

## Nanocellulose-Based Separators in Lithium-Ion Battery (Pemisah Berasaskan Nanoselulosa dalam Bateri Litium Ion)

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Received: 12 December 2023/Accepted: 22 December 2023

### ABSTRACT

Lithium-ion batteries are the essential parts of many portable electronic devices. They draw substantial recognition by virtue of their advantages over other batteries. Separators are critical to the working of lithium-ion batteries. The separator is a key component of lithium-ion battery that isolates the cathode and anode. Though the separator does not participate in the electrochemical process, it plays a major role in the safety of the battery. Nanomaterials are used in many applications in our life. Nanocellulose are sustainable materials used to fabricate parts of energy storage devices. This review discusses the potential of nanocellulose incorporated materials to use as separators in lithium-ion batteries.

Keywords: Bacterial nanocellulose; cellulose nanocrystal; lithium-ion battery; nanocellulose; separators

### ABSTRAK

Bateri litium-ion adalah bahagian penting bagi kebanyakan peranti elektronik mudah alih. Ia mendapat pengiktirafan yang besar berdasarkan kelebihan mereka berbanding bateri lain. Pemisah adalah penting untuk bateri litium-ion (LIB) berfungsi. Walaupun pemisah tidak melibatkan proses elektrokimia, ia memainkan peranan utama dalam keselamatan bateri. Bahan nanoselulosa adalah bahan yang berpotensi untuk pelbagai aplikasi termasuk dalam sektor tenaga. Kajian ini meneroka potensi nanoselulosa untuk digunakan sebagai pemisah dalam bateri ion litium. Kajian ini menyerlahkan penyelidikan dalam bidang ini dengan tumpuan khusus pada bahan nanoselulosa berbeza daripada sumber semula jadi seperti selulosa nanofibril (CNF), selulosa nanohablur (CNC) dan bakteria nanoselulosa (BC) untuk memasang pemisah di dalam bateri litium-ion. Secara keseluruhannya, ulasan ini memberikan pandangan yang lebih mendalam tentang sumbangan bahan pemisah yang digabungkan dengan nanoselulosa kepada keselamatan dan prestasi bateri litium-ion.

Kata kunci: Bakteria nanoselulosa; bateri litium-ion; nanoselulosa; pemisah; selulosa nanohablur

### INTRODUCTION

Many countries are now focusing on the research for the development of batteries to reduce and eventually eliminate carbon emissions. Energy storage devices

convert chemical energy into electrical energy and *vice versa* (Wang et al. 2014). Batteries can be classified either primary or secondary battery. A primary battery can be used for a single time. However, secondary battery

can be used many times. Secondary batteries are also known as rechargeable batteries. Lithium-ion, lead-acid, zinc-air, nickel-cadmium, nickel-hydrogen, and sodium-sulfur batteries are some examples of rechargeable batteries. Lithium-ion batteries have high energy density, low self-discharge rate, long cycle life and high open-circuit voltage (Liu et al. 2019; Sunden 2019). Table 1 compares the characteristics of Li-ion, lead-acid, Nickel-Cadmium, Nickel metal hydride rechargeable batteries.

The lithium ion battery is an energy storage device widely used in many applications. A rechargeable lithium-ion battery with intercalated titanium disulphide cathode and a lithium-aluminum anode was developed in 1976. The modification was continued to develop a new rechargeable battery using lithium cobalt oxide as the cathode and the performance of this battery was better than the earlier batteries (Goodenough 2012). In

1991, Yoshino then realized that charging the graphitic anode with Li from a discharged  $\text{LiCoO}_2$  cathode produced high-energy density Li-ion rechargeable battery (Yoshino 1985). Both researchers awarded the Nobel Prize for Chemistry in 2019 for the development of Li-ion batteries. A schematic diagram of an LIB is presented in Figure 1.

Lithium ion batteries are used in portable electronic devices such as cellular phones, laptop computers, digital cameras, smart watches, drones, tablets and other electronic devices (Goodenough & Park 2013; Kim et al. 2019; Nitta et al. 2015; Tarascon & Armand 2001), stationary energy storage systems, and electric vehicles (EVs) (Marsh et al. 2001). Moreover, lithium ion batteries are applied in backup power supply, military applications (Suriyakumar et al. 2018), and aerospace applications (Zifan et al. 2018).

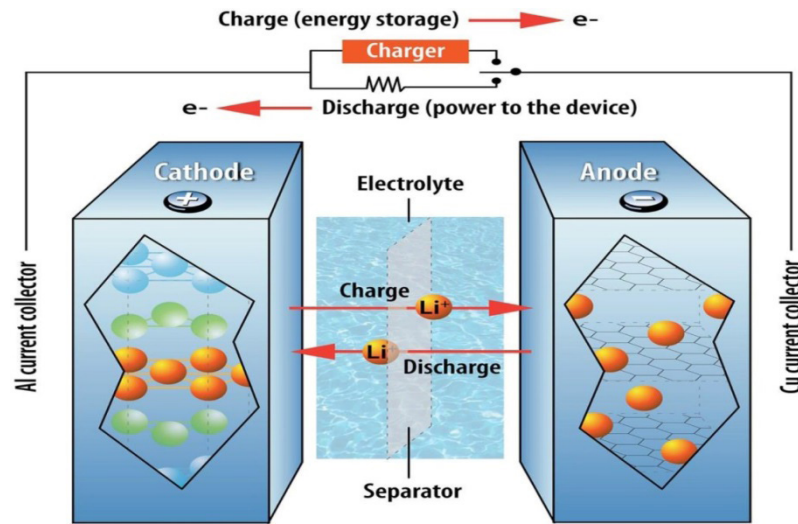


FIGURE 1. Schematic diagram of LIB (Reproduced with permission (Nzereogu et al. 2022))

TABLE 1. Comparison of characteristics of lithium-ion, lead-acid, nickel-cadmium, and nickel-metal hydride batteries (Townsend & Gouws 2022)

Specifications	Unit	Li-ion battery	Lead acid battery	Nickel-Cadmium	Nickel metal hydride
Energy density	Wh/kg	100–265	30–40	40–60	60–120
Power density	W/kg	250–340	180	150	250–1000
Cycle life	Cycles	400–1200	<1000	2000	180–2000
Charge/discharge efficiency	%	80–90	50–95	70–90	66–92

The Li-ion battery consists of cathode, anode, electrolyte and separator. The LIB cathode materials are transition metal oxides containing lithium.  $\text{LiCoO}_2$ ,  $\text{LiMn}_2\text{O}_4$ , and  $\text{LiFePO}_4$  are common LIB cathode materials available commercially. Anode materials used frequently in LIBs include carbon materials such as graphene and carbon nanotubes, transition metal oxides, silicon, and alloys of metals such as Sn, Sb, Al, Mg, and Ag. The electrolyte for LIBs is a mixture of cyclic carbonate esters, linear carbonate esters, and an electrolyte salt compound such as  $\text{LiPF}_6$  or  $\text{LiBF}_4$ . Commonly used cyclic carbonate esters are ethylene carbonate and propylene carbonate, while commonly used linear carbonate esters are dimethyl carbonate and diethyl carbonate. The electrolyte contains lithium ions generated as a result of the electrochemical reaction in the battery. In the course of charging and discharging, the lithium ions in the electrolyte pass from one electrode to the other by crossing the separator, and the electrons traverse through an external circuit (Megahed & Scrosati 1994; Scrosati & Garche 2010).

A separator is a porous electrolyte-permeable membrane that is positioned in the middle of the oppositely charged solid anode and cathode to separate them and prevent electrical short circuit. The porous structure of a Li-ion battery separator is filled with the liquid electrolyte, and it allows the transport of lithium ions between the two electrodes (Arora & Zhang 2004; Lagadec, Zahn & Wood 2019). The separators can be classified as microporous membranes, nonwoven membranes, electrospun membranes, composite membranes, and polymer blend membranes (Barbosa et al. 2021).

Commercial polymers such as polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET), polyacrylonitrile (PAN), and polyvinylidene fluoride (PVDF) and its copolymers such as polyvinylidene fluoride-hexafluoropropylene (PVDF-HFP) are widely used in LIBs as separators due to their chemical stability, excellent mechanical properties, better material properties, and shutdown property (Deimede & Elmasides 2015; Lee et al. 2014).

#### NANOCELLULOSE IN ENERGY STORAGE DEVICES

Cellulose materials are applied in many electrochemical energy storage devices (Teng et al. 2023). The demand for materials prepared from renewable, non-petroleum-based reserves lead to the development of cellulose and its derivatives as sustainable materials for electrochemical

energy storage devices (Kamalesh et al. 2023). Cellulose and its derivative materials, are used in batteries, supercapacitors and other energy storage devices because of their abundance, sustainability, improved electrochemical performance and low cost (Eichhorn et al. 2022). Cellulose is the most abundant polymer in the world. It is a linear homopolymer of -1, 4-linked anhydro-d-glucose units (Kargarzadeh et al. 2018; Klemm et al. 2011, 2005). Cellulosic materials in nanoscale are known as nanocellulose (Maity et al. 2023).

Nanocellulose used as separators, electrodes and electrolytes are used in Li-ion batteries (John et al. 2023). Nanocellulose has advantages over other nanoparticle fillers to use in the polymer due to its natural origin, abundance, renewability, low cost, nontoxicity, and better mechanical properties (Wegner & Jones 2006). Cellulose nanomaterial-based materials are used in battery separators because they offer better ionic conductivity as well as electrochemical, thermal, and mechanical stability (Wegner & Jones 2006; Yang et al. 2012). The better ionic conductivity of nanocellulose is due to the hydrophilic hydroxyl surface groups present in cellulose (Xu et al. 2021; Zhang et al. 2018). Although many review articles written on Li-ion battery separators, few have focused on nanocellulose-incorporated separator materials. This review has been written by considering the growing applications of cellulose nanomaterial in lithium-ion battery applications.

#### ADVANTAGES OF INCORPORATION OF NANOCELLULOSE IN LITHIUM ION BATTERY SEPARATORS

Polyolefin separators have a few disadvantages due to their structure. Most commonly available commercial polyolefin separators, PP and PE have low thermal stability due to their low melting temperatures ( $\sim 165$  °C for PP and  $\sim 135$  °C for PE). Thus, Li-ion battery separator with polyolefin separators can be used only at low temperatures, i.e., below their melting temperatures. Furthermore, the lower dimensional stability of polyolefin separators is by the reason of its porous architecture through stretching during their manufacturing process. The low melting temperature of polyolefin separators is problematic when they are used in LIBs for EV applications. In such applications, the Li-ion battery work at high temperatures, causing dimensional shrinking and melting of the polyolefin separator. In addition, at high temperature, the mechanical strength diminishes and that causes hot spots, which

leads to short circuit and ignition (Heidari & Mahdavi 2019; Nestler et al. 2014; Wang et al. 2019; Yang & Hou 2012). Moreover, the hydrophobic nature due to the lack of polar surface groups and the low porosity of the polyolefin separators brings about decline in absorption of electrolyte and ionic conductivity (Chen et al. 2023; Liu et al. 2018).

The incorporation of inorganic or organic nanofillers into polyolefin separators can make the thermal, dimensional, and mechanical strength of Li-ion battery separator better. In addition, it helps to improve the electrolyte uptake and maintain the porosity of polyolefin separators (Muchakayala et al. 2023). However, such nanofillers are prepared by complex methods; they are not cost-effective or lightweight and they cannot be processed easily. Moreover, inorganic nanoparticles are toxic and they show poor bonding with the organic polyolefin matrix in the composite (Miao et al. 2023; Zhang et al. 2018).

Cellulose can be obtained from nature, either from plants or from microorganisms, in large amounts. It is nontoxic and renewable as well (Trache et al. 2017). The first degradation of cellulose occurs around 275 °C, which provides cellulose nanomaterial-incorporated composite separators with better thermal and dimensional stability than commercial polyolefin separators (Szabó et al. 2023). The materials used in present day batteries are costly and not renewable or sustainable. In this regard, materials of natural origin are increasingly attracting attention to fabricate battery components including separators. The advantages of natural material are wide natural availability, low-cost, and has low environmental impact (Wang et al. 2023).

Cellulose nanomaterial, possesses a large surface area and high surface energy, which provides cellulose nanomaterial-based separators with good electrolyte wettability and ionic conductivity (Xu et al. 2021). Furthermore, the hydrophilic hydroxyl surface groups present in cellulose also contribute toward the improved electrolyte wettability and ionic conductivity (Wu et al. 2023). Cellulose has lower density than steel. However, when the density difference is considered, cellulose is stiffer than steel. Moreover, the stiffness of cellulose is comparable to CNT (Jakob et al. 2022; Kim et al. 2015; Moon et al. 2011; Yan et al. 2023). Furthermore, cellulose nanomaterial-incorporated separators offer high mechanical strength (Mondal et al. 2019; Zhu et al. 2016).

The shape, size, purity, and crystallinity of nanocellulose changes with the cellulose source. These properties determine the specific applications of cellulose nanomaterials. Cellulose can be extracted from wood, plants, tunicates, algae, fungi, and bacteria. The nanocellulose can be classified into three: cellulose nanofibril (CNF), cellulose nanocrystal (CNC), and bacterial nanocellulose (BNC) (Abitbol et al. 2016; Leong et al. 2023; Moon et al. 2011). Figure 2 shows the morphological images of CNF, CNC, and BNC. CNF and CNC are prepared by degrading the plant cell wall structure using chemical/enzymatic or mechanical methods to nano size. On the other hand, BNC can be obtained by low molecular weight sugars by certain strain of bacteria in a controlled environment (Cherian et al. 2022; Pradhan, Jaiswal & Jaiswal 2022). Figure 2 shows transmission electron micrograph images of (a) CNF (b) CNC and (c) BNC.

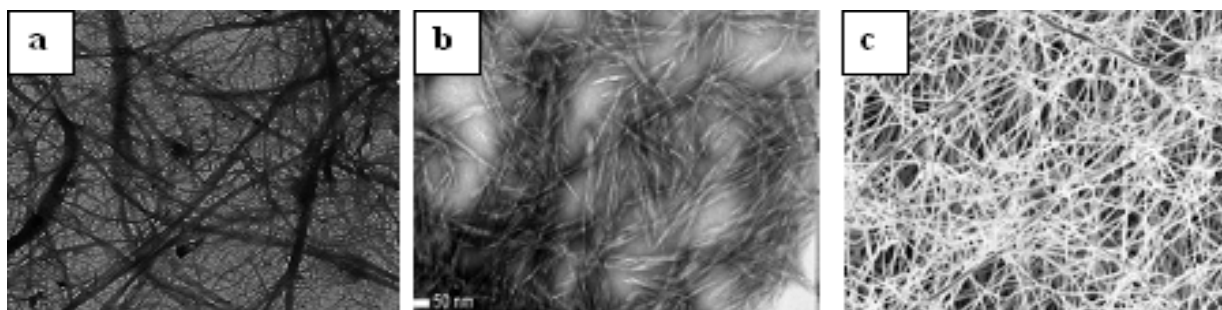


FIGURE 2. Transmission electron micrograph images of (a) CNF (Alemdar & Sain 2008), (b) CNC (Xu et al. 2013), (c) BNC (De Lima Fontes 2019) (Reproduced with permission)

The wide availability, natural origin, renewability, biodegradability, low cost, light weight, nontoxicity, availability of hydrophilic surface groups, easy preparation and processing methods, inherent mechanical strength, and thermal stability make cellulose nanomaterials a better alternative to inorganic nanoparticles and other carbonaceous fillers for use as a nano-reinforcement in composite separators. The fascinating structure and attributes of cellulose nanofibrils (CNF), such as the high aspect ratio, web-like entangled structures, and outstanding mechanical properties, have enable it to use in LIBS. Table 2 compares the properties of CNF, CNC, and BNC. It can be seen cellulose nanocrystals (CNC) is one of the toughest and

stiffest organic material with a high aspect ratio, bending strength of around 10 GPa, tensile strength of up to 7.5 GPa, and Young's modulus of approximately 140 GPa; it is a light weight material with a density of 1.5-1.6 g.cm<sup>-3</sup> (Lei et al. 2020).

CNC could be an ideal candidate for reinforcing the LIBS owing to its promising properties mentioned earlier. Bacterial nanocellulose (BNC) fibers have a huge aspect ratio with a diameter of 20-100 nm, tensile strength in the range of 200-300 MPa, Young's modulus up to 15-35 GPa, high crystallinity of 80-90%, and thermal stability around 360 °C. Owing to the superior attributes of BNC stated earlier, it has recently attracted substantial attention to use in numerous energy storage systems (Lei et al. 2020).

TABLE 2. Comparison properties of CNF, CNC, and BNC (Kim et al. 2019; Klemm et al. 2011; Zhang et al. 2018)

Material	Tensile strength (GPa)	Young's modulus (GPa)	BET N <sub>2</sub> surface area [m <sup>2</sup> g <sup>-1</sup> ]	Diameter (nm)	Length (nm)	Aspect ratio
CNF	2-6	50-160	≈1000	10–80	500–2000	10-100(>300)
CNC	7.5-7.7	50-140	≈200	5-30	≈100	20-100
BNC	2	15-35	≈200	20-100	25-86	—

#### CONCLUSION

Li-ion batteries are influencing our daily life. Li-ion battery separators are widely used as the energy storage device in portable electronic devices. The separator in Li-ion battery prevent the physical contact between cathode and the anode by isolating them. Nanocellulose materials, are used in batteries, supercapacitors and other energy storage devices because of their sustainability and improved electrochemical performance. As discussed in this review, nanocellulose is a suitable functional component to use in high performance Li-ion battery separators. The nanocellulose -incorporated composite Li-ion separators show better thermal and dimensional stability, mechanical strength, electrolyte wettability and ionic conductivity than commercial polyolefin separators.

#### ACKNOWLEDGEMENTS

The authors are thankful to the Government of Malaysia and India for the support. Universiti Kebangsaan Malaysia is also acknowledged for the instrumentation facility and financial support (FRGS/1/2022/STG05/UKM/01/1). Authors also thankful to Mahatma Gandhi University, Kottayam, India for the support.

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