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OVERVIEW OF CARBON MATERIALS FOR USE IN LITHIUM-ION BATTERIES AND SUPERCAPACITORS

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Abstract. This review article focuses on the study of carbon materials utilized as electrodes in lithium-ion batteries and supercapacitors. The research examines three primary categories of materials: activated carbon, carbon aerogels, and nanoporous carbon. The article provides a comprehensive explanation of the operational principles of many types of capacitor systems, such as double-layer electrochemical capacitors, pseudo capacitors, and hybrid capacitors. The carbon materials under discussion are thoroughly examined with a focus on their synthesis processes, structural features, and electrochemical properties. The study investigates the impact of pore structure, surface area, and the presence of functional groups on electrode performance. The impact of heat treatment and chemical modification techniques on the properties of synthesized carbon structures has been investigated, specifically examining parameters such as temperature and duration. The text examines the benefits and drawbacks of each material type, taking into account their individual capacity, cyclical stability, and economic efficiency. The study emphasizes the importance of precise adjustment of the synthesis process to enhance the electrochemical characteristics and showcases the possibility of employing these materials in sophisticated energy storage devices. This review serves as a crucial resource for experts engaged in developing cutting-edge materials for lithium-ion batteries and supercapacitors. It also highlights potential topics for future research in the realm of electrochemical energy storage.

Keywords: lithium-ion battery, supercapacitor, carbon materials, activated carbon, carbon aerogel, nanoporous carbon.

1. Introduction

In the present era, energy technologies are increasingly becoming essential components of both everyday living and several industrial sectors. Lithium-ion batteries and supercapacitors are essential for the storage and utilization of electrical energy, serving as sophisticated electrochemical devices. The efficacy of energy storage and transmission significantly impacts the advancement of contemporary mobile gadgets, electric transportation, and renewable energy sources.

The choice of materials for electrodes plays a vital role in determining the effectiveness of these technologies. This paper offers a comprehensive examination of three primary categories of carbon materials

- activated carbon, carbon aerogels, and nanoporous carbon - focusing on their electrochemical characteristics and potential applications in lithium-ion batteries and supercapacitors.

One category of materials is activated carbon. Activated carbon possesses exceptional electrical conductivity, resistance to corrosion, and affordability as a result of its substantial specific surface area and porous configuration. This article analyses the methods of synthesizing activated carbon-based electrodes, their structural properties, and the impact of activation conditions on their efficiency. Carbon aerogels, a significant category of materials, possess exceptional characteristics in terms of both their lightweight nature and structural strength. An analysis is conducted on the electronic conductivity of carbon aerogels, as well as their potential for increased capacity upon activation. Additionally, this article presents surface modification techniques that are designed to enhance the electrochemical properties of the electrodes.

Nanoporous carbon, which is classified as a third-class material, is formed by carbides. These new materials should be given attention in the development of effective electrodes for supercapacitors and lithium-ion batteries since they have a high specific surface area and porosity. A study was conducted to investigate the impact of temperature, chlorination duration, and synthesis time on the structural features and electrochemical properties of nanoporous carbon.

2. Condenser systems

The Leiden Jar, founded in 1745, is the fundamental model for contemporary electrolytic capacitors. Capacitors are ubiquitous in modern electronic devices and have applications across various domains of electrical engineering. In recent times, a multitude of novel capacitors have been created that function by diverse electrochemical mechanisms. Supercapacitors, sometimes referred to as electrochemical capacitors that utilize the charge energy of a double electric layer (DEL), are rapidly advancing in the realm of contemporary energy storage devices [1]. Electrochemical capacitors, as defined by Brian Evans Conway, are devices that undergo quasi-reversible electrochemical charge-discharge operations. The galvanostatic charge-discharge curves of these devices exhibit a nearly linear shape, indicating that their dependencies are comparable to those of ordinary electrostatic capacitors [2].

Electrochemical supercapacitors function as a bridge between dielectric capacitors, which have the ability to rapidly release a significant quantity of energy, and batteries, which have the capacity to store a substantial amount of energy. Supercapacitors (SC) can be used to complement or replace batteries in situations of high consumption and energy usage. These devices exhibit a greater efficiency in terms of energy conversion or uptake compared to batteries, while they possess a lower energy density. Supercapacitors are essential for controlling and balancing the load, as well as for supplementing or substituting batteries in situations that demand uninterrupted power supply, which is referred to as compensatory power supply [3].

Based on the manner of energy storage, supercapacitors are categorized into distinct classes, each with its own operating principles. These classes include capacitors with a double electric layer, pseudo-capacitors, and hybrid capacitors. Currently, the prevailing devices are capacitors equipped with a dual electric layer. These devices produce energy by creating an electric charge on highly porous materials in the active section of the electrodes. These devices do not experience oxidation/reduction events, which are referred to as Faraday processes. Instead, the charge builds up in pseudo capacitors or redox supercapacitors as a result of fast and reversible reactions that occur on the surface or near the surface. However, both batteries and pseudo capacitors frequently lack enough stability during cycling as a result of redox processes.

Hybrid capacitors are a novel integrated circuit (IC) that merges a battery electrode with either a capacitive or pseudo-capacitive electrode. These capacitors have the characteristics of both a capacitor and a battery. Capacitors can be categorized as symmetrical or asymmetrical based on the design of the cell [4].

2.1 Double-layer electrochemical capacitors

In 1957, Becker initially described the concept of DEL SC in a patent [5]. N.S. Lidorenko spearheaded the creation of the domestic DEL SC at the All-Union Scientific Research Institute of Current Sources. The devices were referred to as "molecular energy storage devices".

The charge builds up in the capacitors of the double electric layer as a result of the reversible adsorption of electrolyte ions by stable electrochemically active materials with a large specific surface area. During polarization at the electrode-electrolyte interface, there is no transfer of charge across the boundary between the electrode and the electrolyte. The charge mechanism involves the surface dissociation of the electrolyte and the adsorption of ions from the solution into the pores of the electrode. Consequently, there is either an

abundance or a deficiency of charge on the electrode's surface. Simultaneously, ions of opposite charge migrate towards the interface between the electrode and the electrolyte to uphold electroneutrality [6].

Helmholtz formulated the initial hypothesis regarding a double electric layer and drew a parallel between the double electric layer and a planar capacitor. Gui-Chapman, Stern, and Grem provided additional support for the concept [7].

During the charge-discharge cycle of supercapacitors, Faraday processes are absent. Instead, energy is solely accumulated by the electrostatic charges present on the opposing surfaces of the double electric layer. Consequently, these devices have the ability to operate for an extended period without experiencing a decrease in active mass, a phenomenon commonly seen in chemical power sources [8]. Although the electrochemical processes in secondary current sources are irreversible, the active mass of the electrodes is steadily diminishing. Therefore, the number of charge/discharge cycles that a battery can undergo during its lifespan varies, ranging from several thousand cycles, depending on the specific type of battery. The DEL SC exhibits excellent reversibility and may undergo a cyclic life of 10^5 - 10^6 cycles without forming new phases or intercalating [9]. The system exhibits quick charge and discharge capabilities with large currents, as it is not affected by polarization issues.

Figure 1 illustrates the structure of symmetrical double-layer electrochemical capacitors, which have two electrodes immersed in an electrolyte and separated by a membrane or separator. Separators are commonly employed in the utilization of diverse polymers, paper, or fiberglass. The electrodes consist of metal current collectors, often composed of aluminum or stainless steel, that have an active mass placed onto them. The active mass comprises a binder and a carbon substance with a highly developed surface. An electrically conductive additive, sometimes referred to as soot, may be added to the active mass in certain circumstances.

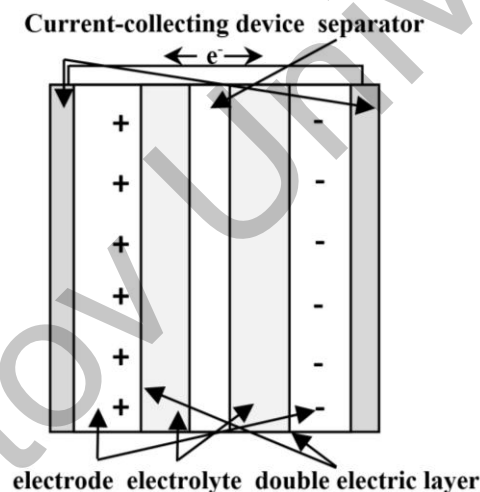


Fig.1. Schematic representation of double electric layer supercapacitor

The electrochemical process in DEL SC can be represented as follows:

- The positive electrode $E_s + A^- \leftrightarrow E_s^+ || A^- + e^-$;
- Negative electrode $E_s + K^+ + e^- \leftrightarrow E_s^- || K^+$;
- Total reaction $E_s + E_s + K^+ + A^- \leftrightarrow E_s^- || K^+ + E_s^+ || A^-$.

The surface of the electrode is denoted as E_s , and there is a double electric layer, represented by $||$, where charge accumulates on both sides of the electrode. The cations and anions of the electrolyte are denoted as K^+ and A^- , respectively [10]. When the integrated circuit is charged, the electrons are facilitated by the external environment to travel from the positive electrode to the negative electrode. The ions migrate towards the electrodes from within the electrolyte. Due to the load during discharge, electrons migrate from the negative electrode to the positive electrode, while ions reenter the electrolyte's volume and return to the surface. The electrolyte-impregnated separator prevents a short circuit between the electrodes while the supercapacitor is charged or discharged, while still allowing the passage of cations and anions. The charge density and electrolyte concentration fluctuate during the process of charging and discharging. The capacitance of a capacitor is determined by the surface area of its plates and the separation distance between them.

$$C = \frac{\epsilon_0 \epsilon S}{d}, \quad (1)$$

where ϵ_0 is the dielectric constant, ϵ is the relative permittivity, S is the surface area of the lining, d is the distance between the plates, C is the capacitance of the capacitor.

In the design of a supercapacitor, the distance between the electrodes is equivalent to the thickness of the double electric layer, which is similar in size to the dimensions of molecules. The magnitude of this value is roughly a few angstroms, contingent upon the dimensions of the ions and the concentration of the electrolyte. The capacitance of these capacitors is significantly lower compared to electrolytic capacitors that utilizes aluminum foil, in which a thin oxide film on the metal surface acts as a dielectric (with a thickness of approximately 10 nm). Furthermore, the utilization of contemporary carbon materials with a substantial specific surface area (ranging from 1500 to 2500 m²/g) enables the attainment of the most extensive surface area for the lining of the capacitor [11].

Figure 1 depicts a SC model comprising two capacitors connected in series. The porous electrodes facilitate the formation of electrical layers that enable current collection. The reason for this is that the ion layer created in the liquid at the interface serves as one of the plates of the supercapacitor. However, it is challenging to get a current collector with such characteristics from the liquid. The formation of the double electric layer occurs on a non-uniform surface, and not all parts of the electrode are equally accessible for wetting with electrolyte [12]. Currently, one of the key areas of research is the investigation of the relationship between the SC capacitance and the features of the electrode surface, including wettability, zero charge potentials, and the presence or absence of functional groups [13].

2.2 Pseudocapacitors

Pseudo capacitors are distinct from double-layer capacitors in that their capacitance is influenced by both electrostatic mechanisms and rapid Faraday reactions involving charge transfer. In this scenario, the active materials' surfaces undergo reversible redox reactions, resulting in "pseudo-intensive" behavior. The operation basis of capacitors of this sort involves the combination of two energy conservation mechanisms: electrostatic contact, which is comparable to what happens in capacitors with a double electric layer, and Faraday reactions, which are similar to what happens in batteries and accumulators [14].

Pseudo capacitor electrodes undergo a variety of Faraday processes, including those that are repeatable.

- For instance, the process of depositing lead or hydrogen onto the surface of titanium or aluminum, as well as the occurrence of redox processes involving transition metal oxides.
- For instance, compounds such as IrO₂, RuO₂, and different combinations of oxides [15] can be used.
- Additionally, electrodes made from conductive polymers can undergo repeated electrochemical doping and dedoping procedures [16].

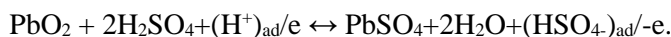
Ruthenium oxide-based pseudo capacitors exhibit a substantial capacity; nevertheless, their practical application is constrained by the exorbitant cost of the constituent raw ingredients. Experiments are carried out utilizing inexpensive substances such as iron, vanadium, nickel, cobalt, and manganese oxides [17]. Interest in carbon materials that utilize a charge accumulation mechanism in a double electric layer is justified due to their increased pseudo-capacity. However, both batteries and pseudo capacitors are frequently regarded as unstable during cycling as a result of redox processes [18].

2.3 Hybrid capacitors

Hybrid systems employ a solitary electrode with a "Faraday function" rather than two carbon electrodes that are identical, in contrast to double-layer electrochemical capacitors. Asymmetric hybrid supercapacitors possess two primary benefits. One reason is the higher specific energy resulting from the battery electrode, and another one is the higher specific power resulting from the capacitor type electrode [19]. When employing a non-polarizable electrode, the voltage of an individual component rises, resulting in an expansion of the operational voltage range. Removing the second capacitor that is linked in series in an equivalent circuit enhances the overall electrical capacitance of the capacitor. This combination of adjustments enhances the specific energy by a factor of two to five when compared to a symmetrical design. An advantage of an asymmetric system is its low self-discharge rate. This is due to the presence of only one carbon electrode in the cell, as opposed to two in a symmetrical capacitor. The carbon electrode is primarily responsible for the self-discharge in the system [20].

Hybrid capacitors encompass the subsequent systems:

- Examples of metal oxides used with aqueous electrolyte solutions include NiOOH|KOH|C [21] and PbO₂|H₂SO₄|C systems. Regarding the latter system, the overall reaction can be expressed as follows:



The DES (Dual Energy Storage) system undergoes recharging at the negative electrode [22].

Graphite [4], LiMnO₂, Li₂MnSiO₄, Li₄Ti₅O₁₂, and other related compounds have the ability to reversibly intercalate lithium ions in non-aqueous electrolytes. These systems closely resemble normal lithium-ion systems. One instance is the suggestion of a hybrid electrode utilizing nanostructured β-Fe OOH [23]. The development of hybrid systems has resulted in the creation of novel electrode materials for lithium-ion batteries. As an illustration, the system consists of nanostructured Li₄Ti₅O₁₂ and a positive electrode composed of activated carbon [24]. Figure 2 illustrates the operational principle of these systems.

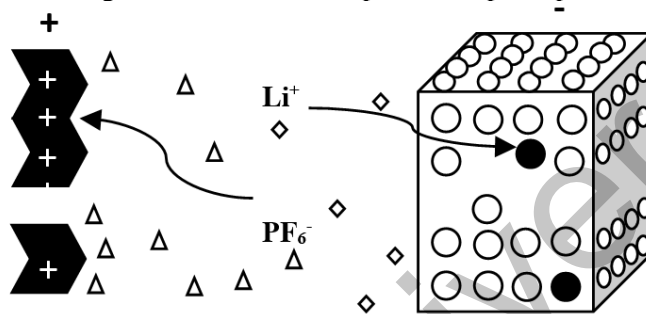


Fig.2. Schematic representation of a hybrid integrated circuit [24]

The lithium titanate electrode offers high power output without the formation of a "solid electrolyte" layer. Furthermore, Li₄Ti₅O₁₂ exhibits minor changes in volume during the charging/discharging process, making it highly resistant to cycling [25]. Following the examination of Li₄Ti₅O₁₂, numerous subsequent experiments were carried out utilizing different combinations of lithium-containing electrodes in conjunction with capacitive carbon electrodes. The emergence of nanomaterials and significant advancements in lithium-ion battery technology are expected to drive the creation of high-performance hybrid supercapacitors. These supercapacitors will utilize a lithium-containing electrode to enable fast charging and discharging.

3. Materials of DEL SC electrodes

Carbon's distinctive chemical and physical properties make carbon compounds appealing for use as electrodes. The properties of this material encompass excellent conductivity, resistance to corrosion, stability under high temperatures, the capacity to regulate the porous structure, affordability, eco-friendliness, and potential for utilization in composite materials. Double layer charging enables the utilization of electrodes that possess highly advanced characteristics such as specific surface area, open porosity, electronic conductivity, and electrochemical stability. This allows for the attainment of a high and reversible capacitance. Currently, ongoing research involves the utilization of activated carbons, nanoporous carbon derived from carbides, carbon fabric, nanotubes and nanofibers, spheroidal carbon structures, aerogels, and other materials [26].

3.1 Activated carbon

Activated carbons (AC) are widely used materials due to their abundant availability, large surface area, and affordable price. Various affordable and readily available raw materials such as rice husks, coconut fibre, sunflower cake [27], animal bones, apricot kernels, straw [28], etc., can be utilized for the manufacturing. The two prerequisites for eliciting a response from an asynchronous machine are carbonation and activation. Conversely, carbonation refers to the transformation of organic matter into carbon through the application of heat in an oxygen-free environment. During the carbonization process, hydrolysis occurs, resulting in the creation of graphite plane pieces. The temperature ranges from 300 to 500 °C. During the carbonization process, it may be necessary to use elevated temperatures, such as 600-1000 °C. The temperature at which carbonation occurs has a significant impact on the composition and arrangement of coal (table 1).

Table 1. Properties of carbon materials after their processing.

Method	Result
Oxidation in the liquid phase (by oxygen-containing acids)	An increase in surface area and porosity, a decrease in density, an increase in the concentration of surface functional groups.
Oxidation in the gas phase (O ₂ , H ₂ O)	An increase in surface area and porosity, a decrease in density, an increase in the concentration of surface functional groups.
Plasma treatment	Increased surface area and porosity, improved wettability, increased concentration of surface functional groups, reduced electrical resistance, increased electrocatalytic activity for the oxygen reduction process.
Heat treatment in an inert environment (argon, nitrogen)	Increase in surface area and porosity, increase in density, decrease in the concentration of surface functional groups, increase in the degree of graphitization.

Functional groups have the ability to influence several characteristics of the supercapacitor, such as the rate at which it discharges on its own, the ability of the electrode to absorb the electrolyte, the resistance at the contact points, and the angle at which the electrolyte wets the electrode surface. Functional groups present at the interface of coal, electrolyte, and current collector might induce chemical reactions, resulting in self-discharge currents and degradation of the electrolyte. Faraday reactions occurring in symmetrical capacitors of the double electric layer typically result in a reduction in the cyclic lifespan of the system. The surface of activated carbon materials may contain functional groups that contain oxygen and hydrogen, small amounts of sulphur, nitrogen, halides, and remnants of organic precursors resulting from incomplete carbonation.

3.2 Carbon Aerogel

Carbon aerogel-based supercapacitors have gained significant popularity. This material possesses exceptional characteristics of both lightness and strength, making it highly distinctive. Additionally, it exhibits an extraordinarily porous surface with a surface area ranging from 400 to 2000 square meters per gramme (m²/g). Aerogels can be produced through the pyrolysis of formaldehyde polycondensation products. The porosity structure of aerogels is determined by modifications in the parameters of the polycondensation process. Aerogels are solid, film-like structures composed of interconnected polymer chains. After pyrolysis, the polymer chains undergo a transformation and are converted into carbon, while maintaining the same structure. Aerogel exhibits a considerably greater level of electronic conductivity compared to activated carbon. Upon activation of the aerogel, there is a significant increase in surface area, resulting in a corresponding rise in capacity. Nevertheless, the holes generated during the activation phase can be excessively narrow for the electrolyte to reach. The literature also presents information regarding the potential for altering the surface of activated aerogel. This modification enhances the ability of the material's pores to be wetted by electrolyte, resulting in improved electrochemical properties of the supercapacitor [29].

3.3 Nanoporous carbon

Nanoporous carbon, also known as NPC or CDC-carbide generated carbon, is a novel carbon material characterized by its exceptionally large specific surface area and porosity. These properties make it highly appropriate for utilization in the electrodes of double-layer supercapacitors and hybrid SC systems. NPC is produced through the process of chlorinating carbides of elements like Al, Ti, B, Si, Mo. This involves exposing the carbides to a continuous flow of chlorine while heating them to a temperature range of 600-1200 °C. In this reaction, the metal is separated from the carbide as a result of the creation of chlorides that have a low boiling point. The carbon rearranges itself into structures that are predominantly amorphous, disordered, and composed mostly of sp² bonds.

For example, for titanium carbide: $\text{TiC} + 2\text{Cl}_2 \rightarrow \text{C} + \text{TiCl}_4\uparrow$.

For aluminum carbide: $\text{Al}_4\text{C}_3 + 6\text{Cl}_2 \rightarrow 3\text{C} + 4\text{AlCl}_3\uparrow$.

The pore size is determined by the composition of the original carbide and can be modified through technological means such as adjusting temperature or chlorination duration. The surface area of these materials, as determined by the BET technique, ranges from 1000 to 2000 m²/g. Micropores, with a size

ranging from 0.7 to 1 nm, mostly develop at a temperature of 800 °C. Mesopores, with a size ranging from 2 nm, are also generated under the same temperatures. As an illustration, the NPC derived from TiC had a pore size ranging from 0.7 to 1.1 nm, and a specific surface area (S_{BET}) of 1000-1600 m²/g. Simultaneously, the specific surface area exhibited an increase as the average pore size decreased.

The scientists from the University of Tartu conducted research where they acquired and examined the characteristics of NPC that were synthesized from various carbides, including TiC, Al₄C₃, B₄C, α -SiC, MO₂C, VC, WC, TaC, NbC, HfC and ZrC. The nanoporous materials derived from vanadium, niobium, tantalum, hafnium, and zirconium carbides exhibit optimal characteristics for use in DES supercapacitors, owing to the favorable ratio between nanopore dimensions and surface area. Simultaneously, the surface area of tantalum carbide NPC (2275 m²/g) ranks among the greatest among nanoporous carbons [30].

Considering the high cost and limited availability of these materials, as well as the combination of surface area and pore size, the most appropriate mass electrical material is NPC derived from titanium carbide at temperatures ranging from 700 to 900 °C.

It is important to mention that, similar to the materials mentioned earlier, not all holes of nanoporous carbon are suitable for usage with solvents. As a consequence of the electrolyte's inability to moisten some pores, a portion of the surface may stay inactive. Therefore, the computed capacity of the double layer on a highly porous carbon material does not align with the real value. Deviation in the incomplete wetting of pores can typically be attributed to the presence of numerous flaws, closed pores, a disordered structure, and surface heterogeneity. To enhance the electrochemical characteristics of the completed supercapacitor, it is imperative to select carbon materials that possess an appropriate porosity configuration for the specific electrolyte. In order to achieve complete infiltration of ions into the electrode's pores and the creation of a deep eutectic solvent, it is necessary for the minimum radius of the pores to align with the significant dimensions of the electrolyte ions. In order to generate a concentrated and spread out section of the double electric layer, it is imperative to augment the size and surface area of the active pores. The presence of larger mesopores also plays a role in the formation of DEL.

4. Conclusions

Ultimately, the authors stress the necessity for additional investigation to optimize the efficiency of carbon material synthesis for the purpose of attaining superior electrochemical characteristics. The article discusses the potential of carbon materials in lithium-ion batteries and supercapacitors, emphasizing the need for a careful balance between specific surface area, porosity, and structural stability in order to develop efficient electrodes. This review article not only provides information for present researchers, but also highlights essential issues for future research focused on creating novel materials for sophisticated electrochemical devices like lithium-ion batteries and supercapacitors.

Conflict of interest statement

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

CRedit author statement:

Dyussebayev M.Sh.: writing—original draft preparation; Khabiev A.T.: supervision; Rafikova Kh.S.: Conceptualization; Afanasyev D.A.: writing—review and editing.

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