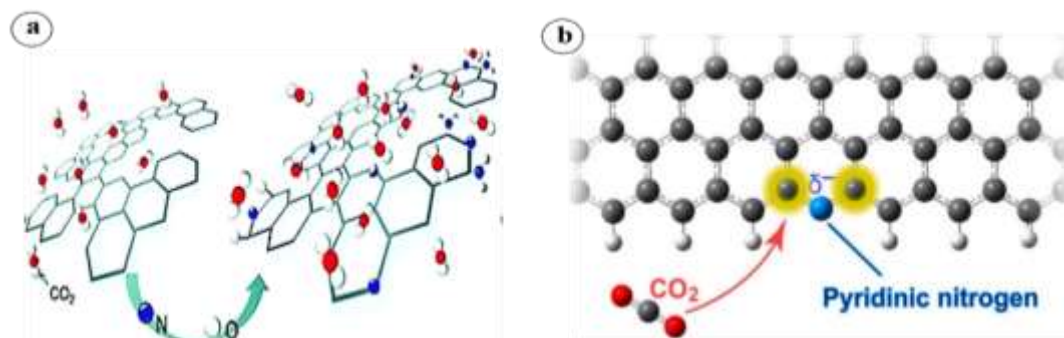


Figure 4. (a) Illustrate the adsorption of CO₂ on the surface of activated carbon. Reprinted by permission from Ref [6]. (b) show CO₂ by chemisorption process of CO₂ on N-doped AC Reprinted by permission from Ref [71].

4.2.3. Lewis acid-base interactions

Nitrogen functionalities, especially those with lone pair electrons, can act as Lewis bases, whereas CO₂ can act as a Lewis acid [72]. The Lewis acid and base reactions involve giving and taking electrons to make coordinate covalent bonds. This interaction improves the material's ability to capture CO₂ [73].

4.2.4. Electrostatic interactions

The presence of nitrogen-containing groups can introduce additional charge distribution on the surface of activated carbon. CO₂, being a polar molecule, can interact with the charged sites through electrostatic interactions, further contributing to its adsorption [74].

4.2.5. Enhanced surface heterogeneity

Nitrogen doping increases the surface heterogeneity of activated carbon, making different active sites for capturing CO₂, (see **Figure 5**). The diverse range of active sites allows for multiple interactions, including physical adsorption, chemisorption, and electrostatic interactions [75].

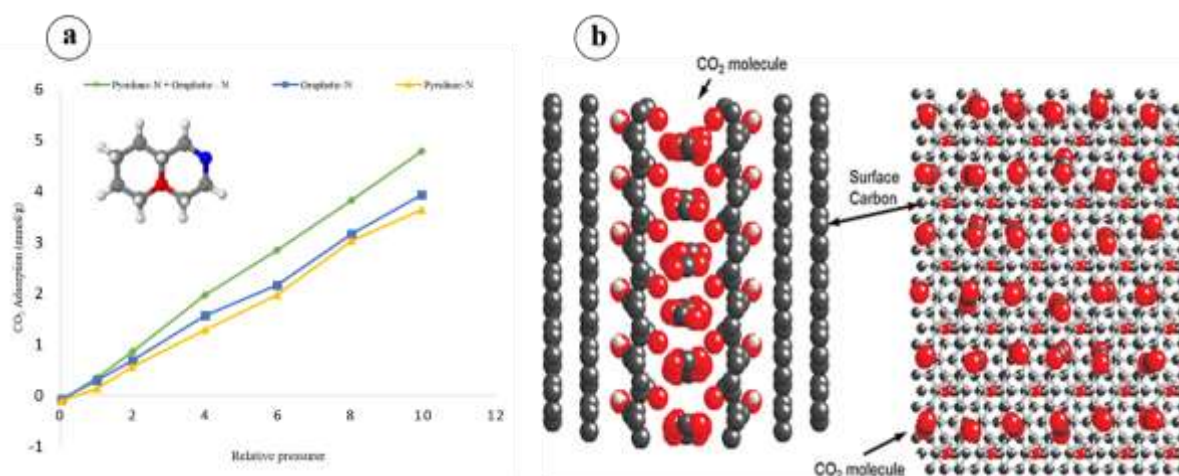


Figure 5. (a) Presents a simulation of CO₂ uptake isotherms at 298 K, exploring a pressure range up to 10 bar. The simulation involves a generated structure, highlighting the presence of Pyridinic-N (shown in yellow), Graphitic-N (depicted in Blue), and a mixture of both (in Brown). Reproduced by permission from Ref [76]; (b) Influence of Surface Variability on CO₂ Adsorption in Microporous Carbon Materials. Reprinted by permission from Ref [75].

4.2.6. Selective adsorption

Nitrogen-doped activated carbon can exhibit selective adsorption, preferentially adsorbing CO₂ over other gases due to specific interactions and affinity for CO₂ molecules [4]. The combination of physical adsorption [6], chemisorption [71], Lewis's acid-base interactions [72,73], electrostatic interactions [74], and surface heterogeneity [75], results in an effective adsorption process for CO₂ by nitrogen-doped activated carbon. This makes such materials promising candidates for applications in carbon capture and storage, as well as other environmental and energy-related processes.

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Interventionary studies involving animals or humans, and other studies that require ethical approval, must list the authority that provided approval and the corresponding ethical approval code.

4.3. Energy storage systems

4.3.1. Supercapacitor

The strategy of nitrogen doping into the structure of carbon-based materials has become the biggest game changer in enhancing the specific capacitance of electrode materials, which in turn has ignited interest in using N-doped AC, in supercapacitors these days [77]. Various aspects including porosity, surface area, and the exact amount and type of nitrogen are the factors which determine the electrochemical performance of N-doped AC electrodes, in terms of the specific capacitance, rate capability, and cycle stability [78]. The most recent research indicates that the nature of N atom arrangement on the surface of N-doped ACs (including pyrrolic N, pyridine N and quaternary N/graphite N, and N oxides of pyridine N) depends on the temperature of heat treatment and on the nitrogen source. Although the most commonly utilized types of nitrogen end up being hydrogenated nitrogen and pyridinic nitrogen, a great number of nitrogen types are usually present in N-doped ACs which are prepared through high-temperature heat treatment (>600 °C) or hydrothermal approach [79]. Although much work has been done to improve the application of N-doped ACs for electrochemical capacitors, it is difficult to find a straight line representing the positive changes between the capacitance of specific Cs and surface area or nitrogen content in N-Acs [80].

The most problematic issues are small size pores inaccessible for ionized solvent molecules, potential nitrogen atom polarity, and pseudo capacitance effects caused by oxygen groups of carbon-based products. In addition to that, the nature of the electrolyte, consisting of concentration, properties, and wettability to the electrodes as well, plays a crucial role in specific capacitance of N-ACs. This complexity stems from the connection of nitrogen to functional groups that are synthesized through conventional methods, which created the hindrance of distinguishing individual role of each group [81]. Besides that, the same carbon materials doped and non-doped with

nitrogen groups can hardly be prepared with the same characteristics, which in turn makes it even harder to clearly understand the practical effect of nitrogen-containing functional groups on capacitance [82]. The role of N-atoms on graphene edges which had initially suggested that this is the factor capable of causing pseudo capacitance has not yet been fully understood or confirmed, in relation to its mechanism. The strongest link between capacitance and surface groups, and the porosity of the carbon enhanced with nitrogen anodes in acidic electrolytes is also reported [83]. In addition to this, bulk gravimetric capacitance correlation was noted with the amount of basal planes whereas normalized capacitance in micropores was determined to directly depend on the dispersion of quaternary nitrogen and pyridinic-N-oxide nitrogen species mainly under increased current densities. Moreover, pyridinic and pyrrolic nitrogen were accompanied by quinone oxygen groups in the core structure of MOFs and were revealed to be the vital capacitance contributors [81]. Building on the results of such research, the further teamwork was focused on the production of “ammonia-activated non-porous carbon materials with low surface area by melamine mica composites”. Along with this, those materials proved their cycle stability and capacitance that was three times higher compared to the performance of untreated composites standing for the significance of nitrogen content and surface chemistry. Nitrogen insertion by carbon-based materials, especially N-ACs, and their mutual relationship with supercapacitors is also under investigation on application performances. Materials that underwent pre-ammonia treatment produced higher nitrogen content as well as increased concentrations of pyrrole-type nitrogen-containing functional groups due to which the pseudo capacitance was improved [84]. Several investigations have disclosed that pyrrole and, sometimes, pyridine natures give rise to pseudo-capacitive effects. XPS provide evidences for this effect; binding energies could be found within the range of 398.0–400.5 eV. There are images of negative clusters of atoms, for instance pyrrolic-N and pyridinic-N, outside the structure of carbon that are linked to pseudo capacitance whereas positive clusters, such as quaternary-N and pyridine-N-oxide are proposed to contribute to electron transfer in carbon (**Figure 6**) [85].

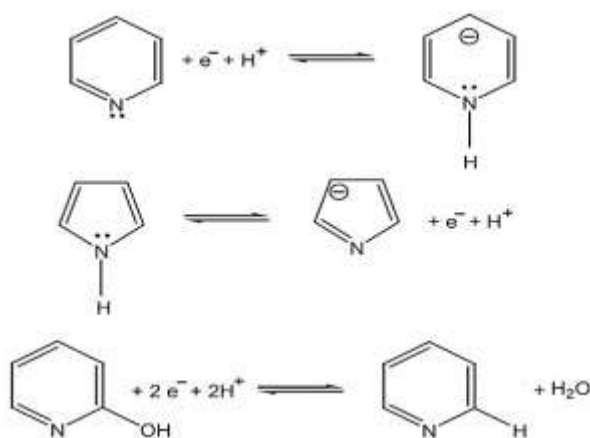


Figure 6. Illustrates potential redox reactions associated with nitrogen species of pyridinic, pyrrolic, and pyridonic types.

Nevertheless, the pseudo capacitance of quaternary-N is an area of disagreement among researchers. Some studies propose that quaternary-N can generate pseudo-

capacitance by interacting with protons in H₂SO₄ electrolyte or by the electrical double-layer formation boosting. Alternatively, some scientists suggest ions' interactions with anions in KOH electrolyte [86]. For example, in a study PPy nanospheres (PNs), carbon nanospheres (CNs), and microporous carbon nanospheres (MCNs) derived from chemical activation of CNs. MCNs, with a developed microporous structure and lower quaternary-N and pyridinic-N functionalities, exhibited significantly improved specific capacitance compared to PNs and CNs. Nitrogen-doped porous CNFs containing N-Q and N-6 displayed reversible specific capacitance in aqueous KOH electrolyte, highlighting the influence of nitrogen functionalities [87]. One of the experiments demonstrated the synthesis of N-doped CNTs (N-CNTs) via chemical vapor deposition (CVD) and the roles of pyrrolic, pyridinic, and quaternary nitrogen groups in acidic and basic electrolytes, respectively. As per their suggestion, N-Q functional groups were found to enhance capacitance through interaction with anions [88]. Intricate mechanisms of redox reactions involving N-containing functional groups were elucidated using *ex situ* XPS coupled cyclic voltammetry (CV) analysis, suggesting electrochemical redox reactions between pyridinic and pyridonic-N, as well as N-O and pyridinic-N [89]. A large variety of papers have been published on the review of capacitance and the porosity of NCs, but the links of the number of basic groups and capacitance is being in the shadows. The principle reason for the lower porosity and nitrogen content of the NC-based materials adopted is the considerable trouble of obtaining the ideal balance between both the high content of pores and nitrogen atoms [77]. Furthermore, other researchers reported a broad KOH activation method that allows for potassium-riched nitrogen-doped porous carbons (NPC's), so that the synthesis methods are the key for getting the functional capacitors to achieve a faster performance in charging and discharging. Because KOH weight ratio to carbon strongly affects the porosity and nitrogen content of the resulting NPCs during their manufacturing, these parameters were found to be more variable between the produced NPCs. With the KOH/carbon weight ratio being increased from 0.5 to 2, specific surface area of the NPCs also rises from 522 to 1913 m²/g whilst the content of N decreases from 10.8% to 1.1%. Another issues present in the aging process would be the falling N/P ratio which leads to a decline of the pyridinic-N content and an increase of the pyrrolic/pyridonic-N, quaternary-N, and pyridine-N-oxide content [90].

Additionally, nitrogen content per unit weight reduced at high KOH/NPC ratio but pyridinic-N and pyrrolic/pyridonic-N namely stayed dominant anyhow in all synthesized NPCs. The electrochemical tests showed that NPC-1, was made with a low amount of KOH: carbon ratio of 1.0 and had the higher specific capacitance at 363 F/g that was tested at the current density of 0.1 A/g in aqueous electrolyte with 1M H₂SO₄. The authors expected this to do something highlighting their success to contribute it to factors such as the greater specific surface area of NPC-1 compared to NPC-0.5 and the higher nitrogen content of NPC-1 compared to NPC-2. The jointly arising tight double-layer capacitance and extra pseudo capacitance are responsible for the system of NPC-1 having the highest specific capacitance [91].

Again, the method of KOH activation was used to prepare NCNPs with hierarchical porous N-doped carbons as the second instance. The capacitance that measured the specific area, rise with the growth in the specific surface areas, and

capacitance normalized by specific surface area ($CA = \text{cm}/S_{\text{BET}}$), increase with nitrogen content mol % (represents the concentration of a component in a mixture, calculated as moles of component per 100 moles) [92]. Similar patterns were observed in the hierarchically porous, nitrogen-doped carbon nanotubes (NCNTs) generated from the KOH activation of those NCNTs retrieved from the polyaniline nanotubes. Under the identical nitrogen composition, the calculations of specific capacitance were near similar, reflecting the changes in surface area only to the certain degree [93]. The above findings thus establish an intricate connection between the role of porosity, nitrogen content and electrochemical performance in nitrogen-doped carbon material, highlighting the importance of having an intimate control over synthesis parameters for achieving the highest specific capacitance in supercapacitor applications [45]. The effects of carbonization temperature on N content, pore structure and surface area are known, and also the influence on specific capacitance (C_{sp}) is well understood [94]. This could be illustrated through nitrogen doped graphitic porous carbon nanofibers (CNFs) acquired from carbonaceous nanofibers coated with polypyrrole (CNFs@PPy) at different temperatures (ranging from 500 to 1100 °C) which exhibited the highest pseudocapacitance at 900 °C [95]. Such high pseudo capacitance was the result of a synergized effect: both nitrogen doping that rendered new carbon species as having better electron donor/acceptor characteristics and a high BET surface area that enhanced the electrolyte ion transportation [96]. The same trends observed in nitrogen-enriched carbon nanowires produced directly from the polypropylene aniline nanofibers using elevated temperatures (started at 600 °C and reaching 900 °C) upon calcination resulted in a decrease in the nitrogen content at increased calcination temperatures. The samples had the highest specific capacity at 700 °C because of the fact that the surfaces area was balanced in its proportion with the nitrogen grown [90]. It has also been reported that smaller pores diameter in the porous carbon spheres administrate higher capacitance and faster rate compare to the larger spheres which having similar surface area. This is due to the less resistance in masses and to charge transfer, the rise in the EDLC performance, and a better way by which the pores are inaccessible to the aqueous electrolyte [97].

Nitrogen-dope carbon structure (NC) is also associated with specific capacitance and rate performance action. The same impurity (nitrogen) of carbon nanoparticles (NCNPs), carbon nanotubes (NCNTs), and carbon nanowires (NCNWs) derived from aniline under various treatment conditions resulted in different electrochemical performances. Varying parameters like smaller particles size, appropriate pore size distribution are keeping a nitrogen functionalities were found to be contributors of better specific capacitances [98]. The selection of electrolyte is also an essential factor for the specific capacitance of dielectrics [77]. For example, in one study, NCs doped by KOH showed improved power output when the acidic based electrolytes were used instead of the basic ones [99]. and the main reason is referred to the fact that carbon surface is markedly more basic in the presence of nitrogen. Next, carbon gels created from the gelatin biomolecules showed that there was higher specific capacitance in the acidic H_2SO_4 solution compared to the basic KOH solution (**Figure 7**). This proves that the type of electrolyte can be changed by the use of pseudo capacitance [77].

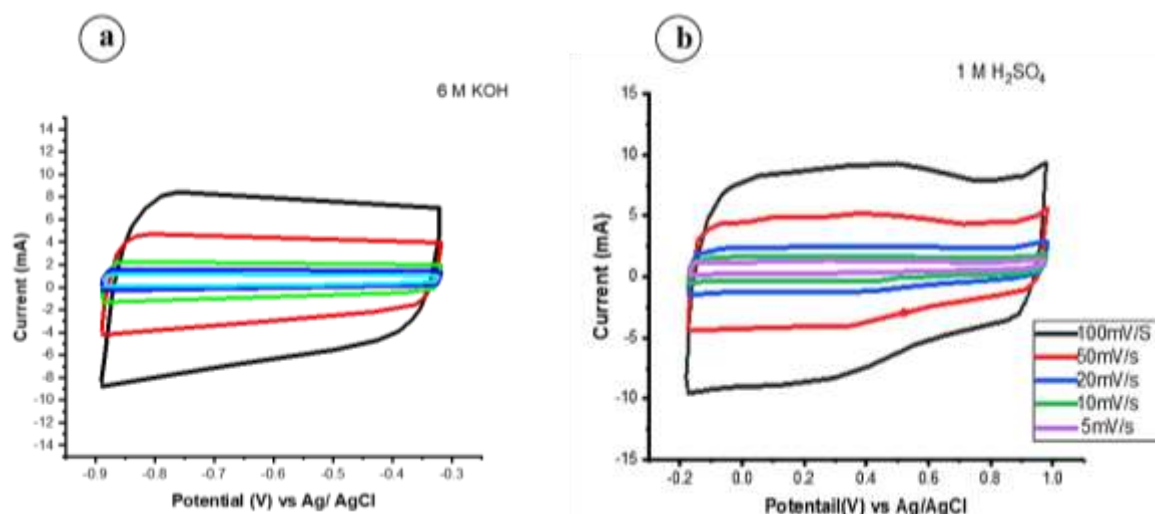


Figure 7. Illustrates the electrochemical capacitive behavior of Nano catalysts (NCs) in 6 M KOH (left column) and 1 M H₂SO₄ (right column) aqueous solutions using a three-electrode configuration. Panels (a) and (b) present cyclic voltammograms recorded at different sweep rates. Reproduced by permission from Ref [100].

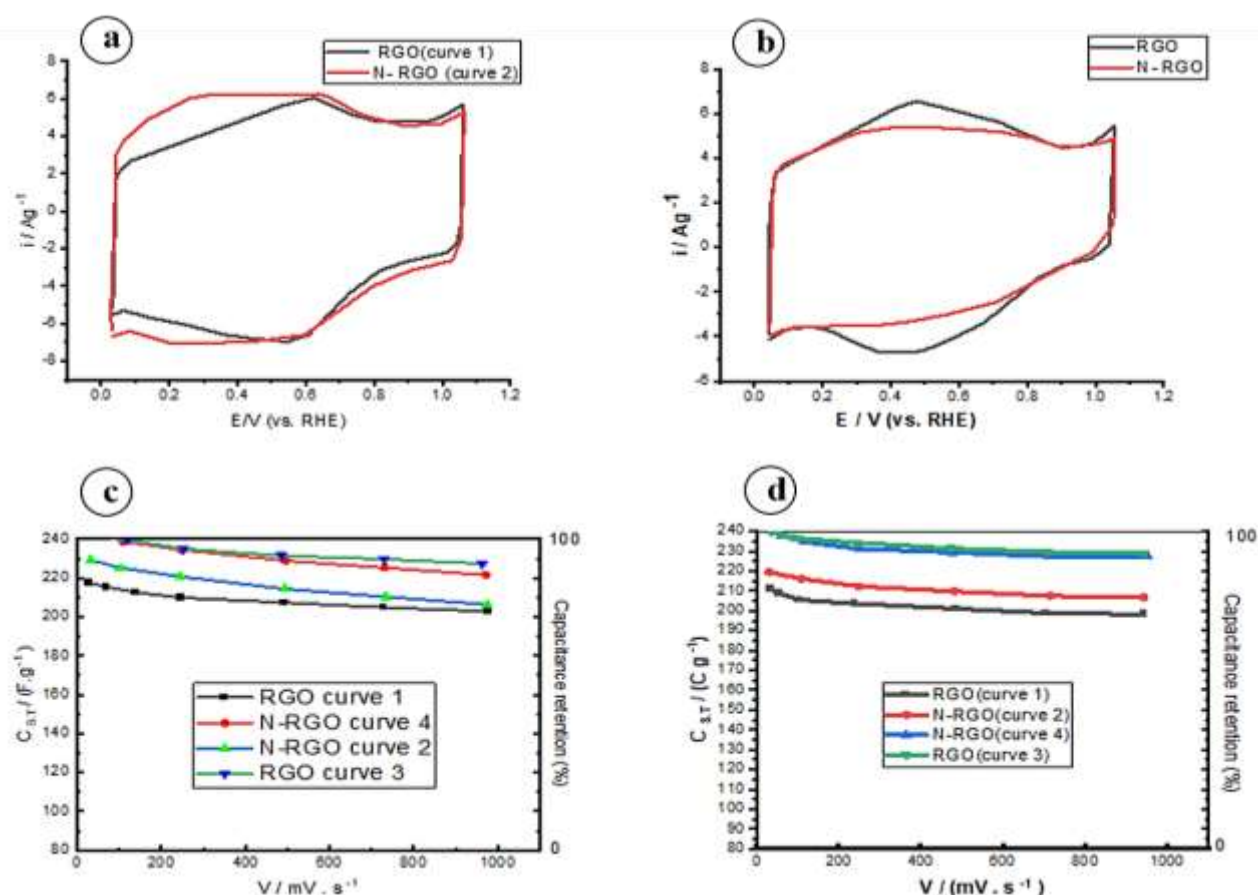


Figure 8. Displays cyclic voltammograms recorded in (a) 1 M H₂SO₄; (b) KOH at a sweep rate of 25 mV/s; and the total specific capacitance ($C_{s,T}$) and capacitance retention (calculated based on the value obtained at 25 mVs⁻¹) against the scan rate of the cyclic voltammetry (CV) for Reduced Graphene Oxide (rGO) and Nitrogen-Doped Reduced Graphene Oxide (N-rGO) in. (c) H₂SO₄; (d) KOH. Reproduced by permission from Ref [101].

These works assemble present together the intricate interplay of synthesis factors, carbonization process, composition, and electrolyte selection in dictating the

electrochemical behavior of nitrogen functionalized carbon materials for super capacitors. The application of a technique termed microwave-assisted hydrothermal synthesis include N-doped rGO and rGO [88]. This study intends to explore nitrogen-based functional groups (N-N and N-O) contribution to the specific capacitance, which depends on the nature of electrolytes (aqueous and non-aqueous). It was discovered that pseudo capacitance was increased mainly by pyridinic-N and pyrrolic-N and pyrrolic-N/pyridonic-N in the acidic medium, as confirmed by the electrochemical studies using alkaline (1 M KOH) and acidic (1 M H₂SO₄) electrolytes (**Figure 8**). Due to double-layer capacity effect of N-rGO in H₂SO₄ that is higher than that in KOH, there are some N-containing groups that enhance the densities of electronic charges of graphene and aid in the adsorption of protons in the acidic electrolyte [88].

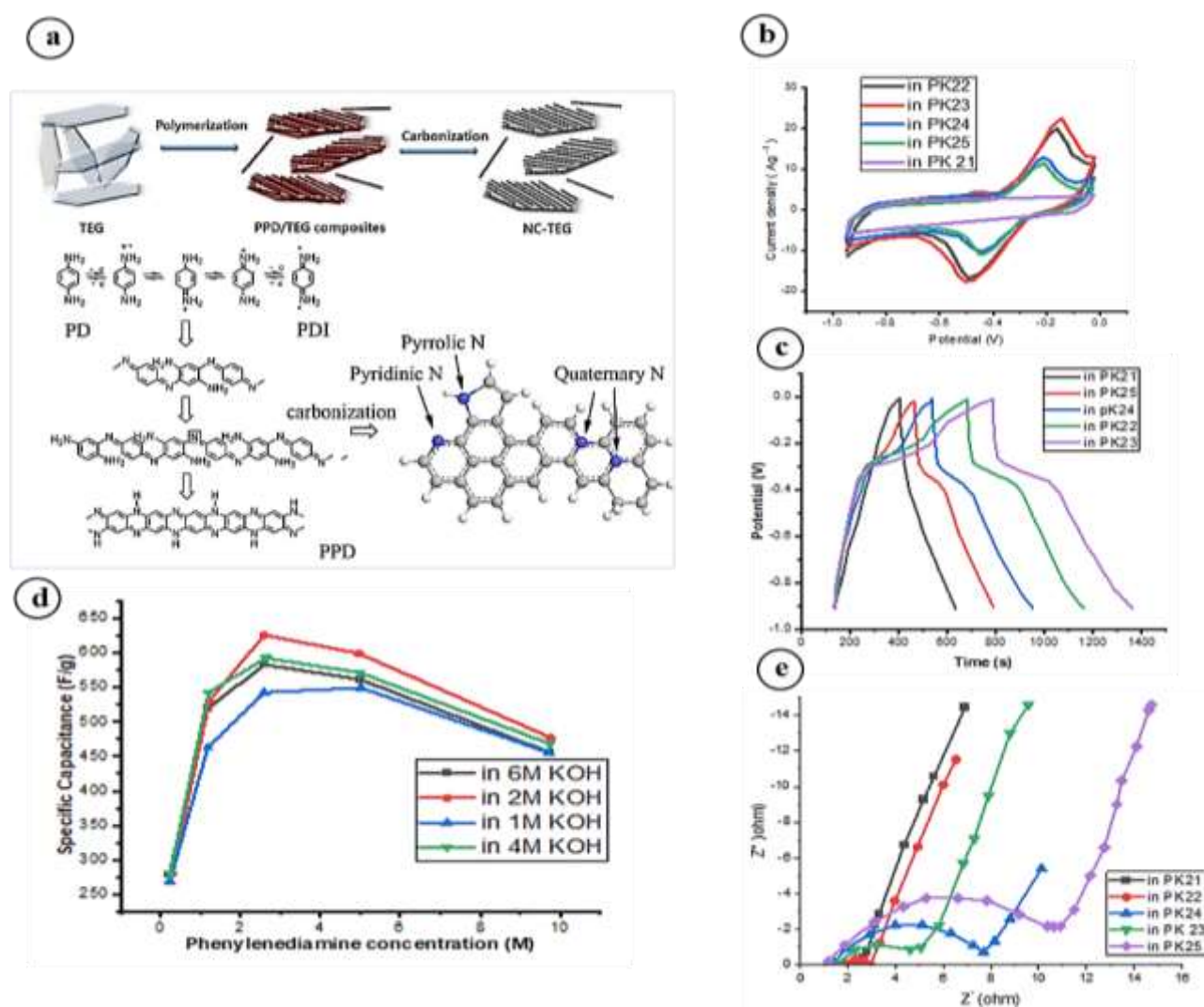


Figure 9. (a) Illustration of the preparation of NC-TEG. Reprinted by permission from Ref [103]. (b) cyclic voltammetry curves; (c) galvanic charge-discharge of the NC-TEG electrode under various concentrations of PD mediator in a 2 M KOH solution. Additionally, (d) compares the capacitance in different electrolytes, and (e) presents the electrochemical impedance spectroscopy (EIS) spectrum of NC-TEG with varying PD concentrations [103].

Contradictory results regarding specific capacitance in acidic versus basic electrolytes have been reported in the literature. Some studies demonstrated higher specific capacitance in acidic electrolytes, while others observed superior performance

in basic electrolytes. For instance, in a research study, 3-D SACFO carbon films of chicken eggshell membranes were developed, which showed higher specific capacitance in KOH than H₂SO₄. Furthermore, it was also determined that the capacity of the N-doped CNTs immersed in the KOH had greater specific capacitance than the ones which were dipped in the H₂SO₄ solution, which shows the importance of selecting an appropriate electrolyte [102]. The effect of nitrogen placeholders on specific capacity in distinct pH solutions is complex in nature and the consequent findings that are released in the literature vary. Continuing work is required to effectively complete the mechanism and further down the line innovative strategies may have to be applied in order to develop the complexity concept in the nitrogen-doping carbons. Yet, besides that, they study how different components of the electrolyte influence the capacitance. The authors demonstrated enhanced faradaic behavior through the epitaxial expression of a nitrogen doped carbon based layer embedded on thermally exfoliated graphene (NC-TEG) and Quinone insertion in the electrolyte into the electronic conductor (PD) (**Figure 9a**). The redox-activity peaks were dominantly observed in a cyclic voltammetry after the incorporation of PD (**Figure 9b**), suggesting a pseudo-capacitive behavior. NC-SC epitomized tech using capacitor and exhibit gradual rise of the specific capacitance with addition of Pd, and reached its maximum specific capacitance in the mixed electrolyte (2 M KOH with 0.025 M Pd) and then showed a downward trend in the electrolyte with higher Pd concentration (**Figure 9d–e**). This study emphasized the importance of electrical composition alongside that of nitrogen as the doping agent in determining the performance of the electrolyte in the capacitive behavior of the carbon Nano-materials [103].

Lastly, the loading mass onto electrodes was discussed as a factor affecting specific capacity and rate performance. In general, an augmentation in loading mass typically leads to a reduction in specific capacity and rate performance. However, limited studies have been dedicated to exploring this aspect specifically in the context of nitrogen-doped carbon electrodes for batteries and capacitors.

4.3.2. Batteries

In an experimental work, Fe₃O₄ was synthesized into nitrogen-doped carbon and acted as a nano-capsule structure with self-filled channels. The Fe₃O₄@NC electrode was compared cycling and rate capabilities with the unmodified Fe₂O₃ electrode, and it exhibited superiority in both aspects. It was shown to function better and it had better stability particularly at high current densities as shown in **Figure 10a,b**. In addition, the ability of the Fe₃O₄@NC anode to be utilized in lithium-ion batteries (LIBs) over a long time was proved to be 480 mAh/g at a high charging density of 20 A/g over 1000 This average was designed to be consistently at 99.9% Coulombic efficiency. With much lower current density of 5 A/g, the reversible capacity attained 873 mAh/g, as **Figure 10c** shows [104].

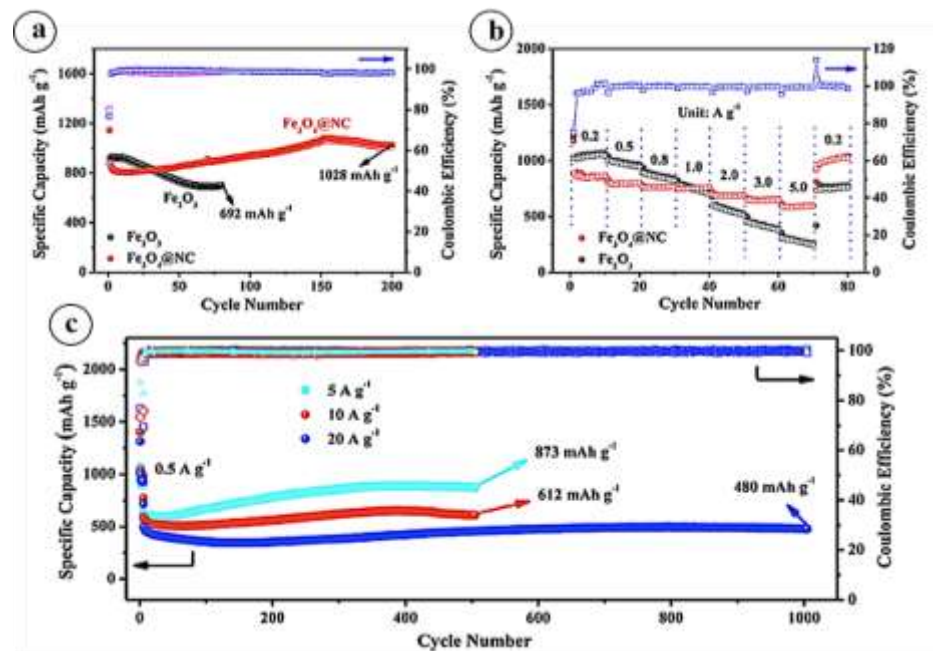


Figure 10. Illustrates the electrochemical performance of Fe₃O₄@NC and Fe₂O₃ as anodes in lithium-ion batteries (LIBs). (a) the cycling performances at a current density of 500 mA/g are presented; (b) showcases the rate performances of both materials in a half-cell configuration against Li/Li⁺. Additionally, (c) provides insights into the cycling performance of the Fe₃O₄@NC anode under different current densities (5, 10, and 20 A/g) throughout 1000 cycles. This comprehensive analysis offers a detailed perspective on how these materials perform in terms of cycling stability and rate capabilities in the context of LIBs. Reprinted by permission from Ref [49].

Moreover, Fe₃O₄@NC anode proved to be practical for high-rate applications, finished to the full charge within 2 min. This implicates the fact that Fe₃O₄@NC nanocarriers is in fact relevant to cases when the current charging is quick [104]. Another research worked on the formation of hollow iron oxide/carbon core-shell nanostructures for lithium ion battery (LIB). To achieve this synthesis, we dealt with Fe₂O₃@polydopamine nanocomposites from the Fe₂O₃ nanotubes and dopamine hydrochloride precursors. Lastly, compositing of Fe₃O₄@N-doped C was accomplished by controlling the carbonization process. In the last step of the Fabrication a series of experiments with Fe₃O₄@void@N-doped C-x composites differing in the void sizes was carried out by etching off Fe₃O₄. The work was performed by investigating the impact of changing etching time on void size. In the electrochemical assessment of LIBs, the sample was the best at 5 Volt for 5 h of etchant when a 2 moles/L HCl acid mixture was used and heated at 30 degrees centigrade. The Fe₃O₄@void@N-Doped C-5 is capable of holding up to 1222 mAh/g after it was discharged at a rate of 200 mAh/g up to 100 cycles [105]. As a separate venture, gallium (Ga) was probed to be a candidate of self-healing anode component. Scientists brought into use a Ga-Sn liquid metal alloy with auto-healing features and wonderful energy and chemical performance applicable with alkali ion battery production. Lips-Ga₂O₃@nitrogen-doped carbon quantum dot (H-Ga₂O₃@N-CQD) nanospheres as a material for lithium ion batteries (LIB's) anode were developed. An

electrode eventually acquired a bulky size with the capacity of 1348.5 mAh/g at 0.1 A/g and after 500 cycles it was able to hold up to the reversible capacity of 700.5 mAh/g under 0.5 A/g [106]. In a similar way, nitrogen-doped porous carbon (NCFC) is obtained from silk cocoon and was used as the cathode (in combination with an ionic liquid) for Li-O₂ batteries. The irregular surface of the NCFC characterized by bending slices demonstrated advanced specific discharge product with more efficient sites available for electrochemical reactions. The Li-O₂ battery functionalized with NCFC is an excellent choice with a high energy efficiency and stability at various current densities surpassing carbon black super P Li-O₂ batteries in terms of the cycle life and over potential. As seen from these results, this compound seems to perform to Li-O₂ batteries and it is certain to be an excellent material for this kind of batteries [107]. This study highlighted that the performance of the electrochemical application improves by adding N through the doping process. Study can be considered as a successful one since increased porosity, surface area and the N moieties preservation were determined without NH₃ removal. The modified PN-HCN with superior is an effect I-S batteries cathode host. It has reached the minimum theoretical capacity of 1620 mAh/g with the highest rate capability and longest cycling stability, which is 789 mAh/g at 0.5C after cycling 200 times Our results show that PN-HCN has not only enhanced performance but also produced surprising results seen in the three comparisons (**Figure 11a–c**). CV-HCN at different current densities (0.05, 0.1, 0.5, 0.8, and 1.0 A/g) is displayed in **Figure 11d**, showing a symmetric signature, which as a good reversibility sign [108].

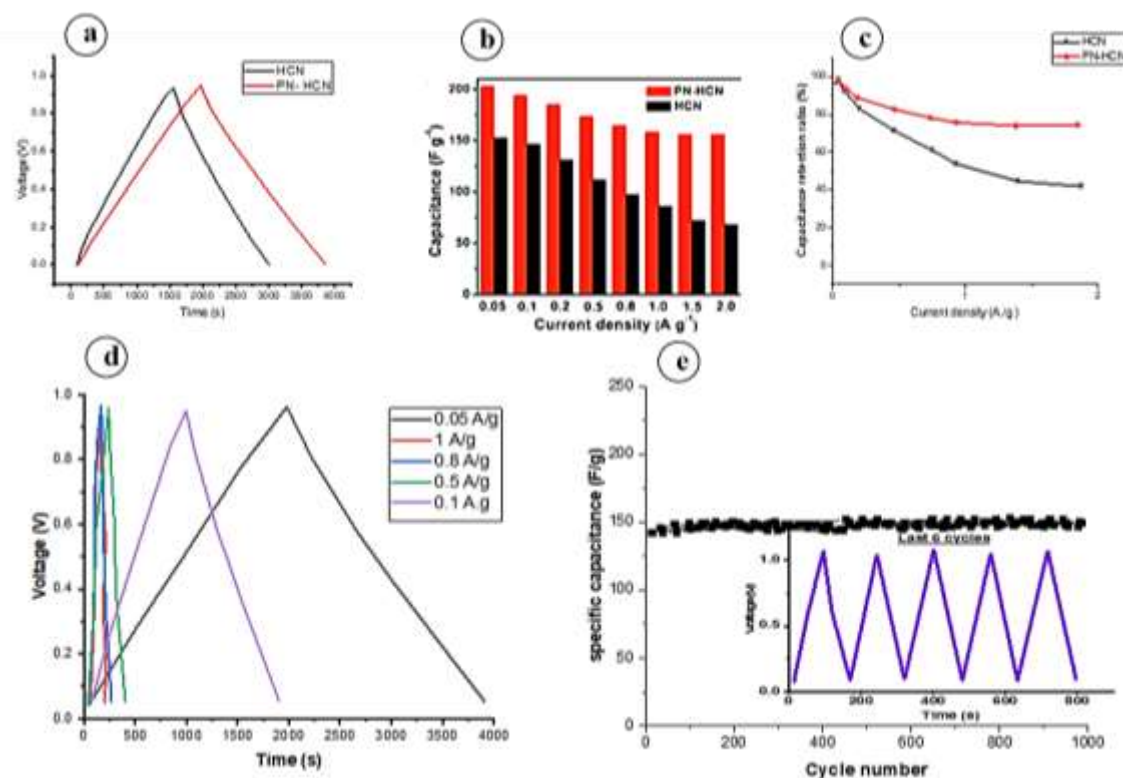


Figure 11. (a) GCD profile at 0.05 A/g; (b) Graphing the specific capacitances and current density; (c) Chart displaying the capacitance retention and current density for PN-HCN and HCN; (d) GCD profiles of PN-HCN at different current densities from 0.05 to 1.0 A/g; and (e) Cycling response of PN. Reproduced by permission from Ref [108].

In the course of cycling above 10,000 cycles PN-HCN has shown excellent cycling stability characterized by the high specific capacitance of 170 F/g, which remains stable and almost the same through such a long cycling (**Figure 11e**). The deliberate structural integration of enhanced porosity, well-maintained N functionality, and a precisely defined nanoscale hollow spherical structure underscores the importance and effectiveness of PN-HCN in enhancing electrochemical performance in Li-S batteries and supercapacitors compared to HCN without NH₃ treatment [108]. Another remarkable experiment is in the list of other scientific analysis, where scientists have pointed out some ameliorations in the area of conductivity inhibition. N-doping is a tool that helps to increase the electrical conductivity of the carbon materials. It can actually be compared with electron facilitation in charge and discharge cycles. This is an indispensable element that leads to increasing the overall battery's workflow. We should also add that nitrogen adsorbed carbon materials participate in the strength and stability increase of the battery, which provides a longer and more feasible work during repetitive charging and discharging [109]. The nitrogen doping causes a large surface expansion and thereby increased active points for action of electrochemical reactions. Thus, within the structure of a battery, these ions can circulate in a confined space allowing for a greater capacity and enhanced storage performance [109]. Besides this, the presence of elements such as nitrogen also allows for the ions to perform effective diffusion within the electrode making the batteries operate faster and the performances better [13]. In different battery types including lithium-ion batteries, sodium-ion batteries, and metal-ion batteries, the Nitrogen-doped carbon materials exhibited their versatility. These features are the evidences of their flexibility which is applied in numerous energy storage systems [110].

5. Conclusion

Nitrogen-doped activated carbon represents a multifaceted solution with profound implications across various domains, underscoring its pivotal role in advancing sustainable practices and addressing pressing environmental challenges.

1) The importance of production of nitrogen-doped activated carbon from plant waste and sustainability in green production cannot be overstated. By utilizing biomass-derived sources such as licorice residue, duck rice husk, walnut shell, feathers, and tea waste, the production process becomes not only cost-effective but also environmentally friendly. This approach not only mitigates waste but also aligns with the principles of circular economy, contributing to a greener and more sustainable future.

2) The significance of nitrogen-doped activated carbon in the adsorption of CO₂ is paramount. Through the interaction between nitrogen and carbon, complex and highly porous structures are formed, greatly enhancing their efficacy in capturing carbon dioxide from various sources. This capability holds immense promise in combating climate change by reducing greenhouse gas emissions and mitigating the adverse effects of global warming.

3) The importance and utilization of nitrogen-doped activated carbon for energy storage in different types, including supercapacitors and batteries, highlight its versatility and potential in the realm of energy storage technologies. From hollow

carbon nanospheres to carbon materials derived from Metal-Organic Frameworks (MOFs), nitrogen-doped activated carbon exhibits superior performance and long-term stability, making it an attractive candidate for next-generation energy storage devices. Furthermore, its catalytic properties and ability to enhance electrode materials in LIBs and ZIBs offer exciting avenues for further exploration and innovation in the pursuit of sustainable energy solutions.

Nitrogen-doped activated carbon stands as a beacon of hope in the quest for a more sustainable and environmentally conscious future. Its production from plant waste, efficacy in CO₂ adsorption, and utilization in energy storage applications underscore its transformative potential in driving the transition towards greener practices and mitigating the impacts of climate change.

Conflict of interest: The authors declare no conflict of interest.

Nomenclature

N-ACs	Nitrogen doped Activated Carbons
MOFs	Metal Organic Frameworks
CVD	Chemical Vapor Deposition
BET	Brunauer-Emmett-Teller
Cs	Specific Capacity
MR-GO	Melamine-resorcinol-formaldehyde Resin/Graphene oxide
PACP	Poly Aniline-Co-Pyrrole
mAh/g	Specific capacity of a battery or an electrode material in energy storage devices (milliAmpere-hour per gram of active material)
A/g	Current density in electrochemical systems or devices (ampere/ gram of active material)
XPS	X-ray Photoelectron Spectroscopy
CNs	Carbon Nanospheres
MCNs	Microporous Carbon Nanospheres
PNs	Ppy nanospheres
CNFs	Carbon Nano Fibers
N-CNTs	Nitrogen doped Carbon Nano Tubes
NPCs	Nitrogen doped porous Carbons
NCNPs	Nitrogen doped Carbon Nano Particles
NC-TEG	Nitrogen doped Carbon based layer embedded Thermally Exfoliated Graphene
LIBs	Lithium Ion Batteries
PN-HCNs	Porous Nitrogen doped Hollow Carbon Nanospheres

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