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REVIEW

Cellulose based smart sensors

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Abstract: Cellulose fibres, cellulose nanofibers, cellulose nanocrystals and cellulose derivatives are all examples of cellulose-based materials that display superior characteristics with a number of desirable properties, including biodegradability, sustainability, biocompatibility, thermal properties , optical transparency, flexibility, high mechanical strength, high porosity, hydrophilicity, a large surface area and broad chemical modification capabilities. "Smart" materials based on cellulose created by the chemical changes and physical incorporation/blending techniques offer numerous advantages, most notably their intelligent responses to environmental stimuli. Conductive networks are formed in cellulose-based composite materials by combining or coating conductive materials with the cellulose components or by directly carbonising the cellulose materials. Numerous nanopaper-based optical sensing platforms are explained and how they can be tailored to exhibit plasmonic or photoluminescent features suitable for sensing applications using nanomaterials or as biomaterials. The responsiveness of these "smart" materials to pH, temperature, light, electricity, magnetic fields and mechanical forces, among other parameters, is also reviewed, as were their applications as drug delivery systems, hydrogels, electronic active papers, sensors, shape memory materials, smart membranes, *etc*.

Keywords: cellulose, sensors, multi-faceted applications, piezoelectricity, smart materials

1 Introduction

Functional materials made from renewable resources have attracted significant interests, owing to easy accessibility, sustainability, environmental friendliness, non-toxicity and renewability [1, 2]. Cellulose is the most prevalent naturally occurring biodegradable polymer, which is currently under-utilised for various applications Cellulose is produced by plants and microbes through biosynthesis [3, 4]. Nanocellulose refers to small fibers with nanodimensions, which is obtained from cellulose through multiple pathways, the most common being alkali or acid hydrolysis. Nanocellulose has emerged as a valuable green material for myriad of applications, varying from reinforcement materials to sensing platforms that are extremely valuable for food, pharmaceutical and environmental protection industries [4–6]. In addition, renewable nanocellulose obtained from biomass showed good mechanical properties, low density, anisotropic structure, optical properties, good mechanical properties, chemical resistance, tailorable surface chemistry and high biocompatibility [7].

Interesting properties of nanocellulosic materials include enhanced water absorption and retainability, low water solubility, biodegradability, increased surface area to volume ratio, and biocompatibility. Cellulosic nanofibrils in the form of nanocellulose can be recovered from lignocellulosic bioresources by a variety of chemical, enzymatic or mechanical methods. Mechanical breakdown of plant biomass [8,9] may result in the formation of cellulose nanofibrils from these cellulose nanofibres (CNFs). CNF is composed of high aspect ratio nanofibrils with a diameter of less than 100 nm. Cellulosic nanofibrils provide the plant with toughness, strength and stiffness [10, 11]. CNFs may be processed into tough solids such as fibrillar aerogels [12, 13], cellular foam [14] and membranes [15, 16] or utilised as reinforcing components in biocomposites due to their high modulus (100 GPa) and network-forming capabilities. CNFs are used in a variety of applications, including rheological fluids, nanopaper reinforcement fillers, and medical membranes [17].

Cellulose nanopaper offer a better mechanical strength, lower coefficient of thermal expansion (CTE) and a higher optical clarity [17–20]. Taniguchi et al. demonstrated the preparation of the first nanopaper from CNFs [21] and followed by casting or vacuum filtering by many groups [22–26]. Tensile strength and tensile rigidity of low-porosity nanopaper were reported to be 230 MPa and 13 GPa, respectively, and could be increased to a maximum of 400 MPa and 46 GPa by partial alignments of the nanofibrils [23]. The dense packing of nanofibrils with

interfibrillar H-bonds offer significant advantages, including improved oxygen barrier properties and resistance to water vapour [24, 26, 27].

2 Structure and properties

Cellulose is a strong, fibrous, water-insoluble material that is critical for preserving the integrity of plant cell walls. Continuous advances in our understanding of the assembly, biosynthesis and structural properties of cellulose have inspired a diverse range of research activities in a broad range of areas ranging from reinforcement materials and sensing platforms to food control, environmental protection and nanomedicine [7, 9, 16]. Cellulose nanofibers (CNF) exhibit interesting physical and chemical properties as functions of their morphology and crystalline structure. Typically, they are characterized with low density, high mechanical strength, chemical resistance, thermal stability, transparency, biocompatibility, and large surface area. The importance of this nanocellulose obtained from renewable resources lies on its fast degradation, low toxicity, and easy reusability. (see Figure 1)

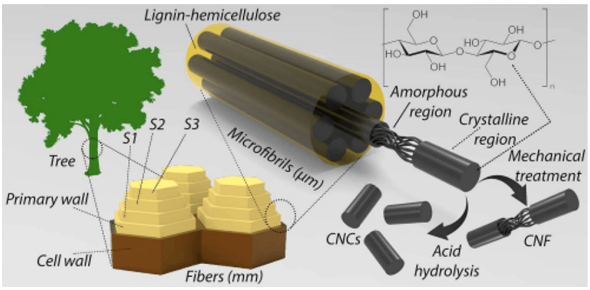

Figure 1 Structure of cellulose depicted hierarchically along with the amorphous and crystalline regions in cellulose fiber (From Dias et al., 2020)

3 Classification of nanocellulose

Nanocellulose is divided into three kinds based on their manufacturing technique, functionality and size. These include cellulose nanofibrils (CNF), cellulose nanocrystal (CNC) and bacterial nanocellulose (BNC). CNF is mostly harvested from plants and chemical techniques are employed to break the link between the tightly attached cellulose bundles into individual fibrils. CNF's great flexibility is a result of its fibrillary structure, which consists of crystalline sections connected by amorphous regions. This structure makes CNF ideal for film production. Crystalline nanocellulose (CNC) comprises rigid rod-like crystallites as compared to CNF and BNC. Acid hydrolysis is utilised to separate the highly crystalline fibre segments from the amorphous portions of CNF. BNC is cellulose fibrils are generated by Gluconacetobacter xylinus species under optimal growth conditions. The bacteria produce BNC as a primary metabolic product with a gel-like consistency [28]. BNC outperforms cellulose obtained from plants in terms of mechanical and thermal stability, tensile strength, purity, and biocompatibility due to its high degree of crystallinity and degree of polymerization. (see Figure 2)

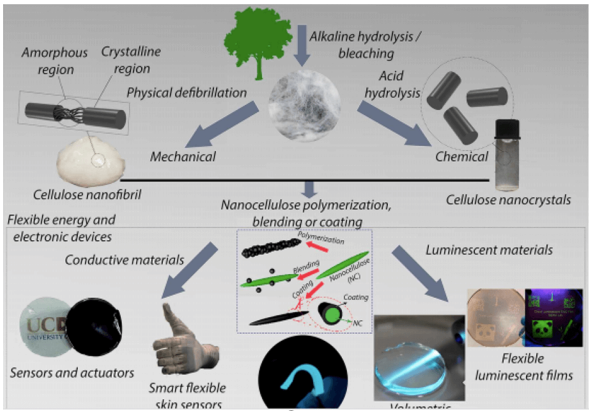

Figure 2 Extraction of CNF and CNC from cellulose, surface functionalization of nanocellulose for electronic materials and flexible energy applications (From Dias et al., 2020)

3.1 Cellulose nanofibrils (CNFs)

Cellulose nanofibril (CNF), also known as nanofibrilloxy nanofibrillate [29] or nanofibrillaire cellulose [30] are the smallest structural unit of a plant fibre, consisting of a bundle of long, entangled and flexible nanofibers stretched cellulose chain molecules ranging in length from 1 nm to 100 nm. It is composed of both crystalline and amorphous domains/regions. Cellulose nanofibers (CNFs) can be isolated/ purified from cellulose fibers of lignocellulosic resources such as wood and forest or agricultural residues by using various chemical, mechanical or chemo-mechanical disintegration processes such as homogenizing, grinding, acid hydrolysis, enzymatic-assisted hydrolysis, 2,2,6,6-tetramethylpiperidine-1-oxyl-mediated or TEMPO-mediated oxidation, solvent-based isolation, electro-spinning and high-intensity ultrasonication [31].

3.2 Cellulose nanocrystals (CNCs)

In comparison to CNFs, cellulose nanocrystals' are rod-like elongated, crystalline forms, which exhibit limited flexibility due to their non-amorphous nature [32]. The amorphous parts of nanofibrillated cellulose are hydrolyzed and removed by acid while the crystalline parts are still maintained. Employing acid hydrolysis, CNCs are generally extracted from cellulose fibre and are also referred to as nanowhiskers, rod-like cellulose crystals [33] and nanorods [34]. The aspect ratio is quite modest, with an average diameter of 2 - 20 nm [35]. The length varies from 100 nanometres to several micrometres. The particles are made entirely of cellulose and have a crystalline concentration ranging from 54 to 88 percent [36]. The degree of crystallinity, the variety of dimensions and the shape of the cellulose are all impacted by its source, preparation conditions and experimental procedure.

3.3 Bacterial nanocellulose (BNC)

BNC is generated by bacteria belonging to the gluconoacetobacter family (*e.g.*, acetobacter xylinium) [37]. These bacteria have the ability to convert glucose to cellulose. BNC distinguishes itself from the other two kinds of nanocellulose as it is formed in the biological processes. BNC nanofibrils have a natural crystalline cellulose I structure with extended hydrogen-bonded polymer chains and high molar mass [28]. The natural nanofibril's Young Modulus is expected to be 150 GPas [38, 39], which is almost equivalent to the value of steel (w200 GPa).

4 Synthesis

In nature, cellulose nanofibrils are generated in an ascending (or bottom-up) manner, with the primary component being the cellulose polymer, which is formed during photosynthesis in green plants and microorganisms. When coupled with other substances, the cellulose chains self-assemble into elementary nanocrystals and nanofibers with multiple strong hydrogen bonds between them, which then lead to robust macrofibrillar structures. Alternatively, scientists have developed ways for physically and chemically removing these nanofibers from natural cellulosic materials (*e.g.* wood, plants and algae), *i.e.*, also known as top-down or descending approach. These chemical and physical methods allow for the fragmentation of fibres through their amorphous zones, resulting in CNC with variable shapes, sizes, and crystallinities according to the approach used. Microcrystalline cellulose (MCC) fibers are the fundamental/intrinsic component of wood. CNFs may be recovered from MCC when the amorphous domains of the fibrils are shattered. CNF can be isolated under mild circumstances by a number of physical processes (*e.g.*, grinding, aqueous counter collision and milling) and oxidation reactions that disrupt the hydrogen bonds and interfibrillary van der Waals forces. CNC is frequently extracted by acid hydrolysis, using hydrochloric acid, sulfuric acids or enzymatic procedures followed by rigorous ultrasonic or mechanical treatments that successfully disintegrate and remove the amorphous areas, releasing just the rod-like crystalline particles [40].

5 Applications in various fields

Nanofibrillar cellulose is a versatile material that may be tailored to particular applications in technology and medicine. As a result, nanocellulose is aligned, processed or functionalized to form a diverse range of materials for a number of applications, including three-dimensional gel-like materials, films, solid powders, suspensions, fibres and surface coatings.

Recently, Nanocellulose has been used to create polymeric materials that exhibit desired characteristics such as stimuli responsiveness, flexibility, barrier properties and mechanical stiffness. In industry, the unique capabilities of nanocellulose incorporated materials were used

for the detection, extraction and photodegradation of contaminants, extraction of oil, industrial catalysts, reinforcing agents and industrial by-products [41]. Nanocellulose has demonstrated great promise in the fabrication of energy storage devices [42]. Nanocellulose is also commonly used in the cosmetic and food industries as a gas barrier, emulsifier and carrier. Apart from this, nanocellulose paves the stage for more sustainable alternatives to fossil-fuel related materials and for developing future biomedical sensors. A widely used bio-healing nanomaterial with superior wound healing and antimicrobial properties for use in artificial organs, skin replacement, blood vessels and implants, transport of medicines, cells and cosmetics [43–48].

Cellulosic nanoparticles have recently extended their analytical applications, potentially functioning as analytical instruments for detecting, collecting, and extracting contaminants [49–51]. Sulfonated NC (s-NC) powders have demonstrated remarkable absorption capacity for metal nanoparticles with antibacterial characteristics in a range of solid phase extraction modalities [52, 53]. Additionally, the capacity of NC to pack densely has been investigated for different reasons. Recently, it was proven that paper-like and gel-like nanocellulosic forms of NC may be used as substrates to adsorb significant amounts of liquid and gas pollutants [54, 55].

5.1 Detection of various environmental conditions

5.1.1 Self-healing sensors

After damage, self-healing sensors restore the structure, integrity and characteristics of the network, thereby increasing the service life of the sensor device [56, 57]. The development of dynamically reversible chemical bonds, such as hydrogen bonds, hosts and hydrophobic contacts, results in the formation of a self-healing hydrogel [58, 59].

A self-healing, highly conductive, stretchy hydrogel was successfully developed for application in flexible strain sensors. CNFs and graphene have been combined to form graphene-CNFs composite, which ensure that graphene nanocomposites (GN) is distributed uniformly throughout the Polyvinyl alcohol(PVA) network. The strength, electroconductivity and elasticity of graphene-CNF@PVA hydrogels were significantly enhanced by the incorporation of graphene-CNF composite. The hydrogen bonding and interconnection of PVA to graphene-CNF, as well as the reversible multicomplexation of the borax-cross-dynamic connection, were used to produce the composite network hydrogel. When combined with graphene-CNF@PVA hydrogel, pressure sensors can detect and monitor body movement in real time, indicating possible usage in wearable sensing equipment [60].

It is possible to stretch the self-healing hydrogels without causing harm to the interface surface. Moldable “NFU” hydrogels retained their amazing autohealing capacity when connected and lifted. The dynamic and reversible hydrogen bonding between GN-OH CNF-Group with borax and PVA is responsible for the self-healing properties of GN- CNF@PVA hydrogels. These hydrogen bonds are rapidly broken and rebuilt, allowing the hydrogel to be reformed and show self-healing properties at room temperature [61, 62]. The GN-CNF@PVA hydrogel-based strain sensor exhibited exceptional electrical stability and reproducibility when used to detect human movement. As a consequence, the hydrogel-based pressure sensor was able to recognise not only simple human gestures but also complex spatial motions, which speaks well for the future development of communication for the deaf and sign language recognition [60].

5.1.2 Strain sensors and piezoelectricity

Strain sensors are used in a variety of applications, including strain measurement, detecting damage in real time and preventing catastrophic collapse. Strain sensors or transducers are categorised according to their functioning mechanism as optical sensors, piezoelectric sensors or resistors. Recent research has been concentrated on developing flexible, low-cost, highly sensitive strain sensors. This has resulted in the creation of a variety of nanomaterial-based strain sensors, including bucky sheets, carbon nanotubes, graphene and zinc oxide nanowires, but the processing and material costs remain expensive.

Due to the piezoresistivity of the conductive substances used, the paper based sensors are strain sensitive. Mechanical deformation of the material network results in a proportionate change in resistance to the strain on the paper substrate. Strain sensing was achieved under static and dynamic loads using a paper-zinc oxide nanostructure covering on the paper substrate. The output current of ZnO coated paper attached to a brass beam was tested to determine its sensing capability in response to changes in strain. Paper strain sensors are well suited for detecting low-frequency signals or static forces of 12 kHz as their natural resonance frequency of 25 Hz is far lower than that of commercial silicone sensors. On the other hand, paper force sensors have a number of cost and manufacturing advantages. Khajeh et al. developed resistive strain gauges based on printing conductive lines of carbon black (CB) on paper using an inkjet. The

long fibres of NBSK paper facilitates the transfer of CB particle suspensions into the paper's large pore size.

Microelectromechanical resistive system (MEMS) sensors were built using a standard office printer and CB was printed and connected to the conductors using commercial silver ink lines of 1/sq. After cutting and folding the paper substrate, a three-dimensional structure containing an inertial sensor and a touch sensor was produced.

Lin et al. demonstrated how to create resistive stress gauges by employing a pencil-on-paper technique. A few leading tracks are simply sketched on a paper resulting in graphic deposits that stick to the paper matrix. Lin et al. constructed paper strain gauges by forming U-shaped solid rectangles on a sheet of office paper. The paper was fractured to create a resistor-equipped beam structure, and its resistance to deflection or bending was altered by compressing/stretching the graphite network in parallel sections of the U-shaped trace. Due to the high concentration of conductive carbon particles in the element created with the HB pencil, the sensor exhibited excellent repeatability and reusability.

By integrating face solution and vacuum filtering technology for building flexible sensors, an electrically conductive hysteropaper hybrid Ag nanowire (AgNW)/cellulose (CNF) sensor system was created. (not clear!) Due to the amphiphilic nature of cellulose, homogenous dispersion of AgNW and the creation of highly efficient electrically conductive networks was fabricated. To evaluate their potential for sensing strain, two different strain sensors were constructed. Between two thermoplastic polyurethane(TPU) sheets, a hot hybrid nanopaper with a unique microfracture structure was generated by pre-training a tensile stress sensor. The varied fracture densities produced by various pre-strains suggested that strain sensing behaviour is pre-strain dependent [63].

Using a vacuum filtering method, hybrids of thermally conductive nanofibrillated cellulose (NFC) and nanodiamond (ND) were created. Cellulose/graphene nanocomposites was also synthesised to study their strain sensing capabilities. Low detection limit and high sensitivity are two critical requirements for optimising conductive polymer composite (CPC)-based strain sensors (*e.g.*, swallowing, heartbeat, pulse) for measuring human physiological signals. To accomplish this, innovative geometrical features, few of them being, microarrays, microcrack structures [64–67] and gaps have been studied, for integrating with CNCs. Microcrack development within CPCs, in particular, has been shown to be the most successful technique. Microcrack structures can be used to enhance the sensing characteristics of cellulose-based CPCs [68].

Facial mixing and vacuum filtering were used to make the flexible, conductive nanopaper Ag Nanowire (AgNW) and CNF (cellulose nanofibre) hybrid. The form, mechanical and structural characteristics of the hybrid nanopaper were carefully investigated. Using the developed hybrid nanopaper, two separate strain sensors (tensile and bending) were fabricated to measure environmental temperatures. The developed AgNW/CNF hybrid nanopaper displayed remarkable multiple sensing capabilities as a result of the change in the AgNW conductive networks in response to external stimuli.

Cellulose nanoparticles have also been demonstrated to have piezoelectric and electroactive characteristics, suggesting that they might be utilised as actuators and sensors in a range of devices such as microelectromechanical systems, robots, speakers and micropumps. Csoka et al. discovered a piezoelectric effect in thin CNC films with an effective shear piezoelectric constant of 2.1/V, which is equal to the effective shear piezoelectric constant of a reference ZnO film. Chemical alteration, can enhance electrically induced bending deformation of bacterial cellulose sheets [62–67].

Additionally, the researchers examined the vibration detecting properties of cellulose EAPap (electro-active paper). The sensor was constructed using piezoelectric cellulose plated plated on both sides with metallic electrodes and covered with thin laminating plates. It was tested by attaching the cellulose EAPap to the surface of an aluminium cantilever beam and measuring its response to beam vibrations. The use of a cellulose EAPap consisting of interdigitated electric electrodes as an acoustic wave sensor for isopropyl alcohol detection has been described, with piezoelectric cellulose acting as both the sensing layer and substratum. The sensor's acoustic wave travels at a speed of 2400 metres per second and has a resonance frequency of 58.2 millimetres per second, which is equivalent to that of other piezoelectric materials [75].

The energy harvesting and sensing capabilities of ZnO-strain paper was shown by synthesising piezoelectric ZnO nanostructures on paper using a wet chemical method [76]. Vertically aligned ZnO nanorods were synthesised on a metal-coated paper substrate and their capacity to function as a flexible piezoelectric nanogenerator was proven [77]. Furthermore, a wet chemical technique was utilized to demonstrate the capacity of ZnO nanorods to absorb energy directly on cleanroom paper [78].

Besides, a low-cost, ecologically friendly hybrid paper suitable for use as a piezoelectric

substrate was created for sensor applications. The manufacturing procedure is straightforward and does not need the use of complex technology. Barium titanium ($BaTiO_3$) nanostructured fibres have characteristics similar to those of wood cellulose fibres. This leads in the formation of a positively charged surface on the wood fibres via the layer technique. When treated wood fibres are submerged in a $BaTiO_3$solution, the negatively charged $BaTiO_3$ bonds to the cellulose fibres electrostatically. The piezoelectric characteristics of the paper were determined using a $BaTiO_3$ concentration and then enhanced by increasing the $BaTiO_3$ loading [76].

Additionally, scientists changed the manufacturing process of piezo-electric paper to incorporate functional wood fibre preactivation and chemical reinforcing As a consequence, a piezoelectric paper with the strength of printing paper was created. The increased piezoelectric coefficient of the CMC(carboxymethyl cellulose)-added piezoelectric paper is largely due to decreased stress absorption in the wood fibre network and better fiber-fiber bonding in the piezoelectric paper [77].

5.1.3 Temperature dependence

Tensile and bending strain hybrid nanopaper displays reproducible and consistent negative temperature sensing behaviour when used as a temperature sensor. Between 290 and 370 K, the variance in relative resistance decreased linearly with increasing temperature, with a typically negative temperature coefficient (NTC). Soon after, it increased again during cooling, nearly paralleling the heating curve. The free electrons in AgNW get activated largely during the heating phase, contributing to the NTC sensing characteristics and resulting in resistance decreases.

5.1.4 Change in magnetic field

Magnetostriction is a phenomenon in ferromagnetic materials that causes physical geometries to alter in response to an external magnetic field. Magnetostrictive effects are frequently negligible in ordinary materials like nickel. The discovery of rare earth iron alloys with significant magnetostrictive effects, such as Terfenol-D, has aided in the development of sensors and energy harvesting systems that need higher energy density, faster reaction time and more precision. The magnetostrictive effect has also been used in transducers for sonar applications [81, 81–83], energy harvesting applications [81, 84–86], active vibration control applications [87–89], and position control system applications [90, 91]. Monolithic Terfenol-D materials are heavy and difficult to shape into appropriate geometries for sensor and actuator production; they remain extremely brittle and can withstand only small loads before breaking [91, 92]. One proposed approach is to leverage the better mechanical properties of the nanocellulose matrix to build a magnetostrictive composite with magnetostrictive particles. The structure of cellulose nanofibers creates a layered substrate for magnetostrictive materials. Since the resulting composite has the form of a film and contains magnetostrictive particles, it is known as a magnetostrictive nanocellulose membrane (MNM) [93].

The Joule effect is a term that refers to changes in the length of a material caused by a change in its magnetostrictive state. As a result, the magnetic field is capable of morphing or activating the flexible MNM. The magnetostrictive opposite effect, or the Villari effect, modifies a material's magnetic susceptibility. Since the film's magnetic state is anticipated to change in response to changes in external mechanical stresses, these variations in magnetic state can create voltage in coil(s) in accordance with Faraday's law, which can then be used to power an electrical load. Additionally, a magnetic field may be used to enhance the magnetostrictive properties of a composite by favourably aligning particles during the manufacturing phase [94–98]. A magnetic field and the Joule action activate the unimorph nanocellulose composite.

The fabrication process for the composite material was generally similar to that for producing CNF described previously [87], with the addition of techniques for adding Terfenol-D particles. A DC stream image was created for each increase in the magnetic field using an optical microscope- Stemi 2000-C, Carl Zeiss AG, connected to a digital camera. After converting the picture sequence to a video file, the sample's angular movement as a function of magnetic field intensity was calculated using an open source video analysis programme, or a tracker [93].

5.1.5 Due to deformation

The sensor concept is based on a nanocomposite of BC that has been injected with ionic liquids(ILs) and covered with a conductive polymer. The structure is cantilevered and the flexural behaviour of the structure is being explored. In reaction to the external mechanical source, the beam deforms and produces a voltage level proportional to the measurand. The frequency behaviour of the system, as well as its step response, have been examined. It is worth noting that the proposed solution is the first demonstration of this class of compounds' sensing

capabilities, which might be used to develop flexible, eco-friendly, biodegradable and low-cost sensors [99].

5.1.6 Fluidic and biochemical sample environment

Paper-based microfluidic analytical devices (PADs) are novel platforms having the capability of handling fluids and analysing biochemical samples on a single-use basis. Microfluidics are preferable to soft polymers for incorporation into paper-based devices due to their volume consumption, low cost, ease of production and portability [100]. Capillary fluid flow is enabled by the hydrophilic properties of paper-based substrates [101]. This is due to the fact that the aggregation of micro/nanometer-sized cellulose fibres acts as a porous material. As a result, the flow behaviour of a fluid on paper is determined by the flow direction (vertical or lateral), composition of the paper (nanopaper, filter, nitrocellulose or chromatographic) and the size and geometry of the prescribed fluidic channel. Filter paper can be used not only for transporting and loading samples, but also for path connection and as an absorbent pad [102]. Antibodies and bioconjugated nanoparticles can be transported, loaded, and immobilised using nitrocellulose paper [103]. Furthermore, nanocellullose paper, a type of paper produced by Acetobacter Xylinum bacteria, has a nanoscale structure of fibres and holes ranging in size from 10 to 100 nm [104]. This nanoscale structure was helpful for embedding MNP in nanopaper in order to incorporate a nanoplasmonic substrate instead of colloidal suspensions [105].

A microfluidic based biosensor fabricated by one-step plotting method was used in forming permanent ink marks on paper substrate. The microfluidic channel fabrication approach was through forming permanent ink marks on paper substrate with the pattern agent being a commercial permanent marker. It was detected through calorimetry. The advantages of this approach was that it did not require expensive equipment or specialized skills [106]. Another approach was through wax printing by printing solid wax patterns on paper substrate followed by heating. It was detected electrochemically, evident in examples such as blood and glucose [107]. Bench top fabrication method is another methodology involving the coating of PDMS onto the assembly of DSA and paper [88].

5.1.7 Dielectric environment – plasmonics

These biomaterials (oligonucleotides and antibodies) or biomimetic materials (molecularly printed polymers) are typically used as biorecognition elements, resulting in reaction zones that probe for the presence of an analyte via highly specific and selective interactions with the previously labelled analyser via plasmonic nanotechnology [109]. These reaction zones serve as concentration zones for biorecognition events labelled with plasmonic nanoparticles, resulting in a visually observed phenomenon that indicates the presence and absence of the target molecule due to the flux control provided by microfluidics on paper and the highly specific interactions enabled by the use of biorecognition samples. Gold nanoparticles, used in pregnancy test are commonly used to report this technique in lateral flow immunoassays [90].

Food deterioration may be monitored using paper-based devices integrating MNPs; this is because food spoiling results in the production of ammonia and other volatile organic chemicals capable of etching AgNPs [111]. Interestingly, this process causes the plasmonic device's hue to shift from amber to grey. Similarly, UV light can promote photodegradation of AgNPs, resulting in a reduction in the size of AgNPs embedded in nanocellulose. This exquisite feature enables the monitoring of acceptable levels of sun/UV exposure via a colour shift in a wearable paper-based device [96].

Analyte composite interactions that alter the shape, size, interparticle distance, refractive index or composition of the NPs make them ideal sensing platforms for plasmonic nanopaper composites. It was anticipated that optical sensing would be accomplished by the assembly or grafting of implanted metal nanoparticles. The potential for optical sensing of functionalized plasmonic nanopaper was assessed using AgNPBC as a colorimetric sensor for iodide or methimazole and AuNPBC as a colorimetric sensor for cyanide or thiourea[98].

5.1.8 Gas environment

Volatile organic compounds (VOCs) are found in a broad variety of industrial processes and consumer items, including food and beverage products. VOCs at certain concentrations can be a severe health and safety hazard; it is therefore important that VOCs are identified and recognised. Gas chromatography mass spectroscopy(GC-MS) is a reliable technique for examining a wide variety of gases and volatile organic compounds (VOCs). However, the device's data processing requires the expertise of skilled professionals. Moreover, a variety of other VOC detection techniques have been developed based on changes in gravimetrical, optical or electrical indicators [113], but these systems have limitations in terms of shelf life, reproducibility and so on [114].

This led to recent research being focused on developing chemical sensors using a paper substratum. The absorption of VOC gas molecules by a VOC-sensitive chemical in paper results in a change in electrical resistance, which allows for the detection of VOCs. The feasibility of using glassy cellulose paper as a substratum to detect ethanol at room temperature for flexible sensors was established. They developed sensors by stacking polydiallyldimethylammonium chloride (PDDA) sensing layer and ITO nanopowder on top of carbon nanotubes. In this application, PDDA works as a binder, improving the adherence of indium tin oxide (ITO) particles and carbon nanotubes to the paper substrate, enabling the combination to be dispensed using a deposition mechanism. The sensors and electrode material are placed onto the paper substrate using a proprietary microcontroller-based deposition technique. As the ethanol concentration rose, the sensor's current signal altered. The discharging and charging of the dual layer at the electrode-sensing material contact alters the sensor's electrical response [114].

Graphene on a copper sheet supported by HDPE were created using a CVD method [79]. The graph was printed on a conventional paper and the paper-based flexible nitrogen dioxide (NO_2) sensor reacted quickly when exposed to NO_2 gas, displaying a reaction time of less than 100 s.

Mechanical drawings of gas sensors were drawn on a paper substrate, with paper serving as the substrate sensing and gold, carbon nanotubes serving as electrode and sensing material respectively. By manually abrading a CNT pellet on a paper substrate, the gas sensor was constructed in two phases manually. Carbon nanotube pellets were initially produced by crushing them in a hydraulic press. The carbon nanotubes were then rubbed between the gold electrodes that had been inserted on the paper substrate. They constructed a variety of sensors from several types of paper such as cardboard, filter paper, weighing paper and evaluated their ammonia sensitivity (NH_3). These sensors exhibited a wide dynamic range and all examined sensors reacted linearly to NH_3 concentrations up to 0.510 ppm but nonlinearly to concentrations greater than 10 ppm. Its performance is equivalent to that of numerous other solution-phase carbon nanotubes. A further research [116] revealed that carbonate nanostructures with selectors are tiny molecules capable of chemically interacting with certain gases/VOCs, therefore enhancing the selectivity, reproducibility and reversibility of their sensors. Additionally, the suggested manufacturing process is cost-effective and solvent-free, enabling for quick prototyping of chemiresistive gas sensors on paper.

A low-cost technique called pen-on-paper (POP) for printing conductive devices on paper. POP is a novel way for dispensing conductive inks through a commercial roller ball pen. A colloidal dispersion of silver ink is provided for writing conductive structures. Silver nanoparticles can also produce silver ink when silver nitrate is reduced with diethanolamine and poly(acrylic acid). Electrical performance of POP-created conductive structures is exceptional with verified electrical resistance. POP was used to create complex shapes on high-frequency devices and paper.

A Polypyrrole paper gas sensor [117] was developed in two easy steps: first, ferric chloride (FeCl3) was manually painted on printing paper, after which the paper was subjected to pyrrole fumes, resulting in the fast formation of pyrene interfacial polymers as a result of the FeCl3 and pyrrole oxidation. The sensing capabilities of this PPy strip paper chip were evaluated using NH3. Doping PPy with poly(sodium-p-styrene sulfonate) significantly improved sensitivity.

Paper chemiresistors were also created by directly reproducing pencil markings on paper by a process called "pencil-on-paper". On printing paper, active sensing components and electrodes were created and the active layers were stabilised using a flexible toy pencil and an HB pencil, respectively.

When exposed to VOCs, the electrical resistance of these sensors altered instantly. When exposed to VOC vapour, the polymer matrix in the device's active sensing layer swells and pushes the graphite particles apart, changing the device's electrical resistance. Their sensor responded differently to each VOC tested, including methanol, acetone, THF, ethyl acetate, hexane and toluene also displayed outstanding selectivity and reversibility, however the limit of detection and detection range remain unknown.

The ability of PbS quantum dots (QDs) demonstrated to detect volatile organic chemicals by successfully incorporating them onto a paper substrate. The device is made in two simple steps: firstly, RF sputtering was employed a gold electrode is produced on a paper substrate and then QDs are integrated layer by layer using a $NaNO_2$ treatment. The device's electrical conductivity changed significantly after being exposed to NO_2, but there was no reaction to H_2S, NH_3, NO and SO_2. Additionally, the researchers studied and evaluated the VOC detection capabilities of devices constructed from a variety of substrates (PET, polymer, paper, Al_2O_3 and ceramic). While retaining a high NO_2 sensitivity, the paper-based device exhibited the quickest reaction of 12 s and recovery time of 37 s. Finally, paper was employed as a low-cost substrate for the construction of VOC sensors, either sandwiched between the paper and the electrode material

or directly into the paper electrode. Numerous low-cost and easy methods for embedding active sensing material on paper have been shown, including the use of a paper pencil, a paper pen and mechanical drawing. On paper, carbonate material demonstrated exceptional selectivity and reproducibility when coupled with a small molecule selector engineered to chemically react with certain gas analytes.

5.1.9 Presence of copper ions

To detect nano- to micromolar copper ions, a nanocellulose biosensor (CySense) based on the copper sensing properties of the CPC is used. CySense, a biosensor based on the detection of the red fluorescent proteins CPC and NFC, demonstrated unprecedented sensitivity to copper ions. CySense is based on the covalent attachment of CPC to nanofibrilliated cellulose oxidized by TEMPO, which acts as a sensing molecule conservator without the need for additional conservation agents. CySense films with reproducible morphological and optical characteristics may be created using a number of methods, including fluorescence and laser scanning. Additionally, the capability to cut laser-based composite films enables the use of NFC-based composite films in a number of sensor applications. CySense is a sensitive sensor that detects physiologically relevant amounts of free copper ions in human serum. This breakthrough provides the way for the development of nanoscale detection systems, robust point diagnostics, biocatalytic systems and bioactive filtration devices with surface patterns [118].

The results indicate the reliability and ease with which nanocellulose-based biosensors can detect changes in Cu^{2+} concentrations in human blood filtrations. Individual biosensor signal responses were assessed using spectrophotometric measurements for certain $CuCl_2$solutions and human blood spiked filtrate in order to precisely quantify the relevant free copper ion concentrations (*e.g.* Wilson's disease or Alzheimer's [104].

5.1.10 Nanoparticles in the environment

It was hypothesised that due to BC's optical transparency, optically active nanoparticles embedded in BC will function better than in alternative substrates frequently used in paper sensors, such as nitrocellulose. Due to the structure of BC, it acts as a preconcentration membrane, displaying virtually all of the enclosed nanoparticles' optical characteristics. This advancement may pave the way for the creation of valuable, novel nanopaper testing methods[112].

5.2 Photoluminescence

Quantum dots are semiconductor nanocrystals with strong excitation spectra, long-term photostability, tailored emission, symmetrical photoluminescence and high luminescence [82]. They frequently exhibit Stokes-shifted emission when exposed to UVs light (having a wavelength between 320 and 630 nm). Quantum dots have been used as optical sensing agents in a broad variety of applications, including labelling and imaging, for a long period of time [120]. After being modified with biomolecules or specific ligands, QDs can use photoluminescence to prove their interaction along with a specific analyte. This interaction is largely mediated by energy transfer between the donor,acceptor molecules and QDs [121, 122]. Due to its efficient photoluminescence, universal and long-lasting quenching properties, graphene-based materials are undergoing significant investigation for energy-based sensing applications [108, 109]. BC functions as a preconcentration membrane due to its structure, allowing its transparency to almost entirely disclose the optical characteristics of implanted nanoparticles. This discovery may pave the door for innovative and beneficial nanopaper-based testing methods. BC showed that the inclusion of trace quantities of optically active nanomaterials like QDs and AuNPs was beneficial in light of this preconcentration capability. The pathogen revealer GO was used to demonstrate the specificity and selectivity of the nanopaper composites as a biosensing platform, while the antibacterial QD-BC was utilised to demonstrate the selectivity and specificity of the proposed nanopaper composites as a biosensing platform [50].

5.3 Physical touch

Due to their light weight, portability and versatility, paper-based touch sensors are gaining prominence. A transparent bilayered nanopaper with a backbone of natural wood fibre was created with an infill of NFC [125]. This hybrid nanopaper's surface durability and optical transmission have been improved. A thin coating of carbon nanotubes was used to conduct the surface. This translucent conductive paper was used to construct a four-wire resistive touch screen. The sensor senses physical touch and transmits the information to the PC through an external controller. The letter 'paper' was successfully drawn, with a stylus pen, on the touch sensor and shown on the PC [126]. The utilisation of CNF spraying and electrospinning to create a transparent hybrid cellulose nano-fiber (CNF) film was depicted through this. The resultant hybrid film is very transparent and has a low thermal expansion coefficient. The hybrid films underwent spin coating employing AgNWs and then utilised as transparent electrodes in a

four-wire resistive touch panel. Additionally, its flexible and transparent touch panel delivers an unmatched sensory experience. Resistance to heat. Furthermore, a highly crystalline nanopaper was developed with greater than 90% transmission and less than 1% haze [127]. To conduct the nanopaper, a single sheet of graphene was deposited onto it via chemical vapour deposition (CVD) using a transparent conductive electrode The same process that is used to manufacture commercial plastic touch screens may be utilised to fabricate a capacitive multi-touch display on a nanopaper substrate. Sensory performance is equivalent to that of a transparent paper touch panel [114].

CNF epoxy films [126] are characterised by the innovative production process of using microporous epoxy nanofibres. Due to the difficulty of filling the micropores of electrospun epoxy fibres ,which were approximately 420 nm in diameter, with CNF, an epoxy 3D network is a unique method for spraying CNF dispersion into distilled water. Not only has the hybrid film been coupled with other components such as silver nanowires (AgNW), but also with transparent organic light-emitting diodes (OLED). Jung et al. [129] pioneered the use of a novel technique to create an integrated, vertically stacked NC tactile sensor. Using layered polymers, nanoparticles and nanocellulose, a platform for sensing pressure and temperature was constructed using the piezoelectric characteristics of cellulose substrates. They achieved stimulus sensitivity by using thermoelectric and piezoelectric methods to create inkjet prints and pyramid-sized cellulose, respectively. A tactile sensor array exhibited long-term bifunctional sensing capacity with a rapid response time and substantial interference between individual stimuli, which is necessary for electronic skin [40].

On the other hand, another group coupled the advantages of the fluorescence and conductivity of an active transparent ZnO SQD(spherical quantum dots) layer with photo-detection nanocellulose to create a hybrid device [130]. A smooth surface with optical quality was obtained while fabricating the paper by filtering an NC gel cake and replacing solvents with isopropanol, followed by pressing and a double-sided drying setup between two smooth fluoride membranes. This transparent SQD-NC paper is used to detect and produce light in electrical systems.

5.4 Metal ion detection

Kacmaz and colleagues were the first to employ the iron-sensitive fluorescent N′-(4-cyanobenzylidene) isonicotinohydrazide (CNINH) synthesised utilising ethylcellulose (EC) nanofibers using the electrospinning process to detect Fe^{3+} at Picomolar levels. The sensors responded quickly (30 s), which is said to be one of the quickest response times among comparable solid state sensors [117, 118]. Furthermore, unlike the Py/CNC sensors mentioned above, the sensor proved excellent at discriminating against other ions typically present in waterways and some physiochemical systems, such as Ca^{2+}, K^{+}, Mn^{2+}, Mg^{2+}, and Na^{+}. The detection limit of the EC sensor was correspondingly lower than that of the Py / CNC sensor and this substantial discrepancy in Fe^{3+} detection sensitivity can be attributed to the greater binding affinity of CNINH dye for EC nanofibers than Py for CNCs.

Li et al. synthesised nanofibers composited with pyromellitic dianhydride (PMDA) and deacetylated cellulose (DCA) for the simultaneous detection and removal of Pb^{2+} ions. The developed sensor was highly selective for Pb^{2+} ions and had a low detection limit. This detection value is within the WHO-recommended limit for acceptable Pb^{2+} contamination. After exposure to Pb^{2+} PbS in the presence of Na_2S, the PMDA/DCA sensor's colour changed from white to yellow-brown. To prevent interfering with PMDA adsorption sites such as Fe^{3+}, Cu^{2+}, Zn^{2+}, and Ca^{2+}, iminodiacetic acid (IDA) was introduced as a masking agent to a sensor. As a result, only Pb^{2+} cations were capable of initiating the characteristic transition from white to yellow-brown. After each treatment with an acidic solution such as HNO3, the sensor can be reused. The nanocomposite PMDA/DCA absorbed Pb^{2+} 12.7 times more than PMDA/paper, confirming the large area of porous cellulose nanofibers. The sole disadvantage of the PMDA/DCA sensor is its prolonged detection time (30 min). However, the developed sensors eliminated the requirement for complex equipment for direct visual colour evaluation. The findings indicated that the sensors developed had a high potential for detecting Pb^{2+} in polluted water.

Recently, Xu et al. developed a sensitive nitrite sensor by coating it with PDDA/CCNC in a poly(3,4-ethylenedioxythiophene) conduction (PEDOT) system and covered on the surface of the GCE [99]. Additionally, the sensors feature a fast reaction time and low detection limits Although these sensors do not have the lowest nitrite detection limit when compared to chitosan/Prussian blue/graphene nanosheets, carbon nanospheres[137], and manganese(II) complexes bulk-modified carbon sensors [138], the PDDA/CCNC/PEDOT sensor is less expensive, easier to construct and has a broader analytical limit. Additionally, the sensors have been demonstrated in real-world applications by detecting nitrite in a variety of pickled vegetables.

Hu and colleagues developed a 2-(5-Bromo-2-pyridylazo)-5-(diethylamino) phenol (Br-PADAP)/CA nanofiber sensor with a detection limit of 50 ppb in seawater for the purpose of developing portable, selective and sensitive colorimetric $UO_2{}^{2+}$ sensors [101]. When the sensors were subjected to $UO_2{}^{2+}$, they exhibited a pronounced yellow-to-purple colour change. Additionally, the developed sensors demonstrated a high degree of selectivity in solutions including the interfering metal ions Fe^{3+}, Cr^{3+}, Cu^{2+}, Pb^{2+}, Cd^{2+}, Zn^{2+}, and Mn^{2+}. This method has the potential to be extremely helpful in real-time detection applications due to its excellent selectivity, sensitivity and detection.

Weishaupt and colleagues developed "CySense", a highly copper-sensitive hybrid film based on cyanobacterial C-phycocyanin (CPC)/cyanobacterial nucleoside triphosphate (CNF). Although it needed a considerable incubation time, the sensor was capable of detecting Cu^{2+} at very low concentrations Other metals, such as Pb or Cd, may obstruct the sensor's response. These elements, however, were found at a concentration 20 times below the detection limit. Finally, the sensor demonstrated a high degree of precision in detecting free copper ions in human serum, indicating that it may be useful in healthcare applications. Yuen and colleagues advanced ion detection for medicinal purposes [103]. They developed a sensor that can be put to human skin in the form of a decal. The developed sensor is capable of detecting cations ions such as Na^+ in the absence of sample collection. The decal is composed of a layer of nanoporous bacterial cellulose, an insulating layer of SU-8, electrodes, and a doped-semiconducting polymer PEDOT/poly(styrene sulfonate) (PSS). The NC layer's property enables effective Na^+ transfer from the bottom surface to the top PEDOT/PSS sensing material. PEDOT is reduced to create an insulating material, which results in decreased electrical conductivity. The NC layer isolates electrical contact from the human body physically while allowing for high analyte delivery efficiency and detecting Na^+ in biofluids [133].

5.5 Electrical transistor-enabled nanopaper

Due of the low cost, recyclability and flexibility of the material, paper is a good substitute for plastics in the construction of flexible electronics. Owing to its great mechanical strength and possible transparency, cellulose nanopaper, a unique form of paper composed of nanoscale cellulose fibres, is a favorable material for the substrate of flexible and transparent electronics. As the technology combines these unique properties of renewable, recyclable resources, cost-effectiveness, light-weight and flexibility, nanopaper-based 'green' electronics is a rapidly growing field of research and development with significant academic and commercial interest. Due to the increased optical transmission and reduced surface roughness of nanopaper, it may support a range of devices that would not be conceivable with conventional paper [134].

However, due to the form stability of nanopaper after processing, device integration may provide considerable problems. It was demonstrated [135] for the first time that tailored nanopapers may be used to fabricate flexible organic field effect transistors (OFETs) with excellent transparency [136]. Cellulose nanopapers with high mechanical efficiency and conductivity in the semiconductor region were produced too. CNFs of bleached softwood pulp were covered with PPy by chemical polymerization in the presence of iron chloride (III).

The mechanical, conductive and thermal properties along with the morphology and structure of nanopapers have been studied. The tensile response of pure CNF nanopaper was exceptionally high. By adding up to 20% PPy to CNF/PPy nanopapers, considerable flexibility was achieved while maintaining excellent mechanical properties. The resulting materials are conductive and robust nanopapers that can be used as flexible biosensors and biodegradable thin film transistors [134].

Koga et al. [137] demonstrated the technique for fabricating extremely transparent conducting networks from cellulose nanopaper. The ease with which their aqueous dispersion could be filtered by the cellulation nanopaper, which acted as both a transparent flexible substratum and a filter, resulting in a uniform coating of conductive nanomaterials, being CNTs and AgNWs. The sheet resistance of the nanapaper AgNW networks was up to 75 times smaller than the sheet resistance of a film made using conventional polymer coating techniques (ethylene terephthalate). According to these findings, filtration coatings result in an even distribution of conductive networks due to perpendicular drainage via paper-specific nanopores, thereby being more beneficial thsn conventional coating processes which result in uneven distribution of conductive nanomaterial and self-aggregation due to the difficult drying method. Additionally, the driving networks have been incorporated into the nanopaper surface layer, exhibiting excellent adherence to the nanopaper substrate and assuring minimal fluctuations in electrical conductivity. This filtering technique is expected to be a viable covering method for a variety of conductive materials, with transparent conductive nanopaper being a possible material for future paper electronics [134].

Although cellulose nanopaper substrates exhibit significant optical haze, this is owing to their

high diffusive transmission and low direct transmission. Xu et al. [138] developed a straightforward method for producing high-tranquillity, low-hazard cellulose nanopapers containing short cellulose nanocrystals (CNC) and long CNF. The researchers were able to increase their total transmission, direct transmission, and diffuse transmission by changing the CNC-CNF ratio in the hybrid cellulose nanopaper. By increasing the CNC content, it is possible to minimise the optical haziness of hybrid cellulose nanopapers and increase their transparency. By covering the hybrid cellulose nanopaper with AgNWs, transparent and flexible electrodes were produced. The electrodes have a low sheet resistance and a high overall transmission To illustrate the electrodes' potential for use in flexible displays, they were used to construct a light emitting diode (LED) [134].

5.6 As an optical sensor platform

Different optical sensing platforms based on nanopapers were reported and how they may be modified to show plasmonic and photoluminescent characteristics suitable for sensing applications. Additionally, these researchers demonstrated the existence of other nanopaper structures, including plates, spots or cuvettes printed or perforated with cellulose nanopapers. Among the platforms were a colorimeter-based sensor made of nanopaper embedded with gold and silver nanoparticles; a photoluminescent sensor made of CdSe/ZnS nanopaper-connected quantum dots; and a potential nanopaper-functionalized upgrade sensing platform made of nanoparticles NaYF4:Yb3 / Er3 / SiO_2 Numerous biologically relevant models were utilised to examine how these platforms' photoluminescent or plasmonic properties can be modulated. Additionally, cellulose nanopaper was shown to be an effective preconcentration platform for the analysis of tiny amounts of optically active compounds. These platforms will pave the way for lightweight, transparent, flexible and easy-to-use optical theranostic devices and biosensors [40, 50, 112, 134].

5.7 As a water filtration membrane

Membrane operations are critical instruments in the fight against water pollution, to provide safe drinking water. This frequently comprises membranes constructed of synthetic polymers generated from ceramics or fossil fuels, the manufacturing of which requires substantial quantities of energy, chemical processes, and solvents. Thus, membranes derived from renewable resources, such as cellulose nanopapers, may be considered. Since ancient times, cellulose paper has been utilised as a filter and membrane material. On the other hand, conventional filter sheets composed of cellulose microfibrils have certain disadvantages when it comes to eliminating nanometer-sized contaminants. Mautner et al. [139] devised a method for enhancing the performance of nanopapers made from paper industry leftovers by including certain functional groups such as and ammonium (NH4) and phosphates (PO4 3). Nanopapers coated with these groups are predicted to exhibit adsorption properties not only in static environments, but also in the presence of nitrate and heavy metal ions. CNFs were effectively transformed to NH4′ or PO4 3 by the glycidy-trimethylammonium chloride and H3PO4 reactions, respectively. After filtering nanocellulose suspensions and filter cake in a hot press, CNF nanopapers of varying grades were obtained. Permeability and rejection values for nitrates and heavy metal ions were determined using filtration experiments. In dynamic filtration experiments, nanopapers with ammonium-modified CNFs captured up to 12 mg of nitrate per gramme of CNFs whereas phosphate groups were able to adsorb up to 20 mg of copper per gramme of phosphorylated CNFs, that were adsorbent in a range of industrial and agricultural batch-wave waste materials with the advantage of short contact time.

Wood-based Nanocellulose sheets have also created nanofiltration with organic solvents, after which, its usage was extended to highly permeable UF(ultra filtration) membranes for water purification. These nanocellulose-based membranes are composed of four different forms of nanocelluloses: celulose nanocrystals, bacterial cellulose, TEMPO-oxidized CNFs and wood nanocellulose. The molecular weight divisions of the membranes are split into a few nanometers of average pore size divisions. At the interface of nanofiltration and ultrafiltration, nanopaper rejection was perforated. Different types of CNFs may be used to control the porosity of nanopapers.

5.8 Hybrid features

Nanopapers with a heterogeneous network driven by upconverting nanoparticles (UCNPs) are very flexible, transparent, luminous and were manufactured in a matter of hours using a moderate-tension extrusion paper method [140]. After removing non-cellulosic components from skin using an efficient extraction technique combined with high pressure homogenization and high frequency ultrasonics, CNF nanopaper was successfully produced. An effective epoxidation procedure was used to enhance the activity of UCNPs by the capping the surface with oleic acid

ligand. Following epoxidation, UCNPs formed UCNP-grafted CNF (CNFdUCNP) suspensions in aqueous medium. The papermaking process demonstrated exceptional features such as outstanding transparency and brightness along with good flexibility in the constructed fluorescent CNFd UCNP hybrid nanopaper. The experimental results indicate that UCNPs were effectively grafted onto the CNF matrix via heterogeneous networks, resulting in a nanopaper with excellent clear and brilliant optical characteristics.

Another hybrid nanopaper with high optical transmission and iridescence under a polarising layer by integrating native CNFs into the cellulose nanowhisker matrix (CNW). The nanopaper is less expensive than tidy, large-scale carbon nanofiber nanopaper. Additionally, the transparent hybrid nanopaper's writing surface is comparable to that of normal paper. When compared to conventional paper, hybrid nanopaper exhibited improved optical characteristics and a smoother surface. Due to the combination of these properties, this nanopaper is an ideal substrate for flexible electrical devices [134].

5.9 Reactive cellulose nanopaper template

CNFs have excellent interfacial adhesion potential owing to the high density of surface hydroxyl groups. A novel nanocomposite system was [141] examined utilising a porous carbon nanotube system soaked with a methanol solution including a melamine-based curing agent for a reagent hyperbranched functional aliphatic polyester. The technique includes substituting a nanocellulose hydrogel for the solvent and impregnation with a polymer solution [142]. The underlying process was predicated on the fact that molecular mobility was reduced in the geometrically restricted polymer network as a result of interactions with CNF surfaces. Nanofibre couplings in a covalent, solid matrix have developed as hyperbranched aliphatic polyesters may chemically react with hydroxyl groups on nanofibre surfaces. In the restricted region within the nanofibre network, the cross-linking polymer matrix exhibited dramatically changed properties, including a much higher Tg [134].

5.10 Biodegradable nanopaper

Developing ecologically friendly and affordable electronics is an efficient way to solve the issue of electronic waste. Transparent nanopaper was coupled with polylactic acid (PLA) electrets to generate a biodegradable and transparent electric nanogenerator based on paper [143]. Due to the nanogenerator's transparency and desirable performance, it may be used to create a smart, self-powered packaging system with minimum visual effect. Additionally, the nanogenerator's self-degradation characteristic has been proven in its native soil, indicating that the nanogenerator is recyclable and does cause pollution in/to the environment. This research will provide new light on the development of environmentally friendly gadgets and energy sources [128, 134].

5.11 Thermally conductive and optically transparent bilayer

The optical transparency, mechanical robustness and biodegradability of transparent nanopapers constructed of CNFs indicate that they might be utilised in electronics in place of plastics. However, due to the high thermal conductivity of cellulose nanopaper and its limited thermal stability at elevated temperatures, it is inappropriate for long-term stability and device dependability. A bilayer nanopaper with a layer of thin boron nitride (BN) nanosheets [144] was developed that is thermally conducting, electrically isolating and optically clear. This form of optically transparent nanopaper is used in a variety of electronic devices, including bilayers that are both electrically isolating and thermally conducting [42].

6 Perspectives and conclusion

CNFs are gaining traction as a renewable and sustainable feedstock for ecologically friendly, bio-based, high-performance materials in the future. Nanocellulose enables the fabrication of pure nanopapers or the integration of pure nanopapers into bioinspired nanocomposites, resulting in remarkable multifunctional characteristics. While great progress has been achieved in the creation of cellulose nanopapers, certain practical issues remain. The major issue is that mechanical separation of CNFs consumes considerable energy. Appropriate pre-treatments are necessary to mitigate this problem prior to mechanical insulation. The second difficulty arises from the hydrophilicity of cellulose polymers. Water removal from nanocellulose dispersions is a difficult problem that demands the development of surface functional methods for optimising nanofibril dewatering. The authors obtained new insights in order to concentrate on drying the CNF suspension, which is important for future applications. The relevance of drying stems from the fact that drying alters the size of the CNF, which has an effect on its characteristics. Further research should finally concentrate on the most effective methods for production, processing,

modification, and drying of CNF. The third objective is to make cellulose nanopapers at high speeds and at a cheap cost [23, 117].

Paper has gained popularity as a low-cost substrate for a variety of sensing devices due to its lower cost than traditional polymer substrates (*i.e.* paper is made from renewable natural resources and thus costs less than traditional polymer or silicone substrates; for example, paper is flammable in nature and hence paper-based devices could be easily and safely disposed though incineration). Concerns like the ruggedness, porosity, and contaminants connected with the manufacture of paper-based electrical devices must be addressed in order to improve their performance. Chemical impurities in paper, as well as environmental variables such as temperature and humidity, may significantly alter the dimensions of paper and the conductivity of paper-based printed conductive structures. The porous nature of paper makes it ideal for energy storage applications. The energy density of paper-based energy storage devices remains substantially lower than that of commercially available systems. On the other hand, paper-based equipment is well-suited to flexibility and environmental friendliness. Additional basic research may be necessary to optimise the performance of paper-based devices [114]. While paper has great promise as a material for disposable point-of-care (POC) devices, design and manufacturing advances are necessary to match the performance of current analytical procedures. The most common issue with paper microflow equipment is that the concentration of the analyte entering the paper microcanal changes owing to evaporation; one solution is to cover the canal region on both the top and bottom with printing toner [100].

Numerous paper-based sensing devices published to date perform wonderfully in laboratory size studies, but fall short in terms of repeatability and reproducibility when compared to commercial devices. As a result, it is important to enhance the performance of these paper devices by modifying their design and manufacturing features. Several studies on the effect of numerous physical properties of paper on sensor performance, such as surface energy, degradation, temperature, and humidity, are critical for bringing paper-based sensing systems closer to market. Researchers in the paper industry, materials science, bioscience and electrical engineering should make a concentrated effort to commercialise prototypes.

Numerous manufacturing techniques for developing leading or hydrophobic barriers in paper substrates have been developed to far for the purpose of fabricating paper-based sensing devices. Most of these activities do not require specialised equipment and may be done under standard laboratory conditions without the use of cleanroom facilities. While these manufacturing processes are extremely efficient on a laboratory scale, they fall short of industrial standards when scaled up to mass production. The paper and pulp industries employ a variety of fibres from a variety of sources to create paper of varied grades. As a result, the effect of paper type on sensor performance should be investigated, as paper properties like as porosity, mechanical damping, and so on are strongly influenced by fibre type and orientation, among other factors. Additionally, the bulk of papers to date have focused on developing scalable manufacturing processes and proving novel applications for paper-based devices. However, extensive mathematical modelling of paper-based devices has not been conducted, despite the fact that paper modelling has the potential to resolve a large number of the difficulties connected with paper-based devices.

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