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# From papyrus to flexible electronic devices: the revolution of cellulose nanofibrils

## De los papiros a los dispositivos electrónicos flexibles: la revolución de las nanofibrillas de celulosa

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## Abstract

The isolation of cellulose nanofibrils as a native element from cellulose fibers, the main component of paper, has provided novel and exciting opportunities for the development of electronic devices that are flexible and more environmentally friendly. An important field of work has targeted the use of cellulose nanofibrils as the support to produce flexible electronics owing to the material's advantageous properties, including high mechanical strength (stronger than most plastics), high optical transparency, and good thermal stability. Moreover, in recent years cellulose nanofibrils have been explored as a functional component for the development of flexible electronic devices, including as a replacement for the dielectric layer in transistors, or as the electrolyte for energy storage devices. Despite significant challenges remaining, including cost, scalability, and moisture sensitivity, due to their remarkable properties and the increasing importance of reducing the environmental impact of electronic devices, cellulose nanofibrils are expected to play a crucial role in the development of next-generation flexible electronics.

*Keywords*: **cellulose nanofibrils, flexible electronic devices, organic light emitting diodes, transistors, energy storage devices.**

## Resumen

El aislamiento de las nanofibrillas de celulosa a partir de fibras de celulosa, el principal componente del papel, ha proporcionado oportunidades novedosas y apasionantes para el desarrollo de dispositivos electrónicos flexibles y más respetuosos con el medio ambiente. Un importante campo de trabajo se ha centrado en el uso de las nanofibrillas de celulosa como soporte para producir electrónica flexible debido a las ventajas del material, entre las que destacan su gran resistencia mecánica (es más fuerte que la mayoría de los plásticos), su alta transparencia y su estabilidad térmica. Asimismo, recientemente se ha explorado el uso de las nanofibrillas de celulosa como componente funcional en el desarrollo de dispositivos electrónicos flexibles, en sustitución de la capa dieléctrica en transistores, o como electrolito para dispositivos de almacenamiento de energía. A pesar de retos importantes pendientes, como el coste, la escalabilidad, y la sensibilidad a la humedad, se espera que, debido a sus propiedades excepcionales y a la importancia cada vez mayor de reducir el impacto medioambiental de los dispositivos electrónicos, las nanofibrillas de celulosa desempeñen un papel crucial en el desarrollo de la electrónica flexible de próxima generación.

*Palabras clave*: **nanofibrilas de celulosa, dispositivos electrónicos flexibles, diodos orgánicos emisores de luz, transistores, dispositivos de almacenamiento de energía.**

## 1. Introducción

The fabrication of paper-like products can be traced back to ancient Egypt before 2000 B.C. At the time, damped strips of a plant of the Cyperaceae family were placed side by side alternating vertical and horizontal layers (Capua, 2015). To this date, the legacy of ancient Egypt remains in the word paper which is etymologically derived from the Cyperus papyrus plant. More than 2,000 years after the invention of papyrus, the first instance of papermaking was reported in China. The method resembled the ones used in the present day: a puree of fibers isolated from hemp, bamboo, or other plants pressed together afforded thin sheets of paper (American Forest & Paper Association, 2021). Since these ancient times, paper has remained a ubiquitous product in our daily lives. Its traditional applications have included printing, publishing, packaging, writing, stationery, arts, and crafts, and so on. However, early in the 20th century, a less commonly known, and less intuitive use of paper, sparked much interest: electrical insulators (Emsley and Stevens, 1994). At this time the application of paper was studied in oil-filled power transformers and power cables. Composed of fibers of cellulose, paper possesses attractive properties to produce electronic devices, including high electrical resistivity, high electrical strength, flexibility, as well as chemical and thermal stability. Moreover, cellulose and paper are readily available from renewable sources (plants) as low-cost products.

In the mid-20th century, the interest in fabricating electronics derived from paper was propelled by the observation of nanosized crystalline cellulose in the cell walls of plants (Nickerson and Harree, 1947). Since then, researchers determined that cellulose is composed of small fibers intricately arranged, also called macro fibers. In turn, the macro fibers are made of tinier ones that reach down the nanoscale, known as “cellulose nanofibrils”, with a diameter 1000 times smaller than a human hair (Fig. 1a). The discovery, and later isolation, of cellulose nanofibrils (CNF), have triggered immense interest from the industry and academia (Thomas *et al.,* 2018). CNF can be produced using bacteria (bacteria species such as Acetobacter) or obtained from biomass feedstock, including waste products such as tree bark, leaves, and corn husk (Kim *et al.,* 2015; Rajinipriya *et al.,* 2018). In the latter, the feedstocks are typically subjected to chemical treatments to remove lignin and other extractives native to the feedstock and obtain pure cellulose fibers. The fibers are further processed by a high-performance grinder (also called supermass colloider) in a fibrillation step, where the energy provided is sufficient for the rupture of the bonds existing between the fibers and yield CNF.

Using processes resembling those of traditional papermaking, such nanofibrils can be assembled to make a nanopaper. Excitingly, a nanopaper exhibits properties radically different from paper. For example, a nanopaper is optically transparent due to the small size of the nanofibrils (Fig. 1). Furthermore, the existence of chemical interactions between nanofibrils through the hydroxyl groups of cellulose induces tremendous improvements in their mechanical strength. CNF nanopapers are 15 times stronger than commercial paper and several-fold more robust than most petroleum-based plastics (polyethylene, polypropylene, polyvinyl chloride, polyamide, for instance) (Kim *et al*., 2015).

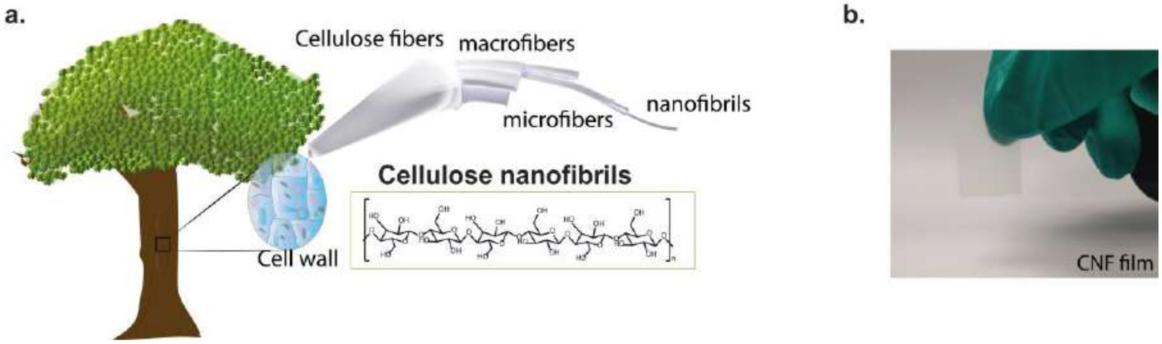

**Figure 1. a.** Architecture of cellulose fibers from wood (Copyright 2023, Elsevier), (Tanguy *et al*., 2023); **b.** Transparent CNF nanopaper.

**Figura 1. a.** Arquitectura de las fibras de celulosa de la madera (Copyright 2023, Elsevier), (Tanguy *et al*., 2023); **b.** Nanopapel de CNF transparente.

CNF nanopapers have emerged as a compelling solution to reduce our dependence on petroleum-based plastics. Meanwhile, the unique properties of CNF nanopapers have expanded the possibilities for the fabrication of electronic devices that are sustainable and environmentally friendly (Hoeng *et al*., 2016; Wawrzyniak *et al*., 2021; Tanguy *et al.*, 2023). This becomes particularly significant as the production of electronic devices continues accelerating, which has caused the accumulation of electronic waste (e-waste) in the environment. Specifically, with a 3-5 % annual growth rate across the world, reaching 53.6 million tons in 2019, e-waste is now one of the fastest-growing waste streams worldwide (Liu *et al*., 2023). Despite the short turnover of common electronic devices (for example, the average use of cellphones is 3 years), the plastic components including casing, circuit board, and display will remain in the environment for more than 100 years. Stemming from the remarkable properties of CNF, nanopapers are expected to provide a high-performance and more environmentally friendly alternative to the previously mentioned plastic components used in electronic devices, thereby contributing to reducing e-waste.

Flexible electronics refer to an emerging class of devices that can bend to various shapes (Corzo *et al*., 2020). The fabrication of these devices contrasts with traditional electronics, wherein rigid substrates such as silicon wafers, epoxies, and polyurethanes, are used as components. Flexible electronics are more versatile as the devices can be integrated onto various objects and surfaces, including clothes, and even the human skin. Most notably, these characteristics enable the design of a novel generation of wearable electronic devices, including flexible devices for computation, sensors, and energy storage. For example, wearable gas sensors combined with energy storage devices can be used to ensure workers' safety in chemical factories or monitor individuals' health in cities, by monitoring gas concentration in real-time and alerting the user in case of exposure to harmful gases.

## 2. Next-generation CNF-based flexible electronic devices

One of the first reports of the use of CNF for flexible electronics can be traced back to 2007 when researchers demonstrated the successful deposition of semiconducting materials onto CNF (Van Den Berg *et al.,* 2007). The obtained system was a flexible semiconducting nanopaper with a combination of high mechanical strength and electrical conductivity. This initial effort paved the path to the development of more complex and flexible electronic devices, for example as organic light-emitting diodes (OLED) used for displays, in the fabrication of CNF-supported flexible transistors (fundamental for computation), and as energy storage devices (supercapacitors and batteries) (Hoeng *et al*., 2016).

### Organic light-emitting diodes (OLED)

In the following years, the fabrication of CNF-supported flexible OLED was achieved (Nogi and Yano, 2008; Okahisa *et al*., 2009), which stood as a remarkable leap as OLEDs are fundamental to the development of modern displays. OLED displays allow for large viewing angles, a high contrast ratio, and are lighter than other technologies (Huang *et al.,* 2020). Thus, the application of CNF permits the design of displays that can be bent, folded, and rolled. OLED technology consists of multiple layers of materials sandwiched between a cathode and an anode. As a voltage is applied in between the electrodes, electrons are injected into the multiple layers of materials comprised in between. Specifically, the positive charges, and negative charges, are injected into the hole transport layer, and the electron transport layer, respectively. The electron and hole recombine in the emissive layer to produce an exciton. The exciton then releases energy in the form of light as it returns to its ground state (Fig. 2). The fabrication of flexible displays requires the integration of OLED components onto a flexible substrate such as CNF nanopapers, which are advantageous as compared to traditional petroleum-based plastics. Specifically, traditional polymeric materials are prone to expand or retract when subjected to changes in temperature, which can cause localized mechanical stresses and deformation either during processing or use. CNF nanopapers have a coefficient of thermal expansion more than 40-fold inferior to that of plastics. This is a significant advantage, as the deposition of the OLED components is traditionally made by thermal processes, wherein repeated changes in temperatures can cause fracture during processing. Heat production by the Joule effect during use can cause a similar phenomenon. Nevertheless, notable technical challenges remain before envisioning the application of CNF as a substrate for OLEDs, including the progressive yellow coloring during aging, and the alterations in shapes (swelling for instance) caused by the deposition of OLED components. Typical strategies to alleviate these limitations have involved the preparation of nanopaper composites, in which CNF are added as fillers into a polymer matrix to reduce thermal expansion and improve mechanical performance (Okahisa *et al.,* 2009; Tao *et al*., 2020).

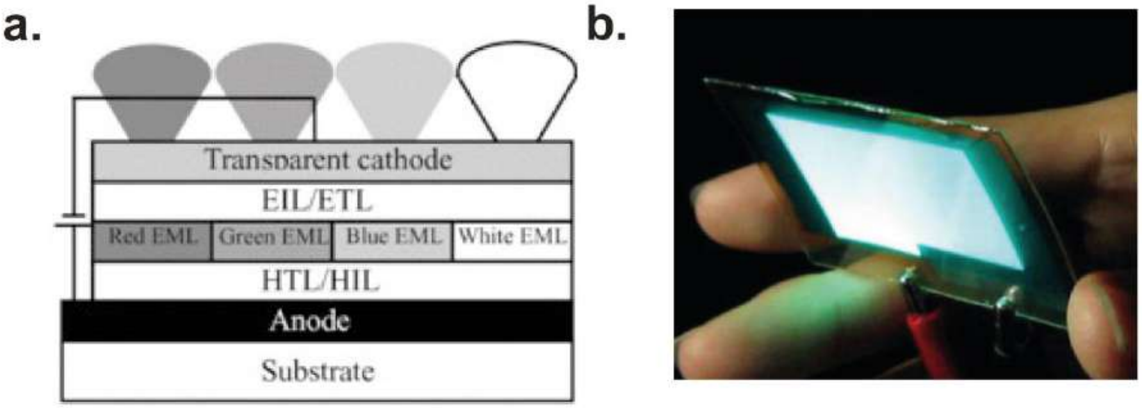

**Figure 2**. **a.** Architecture of OLED: hole injection layer (HIL), hole transporting layer (HTL), electron blocking layer (EBL), emitting layer (EML), hole blocking layer (HBL), electron transporting layer (ETL), and electron injection layer (EIL) (Copyright 2010, Wiley), (Chen *et al*., 2010); **b.** Flexible OLED display supported by a transparent cellulose nanofibrils composite film (Copyright 2008, Wiley) (Nogi and Yano, 2008).
**Figura 2. a.** Arquitectura de una OLED: capa de inyección de agujeros (HIL), capa de transporte de agujeros (HTL), capa de bloqueo de electrones (EBL), capa emisora (EML), capa de bloqueo de agujeros (HBL), capa de transporte de electrones (ETL), y capa de inyección de electrones (EIL) (Copyright 2010, Wiley) (Chen *et al.*, 2010); **b.** Pantalla OLED flexible soportada por una película compuesta de nanofibrillas de celulosa transparente (Copyright 2008, Wiley) (Nogi and Yano, 2008).

## Transistors

Meanwhile, CNF nanopapers have also been explored as substrates for fabricating flexible transistors (Huang *et al*., 2013; Jung *et al*., 2015). These devices can perform logic operations and are the fundamental building blocks for the fabrication of complex circuits in computer processors. Field-effect transistors are the most common technology and are composed of three main components: the source, the drain, and the gate. The electrical current flowing between the source and the drain is controlled by the gate through the application of a voltage (Fig. 3a). The combination of various transistors allows for the fabrication of logical gates that can perform assorted functions (such as AND, OR, NOT).

Thus, the arrangement of transistors in specific configurations allows to perform logical operations and consequently enables the fabrication of processors capable of executing complex tasks. CNF nanopapers have already been explored for the fabrication of various transistor technologies, including thin film transistors, organic thin film transistors, and organic field-effect transistors. The devices could operate when being bent to various angles with only minute alterations in performance (10 % loss in carrier mobility) (Huang *et al*., 2013).

Besides applications as substrates for flexible transistors, CNF nanopapers were recently explored as a functional component in the design of an organic field-effect transistor. Specifically, a CNF nanopaper was evaluated as a replacement for the traditional dielectric materials (typically composed of metal oxides that require complex manufacturing processes and high processing temperatures). This was achieved by transforming a CNF nanopaper into a solid-state ionic conductor through a simple chemical process (Fig. 3b) (Dai *et al*., 2018). As the gate voltage was applied, an electric field was generated by the conducting CNF nanopaper that successfully modulated the current flow between the source and drain. The CNF-based transistor performed a similar function to that of organic field effect transistors integrating dielectric materials. Thus, the CNF structural advantages such as high transparency, temperature resistance, flexibility, as well as the possibility of being converted into solid-state ion conductors have triggered much excitement toward the fabrication of flexible and transparent transistors, and potentially environmentally friendly processors.

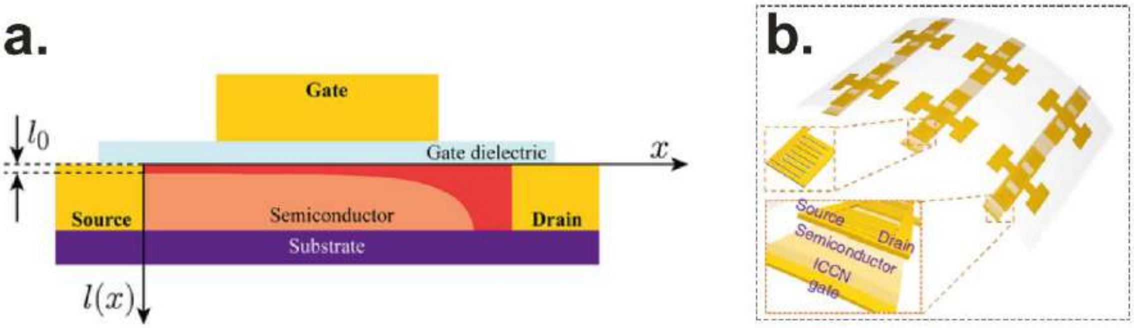

**Figure 3. a.** Architecture of a field effect transistor (Copyright 2023, Wiley) (Luginieski *et al*., 2023); **b.** Organic field effect transistor wherein CNF is used as a replacement to the dielectric layer (ionic conductive cellulose nanopapers, ICCN) (unrestricted reuse from Springer Nature) (Dai *et al*., 2018)
**Figura 3. a.** Arquitectura de un transistor de efecto de campo (Copyright 2023, Wiley) (Luginieski *et al*., 2023); **b.** Transistor de efecto de campo orgánico en el que se utiliza CNF como sustituto de la capa dieléctrica (nanopapeles de celulosa conductores iónicos, ICCN) (reutilización sin restricciones de Springer Nature) (Dai *et al*., 2018).

## Energy storage devices

Perhaps one of the most significant areas where CNF has demonstrated considerable potential is energy storage. The dominant energy storage technologies where CNF has been explored are supercapacitors and batteries. Supercapacitors are generally composed of porous electrodes made of carbon (nano)materials immersed in an electrolyte solution. As a potential is applied to the electrodes, the ions in the electrolyte solution migrate until they reach the surface of the electrodes. Thus, the application of potential causes electrostatic separation of charges between the electrodes and the electrolyte allowing to store and release energy (Fig. 4a).

In contrast, batteries such as lithium-ion consist largely of four key components: cathode, anode, electrolyte, and separator. During charging and discharging cycles, lithium ions move between the cathode and anode materials, converting the chemical energy into electrical energy. The electrolyte acts as the ionic conductor, allowing for the transport of lithium ions between the electrodes. The separator serves as a barrier preventing physical contact between the anode and cathode while facilitating ion transports in the battery cell. Each of these four components plays an important role in determining the final battery performance (Fig. 4b).

While both supercapacitors and batteries can store energy, the differences in energy storage mechanisms yield distinct properties suitable for different applications. For instance, supercapacitors possess a high-power density, translating into the devices being able to deliver high power within a short amount of time. Meanwhile, batteries possess a high-energy density, which means that they can store more energy as compared to supercapacitors, and thus are suitable for powering electronic devices over prolonged periods.

Most notably, CNFs are ideal for the design of flexible electrodes and separators. CNF's high mechanical strength and thermal stability allow the integration of various carbon nanomaterials such as carbon nanotubes, activated carbon, and graphene to obtain flexible composite electrodes for supercapacitors (Chen *et al*., 2018). Interestingly, the presence of small amounts of CNF also enhanced the amount of energy stored by the supercapacitors (Fig. 4c) (Zhang *et al*., 2022). The researchers suggested that the integration of small amounts of CNF improved the porosity of the electrode, which facilitated the migration of ions from the electrolyte to the carbon nanomaterials.

Similarly, beneficial properties were observed when using a CNF polyethylene nanopaper composite as a separator in comparison to a commercial polyethylene. An ideal separator in a battery should be thin, mechanically strong, and electrochemically stable. Another important characteristic is the presence of a highly porous and tortuous structure that prevents the growth of dendritic lithium. Dendritic lithium can cause short circuits in batteries, potentially causing explosions. Researchers observed that the integration of CNF yielded a lithium-ion battery with a prolonged lifetime because of the presence of uniform pores throughout the cellulose nanopapers (Fig. 4d).

CNFs have also attracted a broad interest as solid-state electrolytes. Generally, lithium-ion batteries use organic liquid electrolytes to enable the movement of ions between the electrodes. However, these electrolytes are volatile and flammable, which has led to catastrophic failures including uncontrolled exothermic reactions and explosions. Widely documented examples of such events include Samsung Galaxy Note, Tesla electric cars, and grid stations in South Korea and the USA.

Alternatively, solid polymer electrolytes have emerged as a safer option owing to their capability to dissolve and dissociate lithium-based electrolytes, inexpensiveness, and facile processibility. Nevertheless, conventional solid polymer electrolytes exhibit generally low ionic conductivities that limit the performance of solid-state batteries (for instance, a lower amount of energy stored). In a recent study, CNF nanopapers were subjected to various chemical treatments and transformed into an ionically conducting material. Remarkably, the modified nanopaper outperformed conventional solid polymer electrolytes with an ionic conductivity 10-fold superior to that of the best systems previously reported (Yang *et al*., 2021). This improvement allowed to boost the amount of energy stored by the solid-state battery and was explained by the nanofibrils acting as molecular channels, or cables, that enabled a rapid transport of lithium-ion throughout the electrolyte.

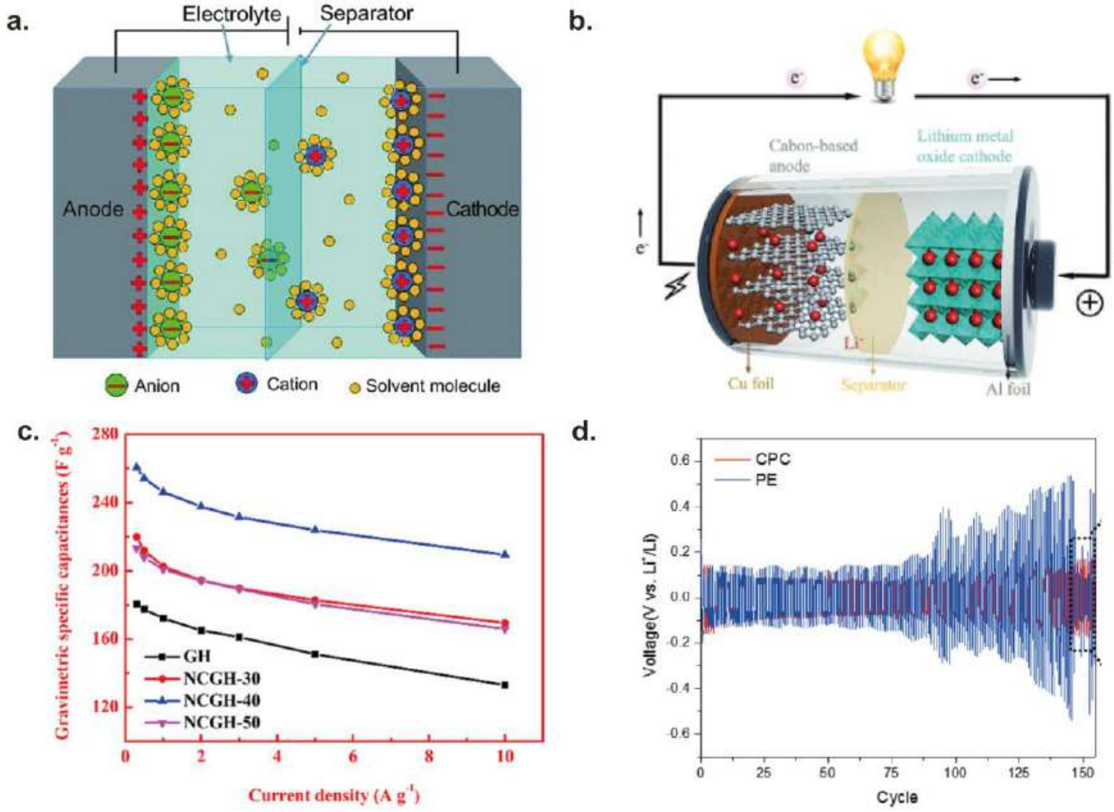

**Figure 4. a.** Energy storage mechanism in conventional supercapacitors (Copyright 2022, Wiley) (Ali *et al*., 2022); **b.** Energy storage mechanism in conventional lithium-ion batteries (Copyright 2020, Wiley) (Zhang *et al*., 2020); **c.** Capacitance comparison of reduced graphene oxide (GH) and nanocellulose/reduced graphene oxide composite hydrogels (NCGHs) with various loading contents of cellulose nanofibrils (Copyright 2022, American Chemical Society) (Zhang *et al*., 2022); **d.** Performance comparison of lithium-ion batteries consisting of commercial separator (PE) and CNF polyethylene composite (CPC) (Copyright 2018, Wiley) (Pan *et al*., 2018). **Figura 4. a.** Mecanismo del almacenamiento de energía en supercondensadores convencionales (Copyright 2022, Wiley) (Ali *et al*., 2022); **b.** Mecanismo del almacenamiento de energía en baterías de iones de litio convencionales (Copyright 2020, Wiley) (Zhang *et al*., 2020); **c.** Comparación de la capacitancia de hidrogeles compuestos de óxido de grafeno reducido (GH) y nanocelulosa/óxido de grafeno reducido (NCGH) con diversos contenidos de carga de nanofibrillas de celulosa (Copyright 2022, American Chemical Society) (Zhang *et al*., 2022); **d.** Comparación del rendimiento de baterías de iones de litio formadas por un separador comercial (PE) y un compuesto de polietileno CNF (CPC) (Copyright 2018, Wiley) (Pan *et al*., 2018).

## 3. Conclusions and perspectives

Despite an impressive array of potential applications, the development of CNF-based flexible electronic devices is still in its early stages. There are major challenges that need to be addressed before these materials can be widely adopted. CNF is currently relatively expensive to produce, which is a significant barrier to its widespread adoption. The scalability of production is another critical issue; as current production methods are not equipped to meet the demands of large-scale production. Additionally, the stability of CNF is a concern, as its properties such as electrical resistance and mechanical strength are altered by the humidity in the surrounding environment.

Despite these hurdles, the potential of CNF-based flexible materials is enormous. With continued research and development, CNF has the potential to revolutionize the electronics industry. This innovative material offers a myriad of possibilities for the future of flexible devices and continues to be a thriving area of research. As sustainability and environmentally friendly solutions gain prominence, CNF's role in flexible electronics is expected to grow, driving innovation, and shaping the future of electronic devices. Despite these hurdles, the potential of CNF-based flexible materials is enormous. With continued research and development, CNF has the potential to revolutionize the electronics industry. This innovative material offers a myriad of possibilities for the future of flexible devices and continues to be a thriving area of research. As sustainability and environmentally friendly solutions gain prominence, CNF's role in flexible electronics is expected to grow, driving innovation, and shaping the future of electronic devices.

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## Conflict of interest

The authors declare no conflict of interest.

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