Cellulose, Nanocellulose, and Antimicrobial Materials for the Manufacture of Disposable Face Masks: A Review

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Cellulose is among the most promising renewable and biodegradable materials that can help meet the challenge of replacing synthetic fibers currently used in disposable N95 respirators and medical face masks. Cellulose also offers key functionalities that can be valued in filtration applications using approaches such as nanofiltration, membrane technologies, and composite structures, either through the use of nanocellulose or the design of functional composite filters. This paper presents a review of the structures and compositions of N95 respirators and medical face masks, their properties, and regulatory standards. It also reviews the use of cellulose and nanocellulose materials for mask manufacturing, along with other (nano)materials and composites that can add antimicrobial functionality to the material. A discussion of the most recent technologies providing antimicrobial properties to protective masks (by the introduction of natural bioactive compounds, metal-containing materials, metal-organic frameworks, inorganic salts, synthetic polymers, and carbon-based 2D nanomaterials) is presented. This review demonstrates that cellulose can be a solution for producing biodegradable masks from local resources in response to the high demand due to the COVID-19 pandemic and for producing antimicrobial filters to provide greater protection to the wearer and the environment, reducing cross-contamination risks during use and handling, and environmental concerns regarding disposal after use.

Keywords: Antimicrobial properties; Cellulosic biomaterials; Cellulose filaments; COVID-19; Fibrillated cellulose; Filter media; Medical face masks; N95 respirators; Personal protective equipment; SARS-CoV-2

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INTRODUCTION

The outbreak of coronavirus disease 2019 (COVID-19) and the associated collapse of the global supply chain have resulted in a severe global shortage of personal protective equipment (PPE)—especially N95 respirators and medical face masks—for healthcare personnel, thereby stimulating the search for local supply sources and innovative solutions. Additionally, the use of non-medical masks by the entire population has been imposed by several jurisdictions worldwide in an effort to mitigate the spread of severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2), which causes COVID-19, and has also led to an increasing demand for masks. Currently, protective face masks are generally made from petroleum-based synthetic fibers, although the use of biodegradable materials with a lower carbon footprint is preferable, provided the protection performance is maintained. The forestry sector, particularly the pulp and paper industry, plays an important role in the context of the crisis caused by the pandemic, as it can supply a wide variety of cellulose-based products for healthcare personnel and the general public. However, few pulp and paper companies in North America produce medical-grade pulp for PPE. For instance, Canada has a single mill producing medical-grade pulp that can be used in the composition of N95 respirators and other PPE by United States manufacturers. This mill has been operating at full capacity since the appearance of COVID-19 (Business Examiner 2020).

For several reasons, cellulose is one of the most promising alternative materials that can be used to partially or entirely replace synthetic fibers traditionally used as filter media in the manufacture of disposable N95 respirators and medical and non-medical face masks. These reasons include its biodegradability, renewability, low cost, abundance, ease of processing, adjustable aspect ratio, strong mechanical properties, and low density, compared to other materials (Tavakolian *et al*. 2020). Both N95 respirators and medical face masks require a combination of high filtration capacity and good performance against the penetration of pathogens. The latter requires, among other characteristics, water repellency, as liquid droplets may contain viral agents (Shimasaki *et al*. 2020), and also bioactive properties including mainly an antimicrobial action (Ristić *et al*. 2011). Face masks should also allow for a low pressure drop (*i.e*., high air permeability) to facilitate breathing (Osman 2020), while being comfortable, soft, light, and non-allergenic to the wearer’s skin (Zanoaga and Tanasa 2014; Qin 2016), all in accordance to regulatory standards (42 CFR Part 84 1995; NIOSH 2014; ASTM F2100-19e1 2019; NPPTL 2020). Both N95 and medical face masks made of synthetic fibers have a high level of filtration of greater than 95%; however, few masks available on the market have antiviral activities, meaning that they filter microorganisms but do not deactivate them. This in turn increases the risk of cross-contamination (from the contaminated mask to the wearer or the environment) during use and handling and raises further environmental concerns about their disposal after use (Zhou *et al*. 2020). Cellulose fibers provide desirable properties to filter media (*e.g*., bulk, permeability, and mechanical strength) and can act as a support structure for the thin and weaker filter media such as meltblown and electrospun matrices (Hutten 2016). Additionally, surface modification of cellulose fibers is considered an excellent option for adding antimicrobial functionality to medical and healthcare products. Although it does not exhibit intrinsic biocidal activities like some biopolymers, *e.g*., chitosan, the cellulose surface is reactive, so it can be chemically modified to graft certain functionalities into its structure (Tavakolian *et al*. 2020). Several technologies have been developed to introduce bioactive compounds, including plant-based products (Catel-Ferreira *et al*. 2015; Gargoubi *et al*. 2020), animal-derived chitosan and chitosan derivates (Ling *et al*. 2013; Ahmad *et al*. 2016), metal and metal compounds (Emam 2019), metal-organic frameworks (MOFs) (an innovative material derived from metal salts) (Nie *et al*. 2020a), inorganic salts, synthetic polymers (Tavakolian *et al*. 2020), two-dimensional (2D) nanomaterials such as graphene (Huang *et al*. 2020), and composite and nanocomposite structures thereof among others, which provide antimicrobial (antibacterial, antifungal, and/or antiviral) properties to cellulose fibers. These antimicrobial materials can be added either during papermaking or as post-treatment coatings to provide effective protection. Furthermore, when cellulose fibers are disintegrated into nanostructures, *e.g*., nanofibrillated materials, many filter properties and characteristics such as filtration efficiency (Sim and Youn 2016) and surface functionalization are enhanced due to their high specific surface. Additionally, cellulose may be combined with other materials, such as polyester fibers, forming composite structures to meet the requirements for filter media (Pan *et al*. 2019). However, cellulose fibers are inherently hydrophilic, so they tend to absorb water, which softens and weakens the filter structure, reduces its useful life, and inhibits filtration performance due to fiber swelling (Mukhopadhyay 2014). This characteristic of the material increases the risk of contamination during use, as viruses can penetrate through a humid mask (Zhou *et al.* 2020), while also providing favorable conditions for the proliferation of fungi and bacteria. Therefore, treatments providing antimicrobial and hydrophobic properties must be applied to cellulose-based masks. In short, the use of cellulose fibers and nanocellulose products from wood pulp offers numerous advantages for N95 respirators and medical face masks, yet it also raises some key technical challenges that must be overcome to meet the requirements for good filter performance.

This paper presents a review of the potential applications of cellulose and nanocellulose materials in the development of biodegradable products to replace the petroleum-based synthetic fibers usually used in the manufacture of disposable N95 respirators and medical face masks. It offers 1) an overview of the structures and compositions of disposable protective face masks, their properties, and standard requirements and particularities of the filter media; 2) descriptions of the cellulose and nanocellulose materials that could be used in the manufacturing of mask filters; and 3) a suite of antimicrobial technologies that could be used for cellulose functionalization and that remain potentially safe for use in face masks. The study is partly a result of a joint effort of researchers and industrial partners of the pulp and paper sector to find innovative solutions for bioproducts obtained from locally and sustainably sourced fiber, which will hopefully contribute to limiting the spread of SARS-CoV-2.

DISPOSABLE FACE MASKS

N95 Respirators

The National Institute for Occupational Safety and Health (NIOSH) approves particulate filtering facepiece (FFP) respirators according to nine filter classes certified under 42 CFR Part 84 (1995): N95, N99, N100, R95, R99, R100, P95, P99, and P100. The N, R, and P designations refer, respectively, to respirators that are non-resistant, resistant, and proof to degradation by oily aerosols. The protection levels of classes 95, 99, and 100 correspond to filtration efficiencies of airborne particles of at least 95%, 99%, and 99.97%, respectively. The N95 is therefore a particulate FFP respirator that is non-resistant to oily aerosols and that filters at least 95% of airborne particles greater than 0.3 μm, which is considered the most penetrating particle size (MPPS). These respirators are classified as protective devices or PPE and are designed to protect the wearer against the inhalation of hazardous airborne particles, including viruses and bacteria. There are two NIOSH-approved N95 FFP types: standard N95 respirators and surgical N95 respirators, which are also referred to as medical/healthcare respirators and cleared by the US Food and Drug Administration (FDA) (NIOSH 2014; NPPTL 2020). North America follows NIOSH specifications for N95 respirators; however, equivalent FFP respirators from Europe (FFP2), China (KN95), and other countries are commercially available in a variety of shapes and sizes.

The N95 respirators have a multilayer composite structure with a central filtering layer displaying electret properties, giving an electrostatic charge to a nonwoven fibrous mat often composed of synthetic fibers. They are generally composed of four layers: two layers (outer and inner) of spunbonded polypropylene of low density (20 g/m^2 to 50 g/m^2 basis weight); a thick, stiff, and dense nonwoven layer (approximately 250 g/m^2 basis weight) that gives support to the meltblown layer and that can be molded to give the shape of the mask; and a meltblown layer with electrostatic properties (Henneberry 2020). The electrostatic charge of the filter media is generally obtained by corona discharge, triboelectrification, or electrostatic spinning. Electret treatments provide many advantages to filter media without increasing structural mass or density (Zhao *et al*. 2020). These advantages include high initial and sustained efficiency during the filter lifecycle, increased submicron particle capture efficiency, filtration efficiency unaffected by relative humidity and long-term storage at high temperatures (54.5 °C), and lower airflow resistance (Mukhopadhyay 2014). The N95 respirator's performance depends on the filtration efficiency as well as on face seal leakage, as respirators must fit tightly to the face of the wearer to create a seal (He *et al*. 2013).

Filtration performance of N95 respirators is evaluated according to 42 CFR Part 84 (1995) and NIOSH standards by polydisperse sodium chloride (NaCl) aerosol testing with an aerosol concentration not exceeding 200 mg/m^3 (typical NaCl concentrations range from 12 mg/m^3 to 20 mg/m^3) at an 85-L/min flow rate using a TSI 8130 automated filter tester (TSI Incorporated, Shoreview, MN, USA) (Rengasamy *et al*. 2011, 2013), which is a photometric detection method that uses a forward light scattering photometer to measure the flux of light scattering from particles (Rengasamy *et al*. 2011). This method determines the penetration or filter efficiency and pressure drop in filter media, filter cartridges, and respiratory masks. The NaCl aerosol must have a particle size distribution with a count median diameter (CMD) of 0.075 µm ± 0.020 µm (size distribution can be determined using differential mobility analyzers) and a standard geometric deviation (GSD) of less than 1.86. Additionally, the NaCl aerosol must be at 25 °C ± 5 °C and 30% ± 10% relative humidity and neutralized to the Boltzmann equilibrium state (the remaining charged particles must have a bell-shaped distribution curve with the center at zero). The 42 CFR Part 84 (1995) standard requires pre-conditioning of the N95 respirators before the test (at 85% ± 5% relative humidity and 38 °C ± 2.5 °C for 25 h ± 1 h) because some types of electrostatic filter media can degrade after exposure to high humidity. The filtration efficiency is the percentage of NaCl particles filtered by the material, while the pressure drop corresponds to the air resistance across the filter material (Zhao *et al*. 2020). The maximum penetration must not exceed 5% for class 95 respirator approval.

Other filtration test methods have also been studied. A comparison between the photometric method and another polydisperse NaCl aerosol penetration method using an ultrafine condensation particle counter (UCPC) to measure the filtration performance of N95 respirators was presented by Rengasamy *et al*. (2011). The authors showed that aerosol penetrations measured by the UCPC were 2 times to 6 times greater than the levels measured by the photometric method, suggesting the need for the development of a more challenging aerosol testing method for N95 respirator certification. In addition to the NaCl aerosol test, N95 respirators must meet other specifications according to 42 CFR Part 84 (1995) and NIOSH standards, such as inhalation and exhalation resistance to airflow (≤ 343 Pa and ≤ 245 Pa, respectively) for both N95 types, synthetic blood penetration (ASTM F1862/F1862M-17 2017) and biological filtration efficiency for the surgical N95 type, and exhalation valve leakage (leak rate 30 mL/min) and force applied (-245 Pa) for the standard

N95 type. Other tests can also be performed, such as surface wetting resistance, microorganism index (bioburden) (Davison 2012), CO_2 clearance, and total inward leakage (TIL), which quantifies the respirator's ability to fit individual facial dimensions to ensure that the respirator fits properly (not a requirement for respirator approval testing) (He *et al.* 2013, 2014; Ramirez 2015).

Medical Face Masks

Medical face masks, also known as surgical masks or procedure masks, are usually composed of three layers of a spunbonded-meltblown-spunbonded (SMS) nonwoven fabric, where the meltblown middle layer provides higher filtration performance compared to the spunbonded layers (Ghosh 2014; Mukhopadhyay 2014). The outer spunbonded layer is typically blue and hydrophobic to prevent liquid droplets containing viral or other hazardous agents from reaching the wearer (Shimasaki *et al*. 2020), acting as a prefilter layer for the meltblown middle layer that provides the final filtration (Mukhopadhyay 2014). The inner spunbonded layer is soft, for the wearer's comfort, and absorbent, to absorb the fluids generated by breathing, coughing, and sneezing, and it usually has no filtration function. The spunbonded layers are generally made of polypropylene, polyester (polyethylene terephthalate, PET), or other thermoplastic fibers (Henneberry 2020). Cellulose fibers such as cotton or rayon fibers, which are made from regenerated cellulose from wood or agricultural residues, and mixes of cellulose/polyester and cellulose/ polyolefin fibers can also be used in the outer and inner layers (Davison 2012; Ciuffreda *et al*. 2020; Medline 2020). Spunbonding is a well-known continuous nonwoven process in the field of textiles. It is fast and inexpensive and combines spinning, sheet formation, and bonding: Fibers are spun, dispersed by deflectors or air streams to form a web, and finally bonded by hydroentangling, needle punching, or thermal or chemical methods (Lim 2010).

The meltblown filter of the middle layer is composed typically of polypropylene; however, other synthetic fibers can be used, such as poly(butylene terephthalate), a type of polyester, and poly(tetrafluoroethylene) (PTFE), a thermoplastic polymer with inherently hydrophobic properties used to form a porous membrane structure (Solvay 2020). The pore diameter in PTFE stretched films ranges from 0.1 μm to 10 μm, providing high filtration efficiency, lightness, high air permeability (breathability), and long service life; however, they are expensive and cannot completely replace traditional masks (Hendrikx 2019). The melt blowing process is a conventional method for producing micro- and nanofibers from thermoplastic polymers. In the process, a polymer melt is extruded through small nozzles surrounded by high-velocity hot air to produce fibers that are randomly deposited to form a nonwoven web (Hiremath and Bhat 2015). A combination of SMS technologies, known as a multi-denier process, is also employed to form a composite structure and reach the requirements of protective textiles (Lim 2010). Spunbonded fibers are coarser, have higher tensile strength, and have a smaller pressure drop than meltblown fibers (Mao 2016). The polypropylene microfibers of the meltblown layers have diameters of approximately 1 μm to 10 μm and a fabric thickness of 100 μm to 1000 μm, which provides high porosity and a limited capacity to filter fine particles (Zhao *et al.* 2020). For this reason, other filtration technologies are employed to improve filter performance. Unlike N95 respirators, which are electret-treated to improve filtration efficiency against small aerosols, the meltblown layer of medical face masks has no electret properties. The filtration of particles is only mechanical and therefore less efficient than in N95 respirators. Medical face masks are hence designed mainly to prevent the projection of droplets expelled by the wearer into the environment, *e.g.*, to prevent surgical infections and to protect others from droplets originating from sick people. Consequently, they offer low protection for the wearer against the inhalation of fine airborne particles.

Prather *et al*. (2020) offer a helpful explanation to distinguish between aerosols and droplets: Aerosols are smaller than 100 μm, remain suspended in the air for long periods (ranging from several seconds to hours), and can be inhaled after traveling more than 2 m and accumulating in poorly ventilated indoor air. In contrast, droplets are larger than 100 μm and fall to the ground in seconds, usually within a distance of 2 m from the source. Transmission of SARS-CoV-2 occurs by both aerosols and droplets (Prather *et al*. 2020). Although the filtering efficiency of medical face masks is generally tested with nonbiological particles, a recent study has confirmed their efficiency in preventing transmission of human coronaviruses and influenza viruses from symptomatic individuals, suggesting that their use could also help control COVID-19 transmission (Leung *et al*. 2020). The study showed that medical face masks significantly reduce the environmental emission of the influenza virus in coarse aerosols (particles > 5 μm) and coronavirus in fine-particle aerosols (≤ 5 μm), with a trend to reduce the coronavirus emission in respiratory coarse aerosols. Medical face masks offer more comfort and greater breathability than N95 respirators during use.

Other filtration mechanisms including antiviral technology and nanotechnology have been developed to improve the performances of N95 respirators and medical face masks. The BioFriend™ BioMask™ provides FDA-cleared disposable N95 surgical respirators and medical face masks with antiviral properties. These masks are composed of four layers, with two active layers designed to inactivate viruses through different mechanisms of action. The outer active (first) layer is composed of spunbonded polypropylene treated with a hydrophilic plastic coating that absorbs infectious droplets and inactivates viruses by exposure to citric acid derivates. The inner active (second) layer is composed of cellulose (rayon) and polyester fibers treated with positively charged copper and zinc ions. These active layers inactivate different subtypes of influenza A and B viruses, paramyxovirus (measles), and SARS-CoV-1, which was responsible for the severe acute respiratory syndrome (SARS) outbreak in 2003. The third layer is composed of meltblown polypropylene filter media, while the inner (fourth) layer is composed of spunbonded polypropylene. The antiviral N95 respirators and medical face masks have the same structure, except for the meltblown material used in the third layer of the N95 respirator, which has increased filtration efficiency to meet the required specifications (Davison 2012). The same antiviral technology is used on the Innonix RespoKare® surgical masks, which inactivate many pathogens, including coronaviruses, but they had not yet been proven efficient for SARS-CoV-2 (RespoKare 2017). Additionally, the Inovenso company has developed a new generation of nanofiber face masks, named Inofilter® 95/99, with a three-layer structure made of PET spunbonded nonwoven fabric in the outer and inner layers (35 g/m^2 basis weight for each layer) and a nanofiber membrane in the middle layer (0.6 g/m^2 to 0.8 g/m^2 basis weight) replacing the meltblown middle layer conventionally used in face masks. The nanofiber membrane is made of a thermoplastic fluoropolymer (polyvinylidene fluoride, PVDF) using a patented hybrid electrospinning technology. The PVDF nanofiber membrane has fibers with a diameter of approximately 224 nm and is designed to provide a high mechanical filtration efficiency of 96% to 99%, inhalation protection, and low pressure drop. Another similar version of the nanofiber filter, the Inofilter V®, provides additional properties such as 99.9% viral filtration efficiency, inhalation and exhalation protection, and resistance to liquids like blood and oil (Inovenso Technology 2018).

Medical face masks must meet five specifications for filtering according to the ASTM F2100-19e1 (2019) standard: 1) bacterial filtration efficiency (BFE), 2) submicron particulate filtration efficiency (PFE), 3) differential pressure (ΔP), 4) resistance to penetration by synthetic blood, and 5) flammability. Medical face masks are classified according to three levels of protection: Level 1 (low), Level 2 (moderate), and Level 3 (high), with differing specifications. The BFE tests measure the percent efficiency of the material in preventing the passage of bacteria through the mask. The BFE test is performed with an aerosol of *Staphylococcus aureus* bacteria (particles of approximately 3 µm) at a constant flow rate of 28.3 L/min (*i.e*., a normal breathing flow rate) (ASTM F2101-19 2019). Submicron PFE tests measure the initial particle filtration efficiencies of materials used in medical face masks using monodispersed aerosols of latex particles, usually at 0.1 µm in diameter (ASTM F2299/F2299M-03 2017). Both BFE and PFE must meet the requirements of $\geq$ 95% for low-barrier face masks (Level 1) and $\geq$ 98% for moderate- and high-barrier face masks (Levels 2 and 3). Differential pressure tests measure the pressure drop across a medical face mask under specific conditions of airflow, temperature, and humidity. The ΔP metric is an indicator of the breathability of the mask, expressed in mm H_2O / cm^2 or Pa/cm^2. The ASTM F2100-19e1 (2019) standard requires a ΔP of < 4.0 mm H_2O / cm^2 (< 39.2 Pa/cm^2) for low-barrier face masks (Level 1) and of < 5.0 mm H_2O / cm^2 (< 49.0 Pa/cm^2) for moderate- and high-barrier face masks (Levels 2 and 3) (Public Works and Government Services Canada 2020). Lower pressures indicate higher breathability or higher air permeability. The breathability of medical face masks depends on several parameters, such as textile type, the applied finish, the thickness, and the number of layers (Rogister and Croes 2013). Resistance to penetration by a synthetic blood is tested according to the ASTM F1862/F1862M-17 (2017) standard, which requires resistance to synthetic blood at pressures of 80 mm Hg, 120 mm Hg, and 160 mm Hg to qualify for the low-, moderate-, or high-barrier classes, respectively. Flammability tests are based on the rate of flame spread on the material. All medical face masks must meet the requirements for class 1 (Nelson Labs 2019).

Filter Media

Protective masks have an air filtration system that can be composed of various filter media. The air filter medium is the part of the filter that separates harmful particles from the air. Air filters are used for a wide range of applications, including vehicle air, commercial and residential indoor air (*e.g*., heating, ventilation, and air-conditioning systems and high-efficiency particulate air (HEPA) filters), clean rooms, laboratory hoods, large baghouse filters, flue gas scrubbers, industrial dust collectors, vacuum cleaner bags, medical protective devices (*e.g*., respirators and face masks) (Mukhopadhyay 2014), non-medical face masks, and reusable cloth masks made with a replaceable filter bag. Nonwoven filter media are formed by dry-forming, wet-laid, or combined technologies using many processes depending on the raw material (polymer or fiber source) and filter end use. Dry-forming processes include air-laid web (used to form absorbent materials, normally from cellulose fluff pulp), dry-laid web (used to form many felt materials used for filtration), melt spinning (used to produce spunbonded and meltblown microfibers from melt polymers), and nanofiber spinning (used to produce nanofibers by electrospinning or centrifugal spinning methods) processes. The wet-laid process is used to form webs from wood pulp, vegetable fibers, fibers of polyester, nylon, rayon, or any other material that can be dispersed in water, whereas combined technologies are used to form multi-layer composite structures, as in SMS technology, and nonwoven-membrane materials using different processes and/or materials (Hutten 2016).

Air filter media are designed to filter airborne particles using various filtration mechanisms. There are five filtration mechanisms by which airborne particles (aerosols) can be controlled: interception, inertial impaction, Brownian diffusion, gravitational settling, and electrostatic attraction (used in N95 respirators). In N95 respirators and medical face masks, filters must have a porous structure designed to maximize the space for filtration while keeping the differential pressure sufficiently low to maintain breathability (Vaughn and Ramachandran 2002; Institute of Medicine 2006). Medical face masks comprise a combined filtration mechanism: Inertial impaction and interception predominate in the BFE test for the capture of large particles, while Brownian diffusion predominates in the PFE test for submicron particles (Tong *et al.* 2016). The N95 respirators are also built to capture particles of different sizes: Large particles (> 1 µm) are captured by interception and inertial impaction, while small particles (< 0.1 µm) are captured by Brownian diffusion, and both small and large negatively charged particles can be captured by electrostatic attraction, which has the advantage of maintaining a low airflow resistance (Institute of Medicine 2006). Protective masks generally use a filtering gradient from the largest to the smallest particles through the mask. This way, the first layer of the filter captures the larger particles and prevents clogging of the pores in the subsequent layers.

Hutten (2016) divided the raw materials used for filter media into four overlapping categories: polymers, fibers, resins and binders, and additives. Polymers used in nonwoven materials include fibers (*e.g.*, meltblown and spunbonded polypropylene, cellulose from wood pulp and vegetable fibers, and regenerated cellulose or rayon), resins, and additives. Fibers comprise natural fibers (*e.g.*, wood pulp, vegetable fibers, and cotton), biofibers (*e.g.*, rayon and lyocell), and synthetic fibers, to name only the most pertinent for mask manufacturing. The most important physical characteristics of fibers for use in filter media are diameter, length, aspect ratio (length-to-diameter ratio), density, linear density (or fiber coarseness, weight in mg per 100 m of fiber), cross-section shape, internal structure (cellular or solid), and strength properties, which include tensile strength, breaking length, stretch or elongation, and stiffness. The ideal properties of the fibers are those that optimize the bulk, air permeability, and pore size of the filter media. When designing filters, the goal is to maximize bulk and air permeability to permit breathability while minimizing pore size to improve filtration efficiency. However, these properties are not directly compatible, as thin fibers provide high density, small pore size, and high filtration efficiency to filter media, but they provide low air permeability. Conversely, coarse fibers provide high bulk and permeability, but they lead to large pore size and thus offer poor filtration efficiency for filter media. In general, the spunbonded web of protective masks is formed by coarse fibers, whereas the fibers are much smaller in the meltblown web. Spunbonded fibers have diameters ranging from 1 µm to 50 µm (ideal range is from 15 µm to 35 µm) and basis weights (grammage) of the filters ranging from 5 g/m^2 to 800 g/m^2 (typically between 10 g/m^2 and 20 g/m^2). Their web thicknesses range from 0.1 mm to 4.0 mm (typically between 0.2 mm and 1.5 mm), and they are characterized by high strength-to-weight ratios compared to other structures and high liquid retention capacities. Meltblown fibers have diameters ranging from 0.5 µm to 30 µm (typically between 2 µm and 7 µm) and basis weights of the filters between 8 g/m^2 and 350 g/m^2 (typically between 20 g/m^2 and 200

g/m^2) (Hutten 2016). These microfibers have high surface areas for good filtration, smooth textures, and circular cross-sections (Malkan and Wadsworth 1993; Hutten 2016).

The aspect ratio affects the quality and performance in nonwoven webs. In general, synthetic microfibers form highly porous webs and are water repellent, but they are advantageous in filter media because they provide low pressure drop and low wettability, due to a low surface energy. Resins and binders for face masks must provide softness to give greater comfort for skin contact and have skin-friendly properties. Additionally, additives may be used in nonwoven filters, including adsorbent materials (*e.g.*, activated carbon—many replaceable filter cartridges for non-medical face masks are constructed with an activated carbon filter layer), flame retardants (required for cellulose-based filters), water repellents (important for the outer layer of masks), antimicrobial agents, and inks and colorants (used for aesthetic and operational purposes) (Hutten 2016). Debonding and softening agents may be particularly interesting for cellulose-based filters used in face masks. Debonder technology is used in paper products, *e.g.*, tissue paper, to prevent excessive bonding between one cellulose fiber and another and to improve the smoothness of the paper (Furman, Jr. *et al*. 2013). Debonders provide surface smoothness and increased softness, bulk, and sheet flexibility, among other advantages, to paper sheets (Solenis 2015).

Filter media properties can be affected by several factors related to fiber characteristics (synthetic or natural source, chemical composition, diameter, geometry, specific surface, and density), filter characteristics (thickness, density, bulk, porosity, and pore size distribution) (Vaughn and Ramachandran 2002), filtration velocity (based on flow rate and surface area), filtration mechanisms, and operational conditions (temperature and humidity) (Mostofi *et al*. 2010). The main properties of filter media are determined according to Eqs. 1 to 6.

The filtration efficiency (E) (expressed in %) is calculated by Eq. 1,

$$E = \left(1 - \frac{N_{\text{down}}}{N_{\text{up}}}\right) \tag{1}$$

where N_{down} and N_{up} are the downstream and upstream number concentrations of the aerosol particles, respectively (Long *et al*. 2018; Lu *et al*. 2018; Pan *et al*. 2019).

The pressure drop across the filter (ΔP) (Pa) is determined by the difference between the upstream (P_{up}) and downstream (P_{down}) pressures (Lu *et al*. 2018; Ma *et al*. 2018; Nie *et al*. 2020a).

In addition to standard filtration efficiency and pressure drop tests to determine the filter media properties of N95 respirators and medical face masks, a filtration quality factor (Q) (generally expressed in Pa^{-1}) is commonly used and calculated as shown in Eq. 2 (Mao *et al*. 2008; Alexandrescu *et al*. 2016; Long *et al*. 2018). Generally, large Q values indicate better filter quality, which conjugates high filtration efficiency (low particle penetration) with low pressure drop (high breathability) (Zhao *et al*. 2020).

$$Q = -\frac{\ln{(1-E)}}{\Delta P} \tag{2}$$

Air permeability is the airflow rate measured through a specific area of the filter media at a specific pressure drop and is generally expressed in $cm^3/(s \cdot cm^2)$, $ft^3/(min \cdot ft^2)$, or cfm. The test is performed by an automatic air permeability testing apparatus according to the ASTM D737-18 (2018) standard. Like the pressure drop test, is an indicator of breathability. The airflow depends on ΔP and filter thickness (t) and is measured by the equation proposed by Whitaker (1986) (Eq. 3),

$$v_0 = -\frac{k}{\mu}\frac{\Delta P}{t} \qquad (3)$$

where v_0 is the airflow velocity, k is the permeability constant, and μ is the viscosity of the air (Pan *et al.* 2019). The air permeability can thus be adjusted by changing the basis weight, thickness, and/or density of the filter. For instance, as filter thickness decreases, air permeability increases (low pressure drop), although the filtration efficiency decreases (Nie *et al*. 2020a).

The apparent density and porosity both affect the filtration efficiency and pressure drop of the filter media. The density of the filter (ρ_f) (expressed in g/cm^3) is calculated from the ratio of basis weight (BW) and t (Eq. 4),

$$\rho_\mathrm{f} = \frac{BW}{t} \qquad (4)$$

where BW is the weight of a unit area, typically measured in g/m^2, while t is often measured in µm (Hutten 2016). Filter media used for disposable applications are produced at a low basis weight, *e.g*., approximately 25 g/m^2 in N95 respirators and medical face masks (Zhao *et al*. 2020).

The bulk (β_f) is also used to characterize the filter and corresponds to the inverse of the density, expressed in cm^3/g (Eq. 5) (Hutten 2016),

$$\beta_\mathrm{f} = \frac{1}{\rho_\mathrm{f}} = \frac{t}{BW} \qquad (5)$$

The porosity (ε) of the filter can be calculated from the basis weight, thickness, and fiber density (ρ_fiber), as shown in Eq. 6 (Long *et al*. 2018). The pore properties (size and distribution) are affected by fiber type (synthetic or natural), fiber dimensions (micro- or nano-scale), and, particularly for pulp fiber sheets, drying conditions to remove excess water from wet-formed sheets (Sim and Youn 2016).

$$\varepsilon = 1 - \frac{BW}{\rho_\mathrm{fiber} t} \qquad (6)$$

The strength properties of filter media include in-plane and out-of-plane properties. In-plane properties comprise tensile properties (tensile strength, stretch or elongation, tensile energy absorption, and tensile stiffness), while out-of-plane properties include bending stiffness, burst strength, internal bond strength, and Z-direction compression. Other properties of filter media are also important. For example, water resistance is important principally for moisture-absorbing materials such as cellulose fibers, as dimensional changes due to moisture absorption affect the integrity of the filter and cause pore closure, increasing flow resistance and decreasing filtration performance (Hutten 2016).

OVERVIEW OF CELLULOSE AND NANOCELLULOSE FOR MASK PRODUCTION

Many filtration technologies and new materials used in face masks have been studied since the occurrence of the COVID-19 pandemic. Among the most important filtration technologies are nanotechnology, membrane technology, and composite and impregnated media structures, as well as adsorbent/absorbent, electrostatic, and antimicrobial media (Hutten 2016). Cellulose has great potential as one of the most versatile and renewable materials suitable for such applications, either through the use of nanocellulose or through the design of functional composite structures, as discussed in this section.

Wood Pulp Fibers

Wood pulp fibers are biosynthesized composites made of spirally wound cellulose microfibrils, which are formed from cellulose chains made of several glucose molecules and assembled through hydrogen bonds and van der Waals intermolecular interactions (Tavakolian *et al*. 2020). Wood-derived cellulose is composed of crystalline (in a high portion of approximately 65%) and amorphous regions, both with accessible and non-accessible parts for moisture sorption, pulping, and chemical modifications. The accessible cellulose comprises the surfaces of crystalline cellulose and almost the entire amorphous regions (Rowell *et al*. 2005). Wood pulp fibers differ from synthetic fibers usually used in protective masks in several ways. Firstly, pulp fibers have irregular shapes and a hollow cellular structure in the cross-section, while synthetic fibers have a regular and solid rounded structure (Pan *et al*. 2019). Additionally, thermoplastic synthetic fibers are hydrophobic, while cellulose fibers are hydrophilic. Masks containing cellulose or paper are not compatible with certain vaporization and sterilization procedures used to disinfect single-use N95 respirators in case of shortage (Bernier *et al*. 2020). Therefore, cellulose-based masks must be designed for single-use and provide antimicrobial and hydrophobic properties, as discussed in a later section.

The main attributes of wood pulp fibers, such as length, coarseness, strength, and uniformity, are largely dependent on the single-fiber characteristics of the raw material (length, diameter, wall thickness, and microfibrillar angle (MFA)) (Watson and Bradley 2009), which vary considerably between wood species. These characteristics also vary at different scales between trees, between annual growth rings of a tree, and within individual growth rings (Schweingruber 2007). Moreover, site, silvicultural treatments, and several other factors affecting environmental conditions can affect the characteristics of the raw material (Macdonald and Hubert 2002). In general, softwood pulp fibers are typically longer (3.5 mm to 5.0 mm) and coarser (40 μm in diameter), with an aspect ratio of approximately 100, whereas hardwood pulp fibers are generally 1 mm to 2 mm length and 22 μm in diameter, with an aspect ratio of 90. Both pulp fiber types are important for the filter media because softwood pulps provide bulk, strength, and permeability, whereas hardwood pulps provide filtration efficiency (Hutten 2016). Fiber coarseness is the relative mass of the cell wall or weight in mg of fiber per unit length, which can be measured using an optical analyzer. Lower fiber coarseness provides higher sheet tensile strength, greater bonding area, and more fiber per ton of pulp (Watson and Bradley 2009). The angle of microfibrils in the S_2 layer of the cell wall also varies at different scales within and between trees (Jordan *et al*. 2005; Auty *et al*. 2018). This property of the cell wall may also affect the mechanical properties of paper sheets, such as tensile strength, stretch, modulus of elasticity, and stiffness. A study with loblolly pine kraft pulp showed that fibers with high MFA are flexible and soft, and they have high stretch and a low elastic modulus, whereas those with low MFA have low stretch and a high elastic modulus (Courchene *et al*. 2006).

Wood pulps are obtained by a chemical, mechanical, or semi-chemical pulping process. The degree of purity (measured by the α-cellulose content) and contamination (lignin) of cellulosic fibers, which vary depending on the pulping process, affect the bulk and permeability of filter media. High-purity pulp has weak bonding and low strength properties, but it provides high bulk and permeability, two important properties for filter media that are directly affected by the level of purity of the wood pulp (Hutten 2016).

Chemical pulping includes the alkaline kraft, the acidic sulfite, and the organosolv processes. The kraft process uses an aqueous mixture of sodium hydroxide (NaOH) and sodium sulfide (Na_2S) (white liquor) to break down the molecules of lignin under elevated temperature and pressure in a digester and separate strong cellulose fibers (pulp) (Bajpai 2017a). Kraft pulp is preferred for filter media because of its strength properties (Hutten 2016), which are particularly important for N95 respirators and medical face masks to maintain structural integrity of the material during use. The sulfite process uses sulfurous acid (H_2SO_3) and bisulfite (HSO_3^-) ions to solubilize the lignin; the pulp mechanical properties from this process are inferior to those of the kraft process (Bajpai 2017a). A special grade of chemical pulp, named “dissolving pulp” or “dissolving cellulose,” can be obtained from the kraft or sulfite process with an acid pre-hydrolysis step to remove hemicelluloses; this pulp has a high chemical purity and is used in the manufacture of regenerated cellulose or rayon to form textile fibers like viscose or lyocell (Polymer Properties Database 2015; Chen *et al*. 2016a). The organosolv processes use organic solvents such as aromatic alcohols (phenols) or aliphatic alcohols (*e.g*., ethylene glycol, methanol, ethanol, butanol, or glycerol) and an acid catalyst (*e.g*., hydrochloric acid (HCl) or sulfuric acid (H_2SO_4)) generally at temperatures less than 185 °C, although higher temperatures may also be used depending on the process and the raw material; these processes provide access to higher-grade lignin than that of the kraft process or sulfite process while still obtaining relatively pure cellulose fibers as the main product (Yoya and Stevanovic 2018). Additionally, a new organosolv process using a Lewis acid catalyst has been developed and patented recently, and it permits more efficient recovery of high-purity lignin applicable in many industrial domains (Stevanovic and Koumba Yoya 2019). This new approach enables better use of lignin as a by-product of the pulping process.

Mechanical pulping consists entirely (or for the most part) of a mechanical process using mechanical force, combined with pressure and temperature, to separate the wood fibers. In some cases, chemicals may be added to the process (chemical-thermomechanical pulp). Mechanical pulping includes pressurized groundwood and thermomechanical pulp processes, which generate fibers and paper with lower strength compared to chemical pulps. Mechanical pulps have lower purity and more contaminants than chemical pulp, and they are therefore less suitable for N95 respirators and face masks. Fluff pulp, also known as fluffing or comminution pulp, is made by a chemical, mechanical, or combined chemical-mechanical process, usually bleached, and used as an absorbent medium in disposable diapers, bed pads, and personal hygiene products (American Forest & Paper Association 2019). Fluff pulp has high porosity, and the hydrophilic properties of cellulose make it a good absorbent material (Rom *et al*. 2007). Fluff pulp also gives bulk to filter media and is an important fiber raw material for wet-laid cellulose filter media. Mercerized pulp, which is obtained by a post-treatment with caustic soda (alkaline solution) of fully bleached pulp (usually southern bleached softwood kraft (SBSK) pulp), has very high purity (more than 99% α-cellulose) and can confer high bulk and high porosity (Hutten 2016) in filter media and nonwoven applications. Because the purity of the pulp is a crucial characteristic for filtration purposes, bleached chemical pulp is generally preferred for the manufacture of N95 respirators and medical face masks, as the bleaching process eliminates all impurities from the pulp.

There are few studies related to the use of cellulose-based filters in protective masks. However, research has shown that filters produced from wet beating of northern bleached softwood (NBSK) and hardwood (NBHK) kraft pulps followed by partial freeze-drying present filtration efficiency and pressure drop values close to those typical of N95 respirators (Mao *et al*. 2008). The authors produced single-layer filters from wood pulp by two forming methods (dry and wet forming), as well as three-layer filters composed of NBSK pulp in the middle layer and a commercial fluff pulp in the outer and inner layers. Wet-formed filters produced from NBHK pulp had the best performance. In this case, *Q* values were close to those of commercial NIOSH-certified N95 filters at the same face velocity (flow rate) of 12 cm/s (corresponding in practice to the breathing that occurs during a heavy workload). Their results also showed that greater beating provided higher capture efficiency and a greater pressure drop to the filters, due to greater surface area and smaller fibril diameter. For a given beating energy, a greater basis weight also provided increased capture efficiency and pressure drop (Mao *et al*. 2008). In short, these results showed that fibrillated cellulose provides the desired filtration efficiency but reduces air permeability, showing that particular attention should be paid to controlling air permeability in the manufacture of cellulose-based masks. In this way, mixed softwood and hardwood kraft pulps could provide a favorable compromise between filtration efficacy and air permeability. Another study proposed a method to form a spider-web-like structure in fibrillated-cellulose-based air filters to improve filtration performance (Lu *et al*. 2018). The fibrillated cellulose was prepared from a suspension of bleached softwood kraft pulp fibers. The spider-web-like structure was obtained from a fibrillated cellulose / water / tert-butyl alcohol (TBA) suspension followed by freeze-drying. The spider-web-like cellulose-based filter exhibited a filtration efficiency greater than 99% for the MPPS, regardless of the TBA content (ranging from 0% to 40%), while pressure drop increased with increasing TBA content. The mechanical properties of the cellulose-based filters were reduced with increasing TBA, which is explained by the weakness of the intermolecular hydrogen bonding interactions between adjacent microfibrils with increased TBA. These results highlight the high filtration potential of microfibrillated cellulose by the construction of spider-web-like structures in cellulose-based filters.

A recent study has compared meltblown polypropylene, as found in commercial N95 respirators and medical face masks, to various paper-based products (paper towel, tissue paper, and copy paper), among other materials (Zhao *et al*. 2020). That study showed that the electret meltblown polypropylene of N95 respirators had a 9.0 Pa pressure drop for 95.94% initial filtration efficiency and a *Q* of 162.7 kPa^{-1}, whereas non-electret meltblown polypropylene of medical face masks had 34 Pa and 16 Pa pressure drops for 33% and 18% filtration efficiencies and *Q* factors of 5.0 kPa^{-1} and 5.5 kPa^{-1}. Compared to medical face masks, paper towel and tissue paper showed moderate filtration performances of 10.4% and 20.2% for similar pressure drops and *Q* values of 4.3 kPa^{-1} and 5.1 kPa^{-1}, respectively. Meanwhile, copy paper had a high filtration efficiency but a very high and unsuitable pressure drop caused by higher bulk density and low porosity. The authors stated that paper towels and tissue paper could be inserted into cloth masks as a filter to increase their effectiveness; however, they have the disadvantage of low mechanical strength. These results serve as a reference for future work on paper-based filters.

Wood-derived Nanocellulose

Nanocellulose technology has great potential for filtering purposes, as it provides materials not only high mechanical filtration efficiency but also the possibility of functionalization into smart materials. Filters composed only of cellulose pulp fibers form relatively large pores due to their micro-scale dimensions in width. However, cellulose nanostructures, which are obtained from pulp fibers by mechanical or chemical treatments, allow better control of pore characteristics, so the filtration efficiency and mechanical properties of filters can be improved. The morphological characteristics and properties of nanocellulose, such as its high specific surface, can improve absorption and capture of small particles because of the small pore size (Sim and Youn 2016), and thus filter thickness can be reduced to allow better breathability (Chua *et al*. 2020). Moreover, nanocellulose provides superior functionalities (*e.g*., antimicrobial properties) to the filter media in comparison with cellulose pulp and other micro-scale polymers (Bajpai 2017b). Nanocellulose from wood pulp also has demonstrated biocompatibility and non-toxic properties for use in materials that are in contact with human skin (Alexandrescu *et al*. 2013).

Nanocellulose is often used as a common term for isolated cellulosic materials with at least one dimension in the nanometer range. It can be obtained from lignocellulosic fibers but also from algae, tunicates, and bacteria (Khalil *et al*. 2014). Nanocelluloses obtained from wood pulp fibers are generally grouped into either cellulose nanocrystals (CNCs) or cellulose nanofibrils (CNFs). However, a new generation of nanocellulose materials has emerged in recent years, which includes hairy nanocellulose (HNC), a new type of CNC (van de Ven and Sheikhi 2016; Tavakolian *et al*. 2020), and cellulose filaments (CFs), a fibrillated material with unique morphological characteristics (Hamad *et al*. 2019).

Cellulose nanocrystals are biodegradable rigid nanorods composed only of the crystalline parts of the cellulose, ranging in size from 2 nm to 20 nm in diameter and 100 nm to 600 nm in length (Beck-Candanedo *et al*. 2005; Tuukkanen and Rajala 2018), having a low aspect ratio (< 100) (Bharimalla *et al*. 2015). Cellulose nanocrystals have strong mechanical properties (tensile strength of 7500 MPa and Young's modulus of 100 GPa to 140 GPa) due to high intermolecular bonding, very high specific surface (150 m^2/g to 250 m^2/g), and high heat stability (low thermal expansion coefficient), which makes them suitable for tissue engineering and cargo carrying (Tavakolian *et al*. 2020). Due to these properties, CNCs also have high potential versatility in terms of functionalization and modification (Trache *et al*. 2020). Cellulose nanocrystals are commonly obtained by concentrated acid hydrolysis using sulfuric acid to react with wood pulp under controlled conditions of acid-to-pulp ratio and temperature (Hamad *et al*. 2019), although other means of production can also be used (Miller 2018). Cellulose nanocrystals are used in various forms, *e.g*., powder, gel, or film (membrane), usually to make lighter and stronger materials with greater durability in a wide range of applications, including paints, packaging, cosmetic bases, pigments, food modifiers, sensors, biomedical sciences, and composites (Tavakolian *et al*. 2020). Regarding filter applications, CNCs have a demonstrated capacity to improve filtration efficiency with a low pressure drop in poly(vinyl alcohol) (PVA) / CNC composite nanofibrous filters produced by electrospinning for HEPA filters (Zhang *et al*. 2019a). The main sources of CNCs are wood pulp, cotton, hemp, flax, wheat straw, rice straw, mulberry bark, ramie, tunicin, algae, and bacteria, among others (Klemm *et al*. 2011).

Hairy nanocellulose is a new class of nanocellulose, formed by nanorods made of crystalline parts and also amorphous chains ("hairs") protruding from both ends, which provides high colloidal stability and unique physicochemical properties, resulting in a wider range of applications than conventional CNCs (van de Ven and Sheikhi 2016). Hairy nanocelluloses are synthesized by oxidizing, solubilizing, and cleaving the amorphous regions of cellulose fibrils (Tavakolian *et al*. 2020). There are different types of HNCs with neutral, negative, and positive charge contents to be chosen for various potential applications (Tavakolian *et al*. 2020), such as transparent films (stronger and more flexible than those made using conventional CNCs), superhydrophobic films (*e.g*., crosslinked with a diamine), security packaging (to manufacture smart coatings and packaging based on green nanomaterials), heavy metal ion scavengers, scavengers of non-ionic pollutants, cellulose hydrogels (hydrophilic polymer networks), humidity switches, polymer reinforcement, rheology modifiers, flocculants, directed metal nanoparticle synthesis, and biomimetic mineralization (van de Ven and Sheikhi 2016).

Cellulose nanofibrils are long slender rod-like particles consisting of alternating crystalline and amorphous cellulose regions. According to a Technical Association of the Pulp and Paper Industry (TAPPI) report, the terms CNF (or nanofibrillated cellulose, NFC) and microfibrillated cellulose (MFC) are used interchangeably, with an overlap in the diameter specifications; while some of these materials contain only particles smaller than 100 nm in diameter, others contain a mixture of particles ranging from the nanoscale to the microscale (Miller 2018). Cellulose nanofibrils are produced from cellulose pulp fibers using an energy-intensive mechanical treatment, although they are usually chemically or enzymatically pretreated to reduce energy consumption. The most commonly reported pre-treatment in scientific studies is a 2,2,6,6-tetramethylpiperidine-1-oxyl (TEMPO)-mediated oxidation of cellulose fibers, which oxidizes the primary hydroxyl groups at the C6 position of the glucose units and converts them to carboxyl groups (uronic acid) (van de Ven and Sheikhi 2016). Cellulose nanofibrils have a high aspect ratio (> 100) (Bharimalla *et al*. 2015) and a high specific surface area, with regions of hydroxylated surface chemistry for possible chemical modification (Klemm *et al*. 2011) interspersed with the crystalline phase (Dimic-Misic *et al*. 2018). However, the degree of surface chemistry and fibril dimensions depend on the CNF source or origin, pre-treatment, and fibrillation procedures (Alexandrescu *et al*. 2013). The main sources of CNFs are wood pulps, sugar beet, potato tuber, hemp, and flax fibers (Klemm *et al*. 2011).

Cellulose nanofibrils and CFs are both fibrillated materials; however, CFs are morphologically different from CNFs. Cellulose filaments are longer and are heterogeneous, comprising cellulose fibrils and fiber fragments (Hamad *et al*. 2019), and they have a higher aspect ratio (greater than 1000) than that of CNFs (Miller 2018). Cellulose filaments can be produced from all types of wood pulps, bleached or unbleached, using a mechanical refining method. Kruger Biomaterials Inc. produces CFs in its commercial plant in Trois-Rivières, Quebec, Canada, using a proprietary technology licensed from FPInnovations Inc., with an annual production capacity of up to 6,000 t of a product traded under the name FiloCell®. This product is used as a reinforcement additive for various paper and tissue grades to improve wet and dry strength, consistency of mechanical strength, and filler retention (Kruger Biomaterials 2018). FiloCell® CFs from Kruger are produced from kraft wood pulp without further application of chemicals or enzymes, with dimensions ranging from 0.1 mm to 2.0 mm in length, 80 nm to 300 nm in width, and 40 nm to 100 nm in thickness (a single kraft pulp fiber produces approximately 1000 CFs). Other characteristics of FiloCell® CFs include a high specific surface of 80 m^2/g, a density of 1.45 g/cm^3, and purity; and they are biodegradable, compostable, thixotropic in solution, lignin-free, and available in several forms (wet pulp, dry and dispersed fluff, water suspension, *etc*.) (Kruger Biomaterials 2018). Cellulose filaments have physical and chemical advantages over kraft pulp, which is often used as reinforcement for weaker pulps. Cellulose filaments have a higher aspect ratio than softwood kraft pulp and more hydroxyl groups on the surface for the same weight of material, leading to improved mechanical properties in many materials, such as polymer composites, cement and concrete products, and paper and tissue paper products (Fairbank 2020).

For filter applications, wood-based nanocellulose materials have been used, for example, in the development of mixed wood pulp / CNF filters (Alexandrescu *et al*. 2016; Sim and Youn 2016), wood pulp / PET composite filters for air filtration (Long *et al*. 2018), and ultrafiltration membranes (Wang *et al*. 2013, 2019) using adsorption mechanisms to remove bacteria, viruses, and heavy metal ions from water (Wang *et al*. 2013). Sim and Youn (2016) investigated the structures and properties of porous sheets produced from pulp fiber (hardwood and softwood bleached kraft pulp) or PET mixed with CNFs. The mixing ratio of CNFs ranged from 0% to 50% at intervals of 10% for each fiber type. Cellulose nanofibrils were prepared from a commercial hardwood bleached kraft pulp and had an average width of 38 nm. Sheets were formed by a wet-laid forming method and dried by different methods (cylinder or freeze-drying). All sheets had a similar basis weight of 36 $g/m^2 \pm 1 g/m^2$ and densities ranging from 0.21 g/cm^3 to 0.58 g/cm^3, depending on the fiber mix, CNF ratio, and drying conditions. The addition of CNFs improved considerably the mechanical properties (tensile strength and elastic modulus) of the porous sheets for all fiber combinations. Porous sheets formed from a combination of hardwood bleached kraft pulp fibers and CNFs showed a high filtration efficiency of 99.94% but also a high pressure drop. The air permeability of the porous sheets decreased as the amount of CNFs increased. However, the application of a freeze-drying method increased porosity and pore size and improved the air permeability of the sheets. The same study also reports the effects of fiber morphology on the apparent density, porosity, and thickness of the filters and, consequently, on their filtration efficiency, pressure drop, and air permeability properties. The PET fibers formed bulkier sheets, while pulp fibers formed denser sheets due to their dimensions and internal structure (solid or cellular). Synthetic fibers have a solid, filled structure, while pulp fibers have a tubular structure with cellular lumens, resulting in greater compactness and density and, therefore, less bulk and porosity than PET fibers. Alexandrescu *et al*. (2016) used CNFs from bleached eucalypt kraft pulp to form CNF-based filters. Cellulose nanofibrils were prepared by the mechanical fibrillation method, with and without TEMPO-mediated oxidation before fibrillation. In the filter formation, CNFs are dispersed using water, but then water removal is one of the challenges for the production of CNF filters because fibrils tend to clump together during drying as they form hydrogen bonds, leading to compact structures, a process known as cellulose hornification. To solve this issue, a freeze-drying process was applied. The TEMPO-treated CNF filter had the best performance ($Q = 0.025 Pa^{-1}$) compared to the untreated CNF filter ($Q = 0.015 Pa^{-1}$). The filtration efficiency (46%) and pressure drop (25 Pa at 5.3 cm/s face velocity) of the TEMPO-treated CNF filters were close to those found by Zhao *et al*. (2020) for meltblown polypropylene found in medical face masks. Long *et al*. (2018) studied the effects of different proportions of lyocell MFC, ranging from 0% to 20% in 5% intervals, on the filtration efficiency and pressure drop of softwood pulp / PET composite filters. Softwood pulp fibers varied from 55% to 75%, while PET fibers had a constant proportion of 25% in the filter composition. All composite filters had a basis weight of 105 $g/m^2 \pm 2 g/m^2$, but filter thickness and porosity decreased slightly as the fraction of MFC increased. Both the filtration efficiency for particles ranging from 30 nm to 2 μm in size and the pressure drop increased with the increase in the fraction of MFC. The authors reported that the optimal fraction of MFC was to 5%, as larger fractions tented to decrease the uniformity and Q factor of the composite filters. For the application of CFs in filter media, an FPInnovations patent recommends a proportion of up to 10% (Drolet *et al*. 2017); however, further research is needed to assess the potential of CFs to improve the properties of N95 respirators or face masks.

ANTIMICROBIAL TECHNOLOGIES APPLICABLE TO CELLULOSE FIBERS

Antimicrobial agents are used to provide biocidal (pathogen elimination) or biostatic (pathogen growth inhibition) activities to many materials. Such properties are important for protective masks because they protect the wearer against harmful bioaerosols, in addition to offering greater comfort to the skin, as the prolonged use of a face mask can cause skin irritation. Additionally, by increasing the temperature and humidity between the mask and the skin, prolonged use creates an environment favorable to the proliferation of fungi and bacteria. As cellulose is a hydrophilic and biodegradable material, cellulose-based masks require antimicrobial treatments, preferably with antiviral properties, given the current context of the COVID-19 pandemic.

Cellulose surfaces, having multiple carboxylic acid groups, have an intrinsic negative charge, which offers the opportunity to perform cationic modifications by adding positively charged materials for antimicrobial functionalization (Alavi 2019). There are many natural, synthetic, organic, and inorganic antimicrobial agents with an affinity for cellulose that can potentially be used in protective masks. These include natural bioactive compounds (*e.g.*, polyphenols, terpenoids, organic acids, and polysaccharides), metal-containing materials, inorganic salts (*e.g.*, quaternary ammonium compounds (QACs)), synthetic polymers (*e.g.*, N-halamines and polyhexamethylene biguanide (PHMB)) (Ristić *et al*. 2011; Timofeeva and Kleshcheva 2011), and composites thereof, including MOFs, which are regarded as the most innovative today (Chua *et al*. 2020; Nie *et al*. 2020a). As highlighted in this section, several of them have antiviral properties, whereas other nanomaterials such as graphene can act as an enhancing agent for other well-known antiviral agents, *e.g.*, silver nanoparticles (Ag NPs).

Natural Products

Polyphenols and terpenoids

Many natural compounds have intrinsic antiviral properties, with proven efficacy against some types of coronaviruses, but they are not yet proven against SARS-CoV-2. An extended list of plant-based products and their mechanisms of action against a large variety of viruses is presented by Lin *et al*. (2014). Particularly for the coronavirus family, the authors highlight antiviral compounds such as phenolic compounds of *Isatis indigotica*, amentoflavone (a biflavonoid isolated from *Torreya nucifera*), myricetin, scutellarein, *Houttuynia cordata* water extract, and active compounds from *Lycoris radiata* (lycorine), *Artemisia annua* (artemisinin), *Pyrrosia lingua*, and *Lindera aggregata*. They also note antiviral activity of saikosaponins (A, B_2, C, and D), which are triterpene glycosides isolated from medicinal plants, against human coronavirus 229E (HCoV-229E). These various compounds inhibit viruses using different mechanisms of action. However, more research is needed to assess the effects of these plant-derived compounds on human health, their economic viability, and safety criteria for their use in protective devices, as the active compounds can vary from one plant to another, depending on local growth, soil conditions, climate, *etc*. Studies have also reported the antiviral activities of curcumin, found in the rhizome of *Curcuma longa*, a plant from the ginger family commonly named turmeric (Moghadamtousi *et al*. 2014), and catechin polyphenols (Catel-Ferreira *et al*. 2015). For instance, the antiviral properties of catechin polyphenols grafted on a cellulose-based filter for masks was studied by Catel-Ferreira *et al*. (2015). The authors recomposed masks by replacing the cellulose layer of a commercial Kolmi M24001 medical face mask with one or two layers of a catechin-grafted cellulose filter produced using laccase as an enzymatic coupling reagent. The antiviral activities of the filters were tested in liquid media of a viral concentration (T4D bacteriophage virus of *Escherichia coli* B), and face masks were compared for filtration efficiency. The catechin grafted-cellulose filters showed a great effect on the viral concentration, while a remarkable increase in virus filtration with a moderate airflow resistance was obtained for recomposed masks compared to the original masks. Another study assessed the antifungal properties of a turmeric-based nanomaterial coating on cellulosic cotton fibers (Gargoubi *et al*. 2020). In this case, cellulose fibers were coated with poly(dopamine), a nanomaterial produced from the final oxidation of dopamine or other catecholamines (Ball 2018), which has biocompatible molecules to enhance the uptake and attachment of bioactive turmeric. The poly(dopamine) interface forms a spider-web-like layer to establish bridges between cellulose fibers and antifungal molecules. The results showed that the turmeric/poly(dopamine)/cellulose composite material inhibited fungal growth, while the morphological characteristics and mechanical properties of the cellulose fibers were preserved (Gargoubi *et al*. 2020). The composite material could have a potential use in protective face masks to prevent fungal skin infections caused by prolonged mask wear (Hamann 2020).

Organic acids

Some commercial N95 respirators and medical face masks present an acid-based medium as an antiviral coating. These masks have an antimicrobial outer layer made of polyester or regenerated cellulose fibers treated with citric acid, sometimes combined with other antiviral agents such as Cu-Zn ions (Davison 2012; RespoKare 2017; Chua *et al*. 2020). Citric acid is also used as a cross-linking agent, to reduce the water retention properties of cellulose fibers in paper and textiles (Rom *et al*. 2007).

Polysaccharides

Several studies have reported the antimicrobial properties of chitosan, a modified natural carbohydrate derived from chitin found in the exoskeletons of crustaceans, mussels, fungi, insects, and some algae (Ristić *et al*. 2011). The antimicrobial activity of chitosan is attributed to positively charged amino groups ($-NH_3^+$) that form polycationic compounds in acid media capable of interacting with negatively charged groups of lipopolysaccharides and proteins on the cell surfaces of microorganisms, causing membrane disintegration and damage to the cell wall. However, the antimicrobial capacity of amino chitosan depends on its degree of deacetylation (ranging from 60% to 95%), molecular weight, and the ratio between protonated and deprotonated amino groups (Kucharska *et al*. 2020). Like cellulose, chitosan is one of the most abundant biopolymers on Earth (Ristić *et al*. 2011). Chitosan is very popular today because it is eco-friendly, renewable, biodegradable, non-toxic, and biocompatible, and further functionalization allows for its use in a wide range of applications (Hahn *et al*. 2020), including biomaterials and healthcare materials (Vaz *et al*. 2014). Moreover, chitosan and chitosan derivates can be applied as carriers and linkers of flame-retardant agents and superhydrophobic coatings (Hahn *et al*. 2020), contributing to two important properties of medical face masks for meeting regulatory requirements. Three-layer non-medical masks, marketed under the name M-Chitosan, composed of an inner antibacterial layer of chitosan are already available on the market (M-Chitosan 2020). The antiviral properties of chitosan nanocomposites, *e.g.*, chitosan nanoparticles / curcumin (Loutfy *et al*. 2020) and Ag NPs / chitosan (Ling *et al*. 2013; Mori *et al*. 2013), and functionalized chitosan (Milewska *et al*. 2013) have been also investigated. The study conducted by Mori *et al*. (2013) revealed no antiviral activity of chitosan against the H1N1 influenza A virus; however, the chitosan matrix had the advantage of preventing the release of Ag NPs, a well-known antiviral agent, from Ag NP / chitosan composites. Meanwhile, Loutfy *et al*. (2020) showed that chitosan nanoparticles can increase the antiviral effects of curcumin in chitosan/curcumin nanocomposites. Additionally, a cationic chitosan derivate (N-(2-hydroxypropyl)-3-trimethylammonium chitosan chloride, HTCC) showed antiviral activity against human coronavirus NL63 (Milewska *et al*. 2013). For paper materials, chitosan derivates, *e.g*., quaternary carboxymethyl chitosan (QCMC), prepared by grafting carboxymethyl groups and quaternary ammonium groups to chitosan chains, have been used as antibacterial agents (Ling *et al*. 2013). Ahmad *et al*. (2016) used chitosan to enhance the affinity of cellulose fibers for Ag ions to produce functional cellulose filter papers. Due to their similar structures, chitosan and cellulose form hydrogen bonds (He *et al*. 2018). Thus, chitosan provides strong bonding and increased mechanical properties of paper materials (Kucharska *et al*. 2020). Ling *et al*. (2013) prepared Ag NP-loaded QCMC / organic montmorillonite (QAOM) nanocomposite coatings for paper surfaces. In addition to greater antimicrobial capacity, QAOM-coated paper exhibited higher tensile, shear, and bursting strengths than uncoated paper. For cellulose-based mask applications, chitosan has often been used in combination with other antiviral technologies to form nanocomposite materials, *e.g*., Ag NPs / chitosan (Imani *et al*. 2011) and MOFs/chitosan (Nie *et al*. 2020a), as presented throughout this section.

Synthetic and/or Inorganic Products

Metal-containing materials

Some well-known and widely documented antiviral technologies have gained considerable uptake since the occurrence of the COVID-19 pandemic. These include metals and metal compounds of silver (Ag), copper (Cu), and zinc (Zn) used in protective masks, principally for reusable cloth masks made of cotton or synthetic fibers. Many metals and metal compounds (*e.g*., metal salts and metal oxides) have been shown to be effective antimicrobial agents with antibacterial (Imani *et al*. 2011; Emam 2019) and/or antiviral properties (Bright *et al*. 2009; Borkow *et al*. 2010; Galdiero *et al*. 2011; Zhou *et al*. 2020). Bright *et al*. (2009) found that Ag and Ag/Cu zeolites inactivate enveloped viruses such as coronaviruses by charge-based interactions with their outer lipid envelope membranes. An FDA-approved antiviral N95 respirator is currently commercialized by Nexera Medical SpectraShield^{TM} using the Ag/Cu zeolite technology. Zeolite is a carrier that controls the release of metal ions (Nexera Medical 2020). Copper oxide particles can also be impregnated into N95 respirators without altering their filtration properties, a solution with demonstrated efficacy against the influenza virus (Borkow *et al*. 2010).

The antiviral properties of Ag NPs against the H1N1 influenza A virus have also been demonstrated; however, the antiviral activity depends on the concentration and size of the nanoparticles. The antiviral effect of Ag NPs was stronger with smaller nanoparticles in the Ag NP / chitosan composites, and antiviral activity increased as the concentration of Ag NPs increased (Mori *et al*. 2013). Since 2012, Biofriend™ Biomask™ has been offering disposable antiviral surgical N95 respirators and medical face masks comprising a cellulose/polyester layer with positively charged Cu/Zn ions (Davison 2012). Many companies provide metal-based antimicrobial technologies, mainly Ag, because of its non-toxicity, eco-friendliness, high thermal stability, and antiviral properties; however, like for Cu and Zn, the high cost of Ag makes the mask-making process more costly. Dupont Silvadur™ technology uses Ag ions to neutralize microorganisms in textiles that are currently used in non-medical cloth (cotton) masks for the general public.

The antibacterial properties of Ag NPs capped with polyacrylic acid (PAA) combined with chitosan have been investigated in wood pulp filters (Imani *et al*. 2011). The authors used a layer-by-layer technique to impregnate filters made of bleached softwood kraft pulp fibers with Ag NPs / PAA / chitosan and showed that, in addition to being efficient, the antibacterial layer did not affect the tensile strength or pore structure of the filter.

MOFs

Metal-organic frameworks are defined as organic-inorganic hybrid crystalline porous materials composed of a regular array of positively charged metal ions or clusters surrounded by organic ligands (Berger 2020). They are a new class of materials with key advantages compared to other metal compounds (Chua *et al*. 2020). The metal ions construct nodes that assemble the organic ligands *via* a molecular building block approach to form a hollow frame structure with an ultra-high surface area. Many structures of MOFs can be synthesized using different metal ions and organic ligands, with numerous applications (Berger 2020). For instance, zeolitic imidazolate framework-8 (ZIF-8), copper(II)-benzene-1,3,5-tricarboxylate (Cu-BTC) MOF (MOF-199), and Ag-based MOFs (Ag-MOFs) are used for antibacterial applications (Ma *et al*. 2018). A recent study investigated the potential of MOF-paper materials as antibacterial composites for air filtration (Nie *et al*. 2020a). The authors used ZIF-8, a three-dimensional framework designed with zinc nitrate hexahydrate ($Zn(NO_3)_2 \cdot 6H2O$) ions and 2-methylimidazole (2-H-MeIM, $C_4H_6N_2$), combined with chitosan gel and mixed with softwood kraft pulp to produce ZIF-8/chitosan/cellulose composite paper filters. Chitosan prevented MOF agglomeration and enhanced the compatibility between ZIF-8 and the cellulose fibers. The ZIF-8/chitosan addition did not affect the tensile index and provided antibacterial properties to the composite paper, but it decreased the evenness index used to evaluate the quality of formation and the uniformity of the paper sheets. The ZIF-8/chitosan/cellulose composite filters of 60 g/m^2 basis weight had 99.68% filtration efficiency for removing particles ≤ 2.5 µm in diameter, compared to a 94% efficiency for untreated paper. The composite paper had an ultra-high specific surface of 139 m^2/g, compared to 0.77 m^2/g for untreated paper. Additionally, the air permeability of the composite papers increased by more than 50% compared to the untreated papers (Nie *et al*. 2020a). These results are encouraging, as filtration and breathability are generally two opposing properties of mask filters, especially for cellulose-based filters. Moreover, Nie *et al*. (2020a,b) state that MOF/chitosan gel can be added directly to the wood pulp and therefore would not require a change in paper manufacturing. Studies have also reported the use of MOFs to produce multifunctional cellulose-based filters with ultra-high filtration efficiency, adsorption properties, and antibacterial properties for the healthcare industry (Ma *et al*. 2018). Ma *et al*. (2018) produced biodegradable MOF/cellulose filters using ZIF-8, MOF-199, and Ag-MOFs by *in-situ* deposition on the cellulose matrix. The MOF/cellulose filters were compared to pure cellulose filters, all made of fibrillated softwood kraft pulp. The ZIF-8 was deposited directly to the pulp suspension diluted in distilled water, reacted in an autoclave at 75 °C for 24 h, and freeze-dried to remove the residual solvent. Similar methods were used for the other MOFs, except that MOF-199 was deposited upon the pulp suspension diluted in composite solvents of N,N-dimethylformamide, ethanol, and water (1:1:1) and reacted at 85 °C for 24 h, while Ag-MOFs were reacted at 120 °C for 72 h. The ZIF-8/cellulose, MOF-199/cellulose, and Ag-MOF/cellulose filters had filtration efficiencies of 98.36%, 98.28%, and 97.34% against 0.3-µm particles, respectively, while pure cellulose filters had only approximately 44% efficiency. However, unlike ZIF-8/chitosan/cellulose composite filters (Nie *et al*. 2020a), ZIF-8/cellulose, MOF-199/cellulose, and Ag-MOF/cellulose filters had higher pressure drops (134 Pa, 131 Pa, and 126 Pa, respectively) compared to pure cellulose filters (approximately 25 Pa). The antibacterial activities followed the order Ag-MOF/cellulose filter > MOF-199/cellulose filter > ZIF-8/cellulose filter (Ma *et al*. 2018). Similar results for filtration efficiency and pressure drop of ZIF-8/cellulose filters were obtained by Su *et al*. (2018).

Quaternary ammonium and other salts

Recently, the Indian Department of Biotechnology's Institute for Stem Cell Science and Regenerative Medicine (Bangalore, India) developed an antiviral face mask composed of cotton fibers impregnated with quaternary ammonium salts, which will be commercialized under the brand name G99+ by the Color Threads company in India. Because SARS-CoV-2 is an enveloped virus composed of a lipid membrane with negatively charged ions, it is expected that quaternary ammonium salts containing positively charged nitrogen could disrupt the virus envelope through the same principle of action as Ag ions, although they have not yet been tested against SARS-CoV-2 (Sachan 2020). Quaternary ammonium compounds are among the most studied functional groups to provide antibacterial properties to cellulosic materials (Tavakolian *et al*. 2020) and other biomaterials and are commercially used in the manufacture of antimicrobial textiles (*e.g.*, cotton, polyester, and nylon) (Ristić *et al*. 2011), which make them a good candidate for antiviral cellulose-based filters. Sodium chloride (NaCl) is another inorganic salt used as an antiviral agent applicable to medical face masks. Quan *et al*. (2017) reported the use of NaCl coatings to treat the middle layer of three-ply surgical masks made of polypropylene microfiber; a surfactant was also used to enhance the wetting of saline solutions on the hydrophobic surfaces of polypropylene fibers. Their results showed that NaCl-coated filters exhibited high efficiency (approximately 85%) against aerosols containing H1N1 influenza viruses through a salt-recrystallization-based filtration system, which completely inactivated different types of adsorbed influenza viruses. Moreover, the NaCl coating exhibited stability at high temperature and humidity, suggesting safe use and long-term storage, in addition to being simpler and less expensive than other antiviral technologies. Sodium chloride coatings on surgical face masks are also currently being investigated at the University of Alberta in Canada (Bogart 2020).

N-halamines

N-halamines are a group of organic synthetic polymeric compounds and can be used as effective antimicrobial, antibacterial, and antiviral agents in protective masks (Ren *et al.* 2018; Huang *et al.* 2019). Huang *et al.* (2019) produced polyacrylonitrile (PAN)-nanofiber membranes with the addition of 5% of a type of N-halamine (1-chloro-2,2,5,5-tetramethyl-4-imidazolidinone, MC) by electrospinning for potential applications in protective masks. Their results showed that PAN/MC membranes have excellent antibacterial efficiency but a 20% decreased air permeability compared to untreated PAN membranes, although both reached breathability requirements. Ren *et al*. (2018) studied the antiviral activities of the nonwoven fabric of a commercial N95 respirator coated with MC concentrations of 0.1% to 1% and showed that higher MC concentrations considerably reduced the presence of the avian influenza virus in the filter and completely inactivated the viruses by disrupting their RNA. According to Tavakolian *et al*. (2020), the cellulose surface can be easily grafted with N-halamine due to the abundance of hydroxyl groups, thus developing durable antimicrobial properties. For instance, an N-halamine precursor (polyethylenimine, PEI) was grafted onto dialdehyde cellulose membranes, followed by chlorination, to prepare antibacterial N-halamine/cellulose membranes for wound dressing materials. The N-halamine/aldehyde-cellulose membrane completely inactivated bacterial pathogens within 5 min even after 15 d of storage and exhibited an increase of approximately 38% in elongation at break and a slight decrease in tensile strength (105 MPa) compared to the ungrafted membrane (112.8 MPa) (Zhang *et al*. 2019b). N-halamine compounds have long-term stability and low environmental impact (Huang *et al*. 2019), are safe for humans, and can be grafted onto other antimicrobial materials such as chitosan.

PHMB

Polyhexamethylene biguanide is another polymer that is widely used as a biocide in numerous applications, including wound dressing, hand washes, personal care products, finishing for textiles (mainly for cellulose-based textiles) (Ristić *et al*. 2011), face masks, medical devices, and surgical instruments (Schorr *et al*. 2007). A Kimberly-Clark patent describes an antimicrobial composition containing a mixture of PHMB hydrochloride and a synergistic coactive agent (*e.g.*, another biguanide, QACs, or natural agents, among others) that can be coated on the outer layer of face masks made of cellulose-based or synthetic polymeric nonwoven sheets, polymeric films, or a combination thereof. Polyhexamethylene biguanide hydrochloride is a cationic biguanide that attracts and disrupts the negatively charged membranes of most microorganisms, and the damage to the membrane increases with the degree of polymerization of the biguanide (Schorr *et al*. 2007). Polyhexamethylene biguanide is characterized by low toxicity and is skin-friendly, non-mutagenic, non-carcinogenic, showing oral tolerance (Timofeeva and Kleshcheva 2011), and odorless to humans, which are important characteristics for face masks (Schorr *et al*. 2007).

Carbon materials

Graphene is a carbon-based 2D nanostructured material with antibacterial and potentially antiviral properties that can also be used as a superhydrophobic agent for disposable surgical masks, as pointed out in a recent study (Zhong *et al*. 2020). The authors proposed a method for functionalizing commercial surgical masks made from polypropylene fibers with self-cleaning and photothermal properties using a porous graphene coating. Functionalized graphene masks were produced from a polyimide precursor film through a fourth-generation laser technique that was used to transfer the hydrophobic graphene layer. These masks were demonstrated to be efficient in bouncing particles and could be reused after solar sterilization, because the SARS-CoV-2 spike protein, which is used as a means of infection, is sensitive to temperature. The photothermal performance of graphene-coated *versus* pristine masks was measured by absorbance, which was maximal for graphene-coated masks while being relatively low for pristine ones, especially in the visible and UV portions of the spectrum. Variations of the surface temperatures of the masks over time were measured by an infrared camera. While the surface temperature of the classical surgical masks remained at approximately 45 °C after 5 min of solar illumination, the graphene-coated surface reached a temperature of 70 °C in 40 s and maintained a temperature of 80 °C after 100 s of illumination, a temperature sufficiently high to inactivate viruses (Zhong *et al*. 2020). Huang *et al*. (2020) report that the antibacterial efficiency of laser-induced graphene masks is improved to approximately 81% and could be increased to 99.998% within 10 min under sunlight, while in common face masks 90% of bacteria remain alive after 8 h, creating a biohazard and environmental concerns when they are improperly used or discarded. Some graphene face masks have recently appeared on the international market. United Kingdom-based PlanarTECH, for example, has developed a washable and reusable face mask coated with graphene and other carbon nanomaterials (Graphene Catalog 2020). Additionally, a three-layer disposable face mask from Transon features an antibacterial protective inner layer of biomass graphene composite (Transon 2019). Regarding cellulosic materials, a study reports the benefits of graphene oxide (GO) and Ag NPs added to dissolved cellulose to improve the physical and antibacterial properties of nanocomposite membranes (Chook *et al*. 2014). Graphene oxide caused the immobilization of Ag NPs in solution, preventing their aggregation and improving their colloidal stability, which resulted in a large specific surface area, thus increasing the antibacterial functionality of the nanocomposite membrane. Chen *et al*. (2016b) state that the most important advantage of GO / Ag NP composite is the Ag NPs' immobilization on the GO sheets, which increases material compatibility and reduces environmental impacts associated with the release of free Ag NPs. Moreover, GO / Ag NP composites have antiviral potential against enveloped and non-enveloped viruses– enveloped feline coronavirus (FCoV) and bursal disease virus (IBDV), respectively– as shown by Chen *et al*. (2016b). They found that GO / Ag NP sheets exhibited greater virus inhibition than GO sheets alone. The latter had viral inhibition against FCoV but not against non-enveloped virus.

CONCLUSIONS

The current review provides an overview of the features of commercial N95 respirators and medical face masks, the desirable characteristics of cellulose and nanocellulose materials for the manufacture of disposable face masks, and the recent advancements in antimicrobial (nano)materials to produce functional cellulose-based filters. The currently produced commercial masks, although generally made of petroleum-based synthetic fibers, offer high filtration efficiency against harmful particles, including viruses and bacteria, and low pressure drop to allow breathability and provide comfort to the wearer. However, few commercial N95 and medical face masks currently provide antiviral properties, increasing the risks of cross-contamination since the contaminated mask may become a passive transmission vector and raising environmental concerns for their disposal after use. These concerns are particularly important for SARS-CoV-2, which is a highly contagious pathogen and able to survive for longer periods on the surface of different materials. Therefore, the challenges related to the manufacture of biodegradable cellulose-based masks are to develop filters capable of providing high filtration efficiency and low pressure drop similar to those found in commercial masks, and antimicrobial functionalities to increase protection against viruses. The targeted cellulose-based filter can be designed by adjusting different blends of softwood/hardwood chemical pulps, as coarse softwood pulp fibers offer bulk, strength, and air permeability, while hardwood pulp fibers improve filtration efficiency. Additionally, the high specific surface and versatility of fibrillated cellulose materials could improve the mechanical strength and enable the capture of submicron particles by filter media, thus permitting the adjustment of filter thickness to maintain breathability. Moreover, nanocelluloses can increase the functionality of the filter and allow for the design of more efficient functional cellulose-based composite filters.

Among the principal antimicrobial technologies applied to masks and compatible with cellulose due to similarities of their structures, chitosan-based (nano)composites appear to be among the most promising. Many chitosan derivates are commercially available and can be combined with many other antiviral agents to form composite structures capable of being incorporated into papermaking processes. Chitosan is eco-friendly, renewable, biodegradable, non-toxic, biocompatible, and antibacterial. Chitosan also enhances the cellulose affinity with antimicrobial agents and acts as a matrix that increases the dispersion and functionalities of other antiviral/antibacterial agents such as Ag NPs and MOFs. Ideally, cellulose-based masks must present antimicrobial properties in the outer and inner layers to prevent pathogenic microorganisms from the environment from reaching the wearer and *vice versa*.

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