Cellulosic fiber nanocomposite application review with zinc oxide antimicrobial agent nanoparticle: an opt for COVID-19 purpose

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Abstract

Cellulosic fiber (CF) in nanoform is emergingly finding its way for COVID-19 solution for instance via nanocomposite/nanoparticle from various abundant biopolymeric waste materials, which may not be widely commercialized when the pandemic strikes recently. The possibility is wide open but needs proper collection of knowledge and research data. Thus, this article firstly reviews CF produced from various lignocellulosic or biomass feedstocks' pretreatment methods in various nanoforms or nanocomposites, also serving together with metal oxide (MeO) antimicrobial agents having certain analytical reporting. CF-MeO hybrid product can be a great option for COVID-19 antimicrobial resistant environment to be proposed considering the long-established CF and MeO laboratory investigations. Secondly, a preliminary pH investigation of 7 to 12 on zinc oxide synthesis discussing on Fouriertransform infrared spectroscopy (FTIR) functional groups and scanning electron microscope (SEM) images are also presented, justifying the knowledge requirement for future stable nanocomposite formulation. In addition to that, recent precursors suitable for zinc oxide nanoparticle synthesis with emergingly prediction to serve as COVID-19 purposes via different products, aligning with CFs or nanocellulose for industrial applications are also reviewed.

Keywords Lignocellulosic · Cellulosic fiber · Nanocellulose · Nanocomposite · Nanoparticle · Zinc oxide · COVID-19

Introduction

Cellulosic fibers (CFs) are naturally available and easily accessible via lignocellulosic or agricultural biomass which are abundantly available. Cellulose within the complex biopolymers are extracted and separated from hemicellulose and lignin via pretreatment processes. Various pretreatment methods routed from conventional methods and some

of them were commercialized for mass productions, leaving the rest being as emerging methods for further fundamental investigations with their complexities (Brodeur et al., 2011; Mankar et al., 2021; Meenakshisundaram et al., 2021; Sarker et al., 2021; Zheng & Rehmann, 2014).

From time to time, the research focus becomes very demanding and in thirst of fundamental theories and models for new products, resulting in producing instead of single entity product but now hybrid products for instance in using CF (Arunachalam et al. 2021; Bhattacharjee & Roy, 2020; Godara & Mahato, 2019; Tilak et al., 2021). CF originally from fibrillated form (Mahadi et al. 2015) evolves from certain particle size to nanosize in research, confirming the enhanced effectiveness as the size gets smaller (Miri et al., 2021). Nanotechnology is adopted in various industries including cellulosic nanoparticle as in nanocomposite fabrications, in support of various applications, for instance recently for COVID-19 purposes. The resistant-like hybrid cellulosic nano or not nanocomposite may be a useful option to cater safe environmental surroundings to human use considering as cellulose as abundant renewables. The “safe”

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intention can be collaborated with metal oxide antimicrobial agents, in an intervention with cellulosic fiber materials, suitable for COVID-19 precautionary steps for instance.

To add to these visions, these CF hybrid products should start from the improvements on CF generation, for instance via pretreatment methods up to the fabrication of nanocomposites, the hybrid, in urge need to have reliable data characterizations for scientific proofs. These CF nanoforms are not only being investigated by its light effect activity along the light spectrum ranges for instance via transmittance or absorption but also via microscopic imaging despite other tests on the mechanical strength of the raw and the new products. Upgrades replacing the conventionally generated CF from natural resources could now be produced via biological generation for instance via bacterial cellulose instead, as novel cellulosic material (Abdelraof et al., 2019; Ghozali et al., 2021).

In relation to COVID-19 purposes, hybrid nanocomposites were incorporated with metal oxides acting as antimicrobial agent, acting as alternative solutions for various applications for instance in biomedical (John Owonubi et al. 2020), food (Karim et al., 2020; Kumar et al., 2018; Lu et al., 2019), fabric/textile (Errokh et al. 2021), or safety industries (Rosendo et al., 2020). Zinc oxide as one of the metal oxides is a highly effective antimicrobial agent, proven by inhibition activity with microorganisms, ensuring a fully workable antimicrobial product. The most recent COVID-19 pandemic strikes urge fast alternative solutions for precautionary and safety issues for human which could be translated into personal protective equipment (PPE), air filters or

even for healthcare products (Ahmed et al., 2020; Ibrahim Fouad, 2021; Karim et al., 2020) as our future emerging techniques adopting the cellulosic availability in hybrid with metal oxide capability (Ramaiah et al. 2021). In this article, various discussions on the mentioned issues are presented and also supported with a preliminary zinc oxide nanoparticle experimented data, in support of COVID-19 purposes.

Cellulosic fibers

This section discusses the process modifications, improvements with related engineering parameters in various industrial applications for vast types of CFs or feedstocks as shown by Table 1 to 3. Firstly, Table 1 shows the summary of CF with investigated research focus for instance for electronic, downstream processing, fabric, energy, fluorescent, food, transportation, building, and architecture as well as material engineering industries. Cellulosic biopolymers are listed with its respective industries. For instance, high performance strain sensor and electromagnetic interference (EMI) shielding application, used cellulose hydrogels or as nanocellulose composites, respectively (Anju, 2021; Wang et al., 2022; Zhang et al., 2022a), representing the electronic industry. The former as being tested for its biomimetic skin with adequate mechanical properties containing nanocellulose or microcrystalline cellulose (MCC) for instance of 50 μm in size where the hydrogel was fabricated chemically via crosslinking and under cryogenically lower temperature of $-20\text{ }^{\circ}\text{C}$. Meanwhile, in the latter, a modified

Table 1 Cellulosic biopolymer research focus and application

Cellulosic biopolymer	Research focus and application	Reference
Cellulose hydrogels, hydrogel fibers, nanocellulose	Electronic/sensor applications; EMI shielding; strain sensor applications	(Anju, 2021; Wang et al., 2022; Zhang et al., 2022a)
Cellular/cellulose membrane, biomembrane, nanocellulose membrane, pollutant adsorbent	Downstream processing industry	(Alipour et al., 2020; Das et al., 2021; Perendija et al., 2021; Wang et al., 2020; Zhang et al., 2022b)
Leaf fibers, sisal fibers, agrofibers, cellulose fibers	Foams, polyester, PLA, biodegradable plastics	(Bendourou et al., 2021; Guimarães et al., 2021; Jabber et al., 2021; Sathees Kumar et al., 2021; Siva et al., 2020)
Modified/regenerated cotton cellulose	Fabric industry	(Khalili et al., 2021; Štular et al., 2021)
Wood-cellulose fiber, graphitic cellufoil, SnS/carbonized cellulose film	Energy industry, battery industries (lithium-ion battery), nanogenerators	(Yi et al., 2021; Yuan et al., 2021; Zhang et al., 2021)
Fluorescent smart materials	Fluorescent application	(Delavari et al., 2020; Kalita et al., 2015; Nawaz et al., 2021)
Dietary fiber, bacterial cellulose	Food industry	(Lin et al., 2020; Revin et al., 2018; Zhu et al., 2022)
CFs	Transportation industry	(Aljubory et al., 2021; Li et al., 2021a)
Cellulose fiber insulator, fire clay bricks, concrete cellulose fiber	Building and architecture	(Pal et al., 2021; Wei et al., 2021)
Reinforced polymer composites, fiber reinforced concrete	Material engineering	(Bansal et al., 2017; Venkatarajan & Athijayamani, 2021; Xu et al., 2020)

nanocellulose composite for EMI, with the reason that CF containing high carbon content, was developed for high shielding effectiveness.

Figure 1 illustrates further the research focusses with the cellulosic biopolymers as elaborated in Table 1. Agricultural-based waste type (agrofibers) for instance from pineapple leaf, sisal fibers, and veld grape were investigated or fabricated as reinforced composites of foams and polyester, polylactic acid (PLA) and also as textile fabrics (cotton) from regenerated cotton cellulose (Bendourou et al., 2021; Jabber et al., 2021; Sathees Kumar et al., 2021; Siva et al., 2020). Banana tree pseudostem wastes and *Eucalyptus* kraft cellulose were few examples of biodegradable product resources being investigated for packaging with biodegradable plastics (Guimarães et al., 2021). Other than that, the CF can also be from bacterial growth for instance from *Achromobacter*, *Alcaligenes*, *Aerobacter*, *Agrobacterium*, *Azotobacter*, *Gluconacetobacter*, *Pseudomonas*, *Rhizobium*,

Sarcina, *Dickeya*, *Rhodobacter*, and *Gluconacetobacter sucrofermentans* B-11267 species microbes (Lin et al., 2020; Revin et al., 2018). The CF-based material produced for architectural design in ensuring that the heat transfer design was optimized using CF insulator type fire clay bricks or in concrete/reinforced concrete, applying strength and safety, was crucially investigated and proven for real applications (Pal et al., 2021; Wei et al., 2021). Polymeric reinforced composites for instance using coir fiber and jute from bamboo were investigated showed high mechanical strength and better results in impact test, Fourier transform infrared spectroscopy (FTIR) test, and Rockwell hardness test (Bansal et al., 2017). Nanogenerators and lithium-ion battery are two instances in energy industries applying cellulose film, foil or fiber for instance from wood-cellulose fiber, cellulose nanofibers, carbonized bacterial cellulose nanofibers with graphene nanoplatelets or with tin sulfide (SnS) nanosheets (Yi et al., 2021; Yuan et al., 2021; Zhang et al., 2021). Fluorescent smart materials support security via fluorescence emission (fluorescent latex nanoparticles, autofluorescent nanocrystals) in anticounterfeiting inks, security documents, sensor, manufacturing, biomedical, and pharmaceutical industries (Delavari et al., 2020; Kalita et al., 2015; Nawaz et al., 2021). In addition to all these, CFs are also fabricated as membrane or a pollutant removal adsorbent in downstream processing industries (Alipour et al., 2020; Das et al., 2021; Perendija et al., 2021; Wang et al., 2020; Zhang et al., 2022b). Besides, CFs from environmental or agricultural wastes are also being innovatively converted as asphalt strength-based product as seen in transportation industries (Aljubory et al., 2021; Li et al., 2021a).

Figure 2 gives an overview of this article content discussing on the various examples of cellulosic fiber sources from agricultural waste as tabulated in Table 2 with various options of pretreatment methods. The cellulosic fiber as isolated via pretreatment methods in form of different

Fig. 1 Cellulosic fiber (CF) with investigated research focus and application network

Fig. 2 Overview of agricultural feedstock to cellulosic fiber hybrid nanocomposite in the search of COVID-19 solutions

Table 2 Pretreatment methods of cellulosic biopolymers for the nanocomposite/material/particle and cellulosic fiber industries

Method and cellulosic feedstock	Pretreatment parameters	Industrial/research application and findings	Reference
Method: chemical; pinecone CFs; pineapple leaf	T = 70 °C; P = ambient; alkali solvent = sodium hydroxide (NaOH) and acidified sodium chlorite (ASC)	Bionanocomposites from nanofibers. Higher NaOH and ASC, greater removal of hemicellulose and lignin	(Rambabu et al., 2016)
Method: chemical; pineapple leaf	T = 80 °C; P = ambient; alkali solvent—sodium hydroxide (NaOH); removal of hemicellulose and lignin	Prior to bleaching with sodium chlorite and acid hydrolysis with sulfuric acid to produce crystalline nanocellulose (CNC)	(Chawalitsakunchai et al., 2021)
Method: chemical; rice husk, coconut husk fiber, food waste, seeds, sea plant, sugarcane bagasse, banana peels	Varying T and ambient P; acidic solvent = hydrochloric acid, sulfuric acid, phosphoric acid, formic acid	Various acidic method to synthesize CNCs or CNF either with single or combinations. Influence the yield, size of CNCs, agglomeration effect, dispersion stability, thermal stability, thermal quality, and thermal strength	(Gopi et al., 2019; Hafemann et al., 2020; Rana et al., 2021; Tibolla et al., 2020)
Method: mechanical-chemical; wheat straw	T and P = ambient; solvent = sodium hydroxide (NaOH); with micro-nano-scale ball milling	Micro-nano-scale ball milling coupling NaOH treatment facilitating the removal of hemicellulose and lignin. Retaining cellulose crystals structure for crystalline transformation. Suitable for cellulose nanomaterial preparation	(Gao et al., 2021)
Method: ionic liquid; oil palm frond, corncob, bagasse powder	T = 90 °C, 130 °C; P = ambient; green solvent = BMIM[Cl], EMIM[DEP], EMIM[Ac], ChOAc	BMIM[Cl], EMIM[DEP], EMIM[Ac], ChOAc. Cellulose crystallinity decreases and facilitates removal of hemicellulose and lignin	(Araújo et al., 2019; Azizan et al., 2021; Mahmood et al., 2016, 2017; Ninomiya et al., 2017)
Method: hydrothermal-chemical; wheat straw, rice straw	T = 170 °C; P = autoclaving pressure and time (t) = 90 min; saturated steam T and at 20 bars; acidic solvent = 0.7% sulfuric acid (hemicellulose solubilization)	Nanocarbons (ANCs) (from hemicellulose filtrate), lignin-containing cellulose nanofibers (LCNFs), lignin nanospheres (LNSs). Hemicellulose filtrate with high xylose content	(Tian et al., 2022)
Method: thermal-chemical-nanogrinding; empty fruit bunch	T = 80 °C and P = ambient; solvent = sodium hydroxide (NaOH) for hemicellulose removal; sodium chlorite for delignification (NaOCl ₂); a steam explosion-sodium hydroxide, sodium chlorite-nanogrinding method	The series of steps until delignification were considered pretreatment prior to crystalline nanofiber (CNF) production via nanogrinding (post mechanical step). CNF production was potentially suitable via nanogrinding treatment	(Supian et al., 2020)
Method: hydrothermal-mechanical; energy-cane bagasse	T = 150–170 °C and P = 1 atm; solvent = deionized water and liquid nitrogen; with disk milling/cryogenic grinding application	Yielded 62% higher xylan recovery and 69.7% higher glucan, with low inhibitor, i.e., acetic acid or furfural generation (i.e., at 150 °C)	(Maitra & Singh, 2021)
Method: physiobiological; microcrystalline cellulose	T = 50 °C and P = ambient; enzyme = cellulase from <i>Aspergillus niger</i> ; dilute acid-microwave-enzymatic (irradiation-chemical-enzymatic) concept	Microwave-assisted dilute acid pretreatment effectively facilitated enzymatic hydrolysis and thermal stability and crystallinity were improved. Nanocrystalline cellulose (NCC) yield was increased	(Qian et al., 2021)

Table 2 (continued)

Method and cellulosic feedstock	Pretreatment parameters	Industrial/research application and findings	Reference
Method: chemical-physical-biological; For-mosan alder biomass	T = 25–180 °C; P = ambient; solvent = sodium hydroxide (NaOH), acetic acid, sodium chlorite (NaClO ₂); enzyme = cellulase complex formula; alkali-acidic-steam explosion-enzymatic concept	The multiple pretreatment series could facilitate the production of NCC	(Ko et al., 2020)
T, temperature; P, pressure; BMIM[Cl], 1-butyl-3-methylimidazolium chloride; EMIM[DEP], 1-ethyl-3-methylimidazolium diethyl phosphate; EMIM[Ac], 1-ethyl-3-methylimidazolium acetate; ChOAc, choline acetate			

sizes up to nanosize can be synthesized with nanoparticles from metal oxides (MeO) for specific type of nanocomposites. The nanocomposite is predicted to be formulated for the search of best solutions for COVID-19 protections. The effectiveness of the antimicrobial agents as synthesized in nanocomposites together with the microbial tests is reported to be investigated with different kinds of analytical characterization and biocidal test activity, respectively.

Process improvements

Process improvements on materials are crucially required with certain chemical intervention and modification for its material or step enhancement for instance via pretreatment or treatment method (Wan et al., 2022). Table 2 shows the summary of research findings including industrial applications as reported by various references with conventional and emerging technologies for the pretreatment of cellulosic biopolymers particularly the lignocellulosic biomass for the nanocomposite, bionanocomposite, crystalline nanocellulose (CNC), nanocrystalline cellulose (NCC), nanocarbon (ANC), lignin containing cellulose nanofiber (LCNF), lignin nanosphere (LNS), and cellulose nanofibers (CNF) for CF industries. Pretreatment with alkali, acidic, water, and green solvents were conventionally imposed singly or in combination with other methods for instance chemical (green or non-green) (Gopi et al., 2019; Hafemann et al., 2020; Rambabu et al., 2016; Rana et al., 2021), physical (grinding/milling or nanogrinding/ball milling) or physical–mechanical-chemical (Gao et al., 2021), hydrothermal-chemical (Tian et al., 2022), thermal-chemical-nanogrinding (physical), hydrothermal-mechanical (Tian et al., 2022), physiobiological (irradiation-microwave-chemical-enzymatic) (Qian et al., 2021), and chemical-physical-biological methods (Ko et al., 2020), emergingly improving the separation or removal of cellulose, hemicellulose, and lignin biopolymers. Such methods were imposed with low to high solvent concentration, pressure (P), and temperatures (T) (from ambient pressure or temperature up to above 20 bar or 180 °C, respectively). Investigated feedstocks investigated were for instance pinecone, pineapple leaf, rice husk, coconut husk, food waste, seeds, bagasse from sugarcane or corn, wheat or rice straws and also from palm trees (empty fruit bunch or oil palm frond). Yield and recovery for instance xylan, xylose, glucan, and cellulose crystal recovery were also observed to increase. Green solvents via methylimidazolium-based ionic liquids facilitating the removal of hemicellulose and lignin for CF is one of the emerging methods for the pretreatment of lignocellulosic biomass as reported. These are the possible pretreatment methods to be adopted either for the isolation of nanocrystalline cellulose from the treated nanofibrillated cellulose with the optimized nanoparticle sizes prior to

the nanocomposite formulation approach. Process improvements with optimized temperature, pH, pressure, concentration of solvents including combinations of processing steps help to produce best outcome of CF pretreatments.

Characteristic and products

The CFs for nanocomposite products were analyzed during preparation for scientific and theoretical justifications. Advanced technological equipment adopting light spectrum exposures are commonly used in various research themes for instance by Fourier infrared transform spectroscopy (FTIR) (Araújo et al., 2019; Mahmood et al., 2016; Ninomiya et al., 2017; Rambabu et al., 2016), X-ray diffraction (XRD) or powder X-ray diffractometry (PXRD) (Araújo et al., 2019; Azizan et al., 2021; Mahmood et al., 2017; Ninomiya et al., 2017; Qian et al., 2021), and X-ray computed tomography (CT) (Mahmood et al., 2017; Qian et al., 2021; Rambabu et al., 2016). Other than that, microscopic analyzer types of scanning electron microscope (SEM) (Araújo et al., 2019; Azizan et al., 2021; Ko et al., 2020; Mahmood et al., 2016; Ninomiya et al., 2017), environmental scanning electron microscope (ESEM) (Rambabu et al., 2016), field emission scanning microscope (FESEM) (Supian et al., 2020), and transmission electron microscope (TEM) (Qian et al., 2021) were also explored. The thermal stability of the nanocomposites or cellulosic-based materials can also be investigated for its thermal stability via thermogravimetric analyzer (TGA) (Mahmood et al., 2017; Qian et al., 2021; Rambabu et al., 2016).

Table 3 presents various analytical characterization purposes for the CF or the composite from various CF for instance via oil palm frond cellulose fibers, natural fibers, bagasse powder, oil palm frond CFs, pinecone CF, banana tree pseudostem, corncob CF, banana peel starch, or empty fruit bunch. FTIR detects functional groups available which characterize the change of the composition of the materials analyzed by its arbitrary intensity of absorbance or transmittance of light at different wavelength [cm^{-1}]. The functional groups are defined by the chemical bonds between oxygen or carbon to hydrogen, carbon to oxygen or hydrogen, aromatic rings and carbonyl bonds. The morphological characteristics of the CFs giving microscopical imaging depending on the instrument's specifications for instance with SEM (up to 500 $\times$), ESEM for natural state imaging (up to 100,000 $\times$), FESEM (up to 20,000 $\times$), and with TEM for high-resolution near atomic imaging using electron beam transmission. The X-ray analysis can be based on diffraction or diffractometry via XRD or PXRD and computed tomography (CT), allowing the former to apply Segal equation (Segal et al., 1959) for crystallinity index while the latter to give information on the 3D volume data of the investigated CF. In addition to that,

the tensile strength together with storage and loss modulus were investigated for strength and stiffness of CF prior to or post pretreatment and fabrication of nanocomposite or bio-based film (Guimarães et al., 2021). The roughness values of the new nanocomposite films fabricated can be determined from the topography images on the scanned surface in the nanoscale resolution from atomic force microscopy (AFM) readings (Tibolla et al., 2020).

Nanocomposite with synthesized nanoparticles

This section focuses on three exemplary preparation methods of nanocomposite and nanoparticles (NPs) or nanocomposites with synthesized NPs as summarized by Table 4. Nanocellulose (CNP) hybrids with metal oxide (MeO) applications were proven to be enhanced for instance in photocatalytic treatment (Shi et al., 2021), food industry (Reis et al., 2021), and biomedical industries (Oprea & Mihaela Panaitescu, 2020). In these three examples, various cellulosic feedstock ranging from soft wood, cellulosic waste and to bacterial cellulose were investigated and fabricated in hybrid with common steps starting with pretreatment of cellulosic biomass for instance ultraviolet (UV) treatment or steam explosion and via generation of cellulose (for bacterial mode), to synthesis of MeO NPs either in situ or ex situ and finally the final preparation of the readied nanocomposites of MeO/CNPs.

Methyl orange dye was treated in a photocatalysis with Ag-ZnO/cellulose nanocomposite (acting as nanocatalyst) which was found to have enhanced photocatalytic activities, improved recovery, catalyst stability, and reusability (Shi et al., 2021). Bionanocomposite from plain parchment fiber for instance polycaprolactone (PCL) reinforced nanocomposite with ZnO nanoparticle was investigated for food packaging film with enhanced mechanical strength, thermal stability, characteristics, and gas barrier permeability coefficient for carbon dioxide (Reis et al., 2021). Another example of nanocellulose with various types of MeO NPs of zinc oxide (ZnO), titanium dioxide (TiO_2), copper oxide (CuO), magnesium oxide (MgO), and magnetite was fabricated either with in situ or ex situ method which was imposed on bacterial cellulose (BC), with the former, having the precursor being trapped inside NC network, and later reduced to MeO. In the latter, only by adding the synthesized MeO to NCs via suspension, dispersion, and homogenization at not extreme temperature but with optimized time to incorporate NCs into BC network (Oprea & Mihaela Panaitescu, 2020). The BC-MeO hybrid, namely, BC-ZnO, has reliable antibacterial characteristics investigated for wound healing cases in biomedical application. Above-all, besides knowing that various precursors could be investigated for these three

Table 3 Analytical characterization for the CF

CFs or nanocomposite	Analytical measurement	Purpose	Reference
Nanocomposite from CFs	Fourier transform infrared spectroscopy (FTIR)	To detect the functional groups for O–H, C=O, C–O, C–O–C, C–H, C–O, C–OH, CH ₂ , aromatic ring, carbonyl	(Alipour et al., 2020; Araújo et al., 2019; Mahmood et al., 2016; Ninomiya et al., 2017; Rambabu et al., 2016)
Oil palm frond cellulose fibers, natural fibers, bagasse powder, nanocrystalline cellulose	X-ray diffraction (XRD); powder X-ray diffraction (PXRD)	To analyze the crystallinity index, i.e., using Segal equation (Segal et al., 1959)	(Araújo et al., 2019; Azizan et al., 2021; Mahmood et al., 2017; Ninomiya et al., 2017; Qian et al., 2021)
Oil palm frond CFs (OPF)	Scanning electron microscopes (SEM)	To observe morphology of CFs, i.e., up to 500× or higher resolution (imaging)	(Araújo et al., 2019; Azizan et al., 2021; Ko et al., 2020; Mahmood et al., 2016; Ninomiya et al., 2017)
Biocomposite, pinecone CF	Thermogravimetric analyzer (TGA)	To analyze the thermal stability in the range temperature, i.e., of 50 to 800 °C	(Mahmood et al., 2017; Qian et al., 2021; Rambabu et al., 2016)
Modified bagasse composite	X-ray computed tomography (CT)	Using E-ray microscope for 3D volume data	(Ninomiya et al., 2017)
Pinecone CF	Environmental scanning electron microscope (ESEM) analysis	To measure the cellulose nanosuspension, i.e., up to 100,000× for natural state imaging	(Rambabu et al., 2016)
Pinecone CF, Banana tree pseudostem bio-based films	Tensile strength	To measure the tensile strength of the biocomposite/bio-film	(Guimarães et al., 2021; Rambabu et al., 2016)
Corn cob CF	Dual-cantilever bending mode for storage modulus (E') and loss modulus (E'')	To analyze the dynamic mechanical analysis via dynamic Young's modulus value for material's stiffness	(Araújo et al., 2019)
Nanocrystalline cellulose, nanocellulose composite	Transmission electron microscope (TEM)	To observe the morphology of the nanocellulose via electron beam transmission	(Alipour et al., 2020; Qian et al., 2021)
Empty fruit bunch (EFB)	Field emission scanning microscope (FESEM)	To observe the morphology of the nanofiber, i.e., up to 20 k×	(Supian et al., 2020)
Banana peel starch nanocomposite	Atomic force microscopy (AFM)	To observe the topography of the nanocomposite films, determining the roughness values	(Tibolla et al., 2020)

Table 4 Three exemplary applications for synthesized nanoparticle or nanocomposite

Nanoparticle/nanocomposite	Synthesis method	Application and benefits	Reference
Ag-ZnO/cellulose nanocomposite	Cellulose source: soft wood bleached kraft pulp; composite method: suspension reaction plus ultrasound dispersion and UV treatment; steps = CF fibrillation, mixed suspension with ZnCl and NaOH, centrifugation, washing, freeze drying, drying, dispersion with AgNO ₃ liquid, UV reaction, washing, final nanocomposites	Application of waste treatment industry (example of dye of methyl orange) for a photocatalysis with excellent photocatalytic activities, stability and reusability	(Shi et al., 2021)
Polycaprolactone (PCL) reinforced nanocomposite with zinc oxide nanoparticle	Cellulose source: plain parchment fiber (PAR); composite method: steam-exploded microfibrillated cellulose modified with ZnO via homogenization. ZnO synthesis: zinc nitrate tetrahydrate (Zn(NO ₃) ₂ ·4H ₂ O) with ammonium hydroxide NH ₄ OH solution; NaOH pretreatment at T = 120 °C, time (t) = 1 h, agitation rate = 200 rpm (pretreated cellulose CFA). Steam explosion at P = 2 bar, T = 120 °C, and N = 200 rpm (ZnO + CFA). Mechanical homogenization t = 1 h, N = 8000 rpm	Application of food industry producing bionanocomposite fabricated for food packing with an enhanced in thermal stability, mechanical strength, Young modulus and improved barrier properties (lower permeability coefficient on CO ₂)	(Reis et al., 2021)
Nanocellulose hybrid with metal oxide nanoparticles	Cellulose source: bacterial cellulose (BC); composite method: with various MeO NPs, i.e., ZnO, TiO ₂ , CuO, MgO, magnetite. Step: (ex situ) by synthesis of MeO NPs and adding to NCs (suspension, dispersion, homogenization into BC network), T = 50 °C, t = 24 h. Step: (in situ) by trapping precursor inside NCs network, reduced to MeO. By impregnating MeO precursor in 3D network of BC, later ultrasonic treatment with hydroxide ions for smaller crystal nanosize incorporation	Application of biomedical—serving as antibacterial properties/formulations for wound healing. Applies electrostatic interactions between positively charged Zn ²⁺ ions and negatively charged hydroxyl groups on the polymeric chains	(Oprea & Mihaela Panaitescu, 2020)

UV, ultraviolet; NaOH, sodium hydroxide; Ag, argentum; ZnO, zinc oxide; ZnCl₂, zinc chloride; AgNO₃, argentum nitrate; TiO₂, titanium dioxide; CuO, copper oxide; MgO, magnesium oxide; CO₂, carbon dioxide; CFA, after alkali pretreatment cellulose; P, pressure; T, temperature; t, time; N, agitation rate

examples with various types of pretreatment methods, it is also known that the final optimized NPs or nanocomposite sizing, methods such as freeze drying, homogenization, and ultrasonic treatment were adopted to crucially influence the final sizing of the NPs, to enhance the effectiveness of the nanocomposite for the intended applications in various industrial applications.

Metal oxide antimicrobial capability

The metal oxides have been investigated for the antimicrobial properties and have the capability to ensure non-interaction of any microbial growth on specific surfaces. Table 5 summarizes on three exemplary microorganisms which are *Escherichia coli* (*E. coli*), *Staphylococcus aureus* (*S. aureus*), and *Pseudomonas aeruginosa* (*P. aeruginosa*), used as model application microbes investigated on antimicrobial activity with synthesized nanoparticles. The antimicrobial activity was conducted on agar medium containing optimized concentration of specific nutrient medium components and culture growth temperature. The setup varied according to the optimal specific growing rate microorganisms investigated, ensuring the predicted highest growth if the MeO NPs fail to work accordingly. In addition to that, the antiviral activity on viruses is reported in this table for instance on the influenza virus H1N1 and SARS-CoV-2 (severe acute respiratory syndrome coronavirus 2) virus with different approach of quantitative antiviral evaluations.

Nanocomposite containing 50% graphene oxide (GO) from a copper oxide-graphene oxide (CuO-GO) has the most optimal antibacterial activity in comparison to other combinations of 5%, 15%, 25%, and 75% of GO (Ahmadi et al., 2021). The percentage of antibacterial activity (R) was defined by how much high R deviation $(B - A)/B$ in reference to the number of colonies counted in the control (B) to the one having the MeO (A) assuming A is lesser than B via colony-forming unit (CFU). Zinc peroxide (ZnO_2) nanoparticle has great antimicrobial/anti-inflammatory potentials against multi-drug and multi-virulent *S. aureus* isolate in comparison to zinc oxide (ZnO) and titanium dioxide (TiO_2) nanoparticles (Ali et al., 2021b). The antimicrobial activity was reported based on the mean diameter inhibition zones rather than the R percentage deviation theory. For the *P. aeruginosa* antimicrobial activity test, the lowest concentration of antimicrobial agents (MIC) visually inhibiting bacterial growth by 99% was experimented in a liquid culture suspension containing nutrient broth (Ali et al., 2021a). The antiviral activity by the H1N1 virus with the presence of ZnO-PEG (zinc oxide-polyethylene glycol) nanoparticles was shown to be higher, comparatively referred to with ZnO-PEG (16–20 nm) rather than only ZnO NPs (20–50 nm) (with the same non-cytotoxic state with ZnO) (Ghaffari et al., 2019). Besides

these identifications on antimicrobial/antiviral evaluation, plaque inhibition assay was also identified via microtiter scale which was conducted on SARS-CoV-2 virus strains with the presence of ZnO NPs allowing highly pathogenic conduct steps to be as minimal volume as possible.

Zinc oxide exemplary synthesis

To observe the capability of metallic NPs, an experiment on extracting zinc oxide (ZnO) NPs had been conducted and characterization was investigated by using Fourier transform infrared spectroscopy (FTIR). In this article, an example of investigation on pH effects on the ZnO formation is presented. FTIR spectral range reading became the indicators of the suitable pH for ZnO nanoparticle formation.

The synthesis of ZnO nanoparticles by chemical precipitation was conducted having the reaction between 1 M zinc nitrate (ZnNO_3) and 25% (w/v) ammonium hydroxide (NH_4OH). NH_4OH in dropwise was monitored for its final varying pH sets of experiment of 7 to 12 with an increment of 1. Three hours of constant stirring producing distinct precipitate with filtrate neutralization of pH via filtration from several washing steps. ZnO NP formation was enhanced with approximately 5-min boiling of the filtrate.

In the mid-IR spectroscopy of 400 to 4000 cm^{-1} , the ZnO functional spectrum was observed due to the absorption of IR via vibrations for instance stretching, bending, and contracting (Viter & Iatsunskyi, 2019). Figure 3 (left) depicts the transmittance [%] of the synthesized ZnO particles at various pH of 7 to 12, for the wavenumber of 300–4000 cm^{-1} . While in Fig. 3 (right) indicates the scanning electron microscope (SEM) photo images of scanned synthesized ZnO at pH 7, 8, and 9 only. In Table 6, previous researches reported various indications of ZnO vibration from IR absorptions at 370 cm^{-1} (Li et al. 2005), 472 cm^{-1} (Viter & Iatsunskyi, 2019), 608 cm^{-1} (Hakim et al., 2020), and 704 cm^{-1} (Purwaningsih et al., 2016), also supporting ZnO fingerprint region (below than 1200 cm^{-1}) for metal cm^{-1} oxides by virtue of interatomic vibrations (Aboorvakani et al., 2020; Sawant & Bamane, 2018; Tiwari et al., 2018). The sharp peaks at ~ 1637 cm^{-1} for pH 7, 10, 11, and 12 may indicate traces of water as shown by the OH bending of the hydroxyl group (Ali et al., 2016; Purwaningsih et al., 2016; Román et al., 2019). The broadening of O–H bands of pH 10 to 12 indicated excessive amounts of OH^- ions via NH_4OH pH adjustment during the ZnO NP synthesis (Ribut et al., 2019), indicating stretching of O–H being present in NPs. As seen at pH 9, the abrupt peak at 595 cm^{-1} indicated the presence of ZnO NPs and the shifted peaks are expected to be also observed at a lower wavelength for other pH conditions only when the wavelength spectrum is lowered below 400 cm^{-1} . Table 6 indicates the compilations of FTIR group

Table 5 Specific microbes and antimicrobial activity on metal oxide nanoparticles

Microbes	Metal oxide (MeO) examples	Identification method	Reference
<i>Escherichia coli</i> (<i>E. coli</i>)	Copper oxide-graphene oxide (CuO-GO) with 5%, 15%, 25%, 50%, 75% GO	T = 37 °C; colony medium = liquid (<i>E. coli</i> suspension + MeO) and agar culture medium (<i>R</i> test); nutrient medium = peptone, sodium chloride, yeast extract	(Ahmadi et al., 2021)
<i>Staphylococcus aureus</i> (<i>S. aureus</i>)	Antimicrobial activity = colony-forming unit (CFU) method using percentage of antibacterial activity Zinc peroxide nanoparticle (ZnO ₂ NP); zinc oxide nanoparticle (ZnO NP); titanium dioxide nanoparticle (TiO ₂ NP)	Antimicrobial activity = colony-forming unit (<i>R</i> equation) T = 37 °C; colony medium = agar medium; nutrient medium = Baird Parker + egg yolk tellurite emulsion	(Ali et al., 2021b)
<i>Pseudomonas aeruginosa</i> (<i>P. aeruginosa</i>)	Antimicrobial activity = mean diameter inhibition zones Zinc oxide nanoparticles (ZnO NPs)	T = 37 °C; colony medium = nutrient broth agar with glucose and nutrient broth (<i>P. aeruginosa</i> suspension)	(Ali et al., 2021a)
Influenza virus A (H1N1)	Antimicrobial activity = minimum inhibitory concentration (MIC) ZnO NPs and ZnO-PEG nanoparticles	T = 35–37 °C; t = 24 h; 5% CO ₂ incubation; MDCK-SIAT1 cell culture medium suspension with MTT reagent and RPMI. Microtiter well plate experiment type	(Ghaffari et al., 2019)
SARS-CoV-2: HAV HM175 strain (ATCC VR-1402) virus on Vero cells	Antiviral activity = via real-time polymerase chain reaction (PCR) ZnO NPs	PCR assay for influenza viral loads T = 37 °C; t = 2 days; 5% CO ₂ incubation; microtiter well plate experiment type	(Attia et al., 2021)
	Antiviral activity = via plaque inhibition assay (MTT colorimetric assay)		

MTT, 3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyl-2H-tetrazolium bromide; RPMI, Roswell Park Memorial Institute medium; MDCK-SIAT1, Madin-Darby canine kidney cell sialic acid receptor; CO₂, carbon dioxide

Fig. 3 ZnO synthesized particle FTIR spectrum data (left) with SEM images (right) of at pH 7 (right-top), pH 8 (right-middle), and pH 9 (right-bottom) at 500×

Table 6 FTIR wavelength and functional groups for ZnO synthesis investigations

ZnO sample T [°C]	FITR wavelength [cm ⁻¹]	Functional group	Reference
400–600	370	ZnO vibration	^a (Li et al., 2005)
400	482	ZnO stretching vibration	^b
	3900 to 380	Water (H ₂ O) molecule present in thin films	(Khan et al., 2011)
480	472	ZnO vibration	^c
			(Viter & Iatsunskyi, 2019)
70	3600–3200	OH stretching	^d
	675–1000	C–O stretching	(Hakim et al., 2020)
	608	ZnO stretching	
400, 500, 600	485, 471, 468	ZnO stretching vibration	^e
400, 500	3352	OH stretching vibration (from water)	(Vaishali et al. 2021)
500	1027–3126	Symmetric stretching vibration bonds of N–O (nitrate complex)	
600	1573	Vibration of –CH ₂ group	
600	2871	C–H stretch	
85	704	ZnO vibration	^f
			(Purwaningsih et al., 2016)
90	3000–3750	O–H stretching; H ₂ O in ZnO	^g
	1634 and 1398	O–H bending; H ₂ O molecules	(Román et al., 2019)
	400–600	ZnO stretching vibration	

^aCalcination temperature from sol–gel method

^bAnnealing temperature from spin coating film

^cCalcination temperature

^dEvaporation of solvent on ZnO film

^eCalcination temperature from self-combustion synthesis method

^fDrying of ZnO NPs at 100 °C

^gDrying of ZnO NPs at 80 °C

detection with varying condition either with calcination or without calcination (i.e., drying or evaporation).

Review on zinc oxide nanoparticles with different precursors

Different NP sizes formed are reported during the synthesis of ZnO using different precursors. In Table 7, zinc acetate ($\text{ZnC}_4\text{H}_6\text{O}_4$), zinc nitrate (ZnNO_3), and zinc sulfate (ZnSO_4) were among the precursors investigated via different ZnO synthesis methods for instance green synthesis, sol–gel, and precipitation methods, producing different sizes of particles. With $\text{ZnC}_4\text{H}_6\text{O}_4$ as precursor via sol–gel method, as reported, as small as 10 nm of ZnO particle was formed (Fallah et al., 2011; Gong et al., 2019). While with precipitation method, considerably small particles were observed of 20 nm (Rădulescu et al., 2016) and also 140 nm (Roy et al., 2021). Considering another common precursor, $\text{Zn(NO}_3)_2$, as being used in precipitation and green synthesis methods, a comparable indication of small particles in the range of 20 to 65 nm was reported and observed (Fakhari et al., 2019; Khalid et al., 2017; Nithya & Kalyanasundharam, 2019; Rădulescu et al., 2016; Shnoudeh et al., 2019; Varaprasad et al., 2016). Despite by using $\text{ZnC}_4\text{H}_6\text{O}_4$ which was likely to produce a large nanoparticle size of 140 nm (Roy et al., 2021), it also was capable to form comparably small NPs ranging from 4 to 32 nm via green synthesis route (Attia et al., 2021; Fakhari et al., 2019; Attia et al., 2020; Rajan Abhinaya & Padmini, 2019; Ratney & David, 2016). Thus, for an ideal nanoparticle size of at least below 100 nm, optimizations must be carefully planned. However, the varying ideal NP sizes formed in varying methods of ZnO synthesis reported do not solely depend on types of synthesis but also being influenced by the concentration of precursors and reaction temperature, besides pH as investigated in Fig. 2.

The proposed chemical bonding model between nano ZnO and cellulose

A model of chemical interaction between nano ZnO with cellulose was described by Li et al. (2021b). Cellulose was dissolved in 1-allyl-3-methylimidazolium chloride; AmimCl ($\text{C}_7\text{H}_{11}\text{ClN}_2$) containing 1 wt% of ZnCl_2 during heating at 80 °C and subsequently degassing at 80 °C. Each Zn^{2+} formed a coordination bond with hydroxyl groups of cellulose that provides one pair electron. Bonded Zn^{2+} was converted into Zn(OH)_4^{2-} after excessive addition of NaOH. Zn(OH)_4^{2-} was transformed into ZnO during sonometric degassing. The evidence for ZnO-cellulose bonding was deduced from ATF-FTIR spectra that shift from 3369 cm^{-1} (stretching OH bond in pure cellulose) and into 3337 cm^{-1} (stretching OH bond in the presence of ZnO). Also, new peaks at 1558 cm^{-1} attributing to the stretching and bending vibration of the bridged hydroxyl groups on the ZnO. The existence of hydrogen bonding between ZnO and cellulose was also confirmed based on X-ray photoelectron spectroscopy (XPS) data (Li et al., 2021b).

Special focus on COVID-19 with nanotechnological strategies on combatting opportunities

All possible combating opportunities against COVID-19 virus are highly recommended on either the readily available or proposed products both as the protective personal equipment (PPE) or protective personal clothing (PPC) with certain criteria as illustrated in Fig. 4. They must be safe, effective, affordable, sustainable besides being highly durable and readily available worldwide. The products surely to have been proven with reliable scientific proofs which are technologically sound and feasible. Above-all, some products should be offering antimicrobial and antiviral capabilities

Table 7 Zinc oxide nanoparticle synthesis with different precursors

Nanoparticle size [nm]	Synthesis method	Precursor	Ref
4, 8–12, 10–32, 21.49, 25	Green synthesis	Zinc acetate	(Attia et al., 2021; Fakhari et al., 2019; Attia et al., 2020; Rajan Abhinaya & Padmini, 2019; Ratney & David, 2016)
10, 50	Sol–gel	Zinc acetate	(Fallah et al., 2011; Gong et al., 2019)
140	Precipitation	Zinc acetate	(Roy et al., 2021)
20, 25, 30, 38, 65	Precipitation	Zinc nitrate	(Khalid et al., 2017; Nithya & Kalyanasundharam, 2019; Rădulescu et al., 2016; Shnoudeh et al., 2019; Varaprasad et al., 2016)
25.26, 65	Green synthesis	Zinc nitrate	(Fakhari et al., 2019; Nithya & Kalyanasundharam, 2019)
13–23, 25	Precipitation	Zinc sulfate	(Khalaf et al., 2019; Mohan & Renjanadevi, 2016)

Fig. 4 Criteria for proposed or already available COVID-19 products

which meant to be used as not as single use but with multiple repetitions including with highly protective against the pathogenic virus on eyes, nose, and mouth besides being comfortable to be worn (as for PPE). Antimicrobial strategy can be programmed either to kill (destroy), suppress, or inhibit the pathogenic virus and bacteria (Karim et al., 2020) intending to also fulfilling these criteria mentioned for combating further risk of exposure and cross-infections by the intended product or equipment without being destroyed.

In combatting the wide spread of COVID-19 virus, it is crucial to hinder or dampen the transmit from one another or from one surface to other surfaces. Not only by considering the direct contact faced by the healthcare workers with patients but also among people in the community as whole. Thus, special approached products or equipment which use certain strategies for instance nanotechnological concepts are significantly required. Personal protective equipment (PPE) or personal protective clothing (PPC) like face mask, respiratory mask, goggle, face shield, glove, apron, coveralls, or footwear covers are the most common required items to be adopted for this purpose.

Table 8 presents some of the examples for this matter. Recently, antimicrobial functions or the biocidal killing of the pathogenic microorganisms are proposed and innovated in face mask as nanomaterial hybrid composites. Despite it being as highly comfortable for the wearer in breathing but also as having the self-killing function being offered can be

considered as a significantly required item to have (Campos et al., 2020). Technologically, coating the antiviral agents by using the atomic layer deposition (ALD) method on the respiratory mask as innovated allows to create the hydrophobic surfaces against the virus considering the short time span of its diffusion or penetration onto the surfaces (Shirvanimoghaddam et al., 2021). NanoHack mask made from copper nanocomposite is made to protect the wearer from airborne particles from the surrounding other than another type of nanocomposite as innovated containing antimicrobial additives for rapid prototyping medical device manufacturing as reported (Zuniga & Cortes, 2020).

Patients with chronic or non-chronic wounds need medical treatment for fast healing and wound dressing with antimicrobial agents is visioned not only to function for self-healing via transdermic contact but also to avoid the infected microbes from patients to be transferred to the surrounding. Thus, nanofiber-based electrospun wound dressing created via electrospinning may become a hybrid polymers which are high biocompatible to human and also biodegradable with nontoxic components (Jatoi, 2020; Sylvester et al., 2019). Apart dealing with the patients, possible antimicrobial-based products are also available in healthcare working environment like plastic waiting room chairs and toilet seat with pure copper (70%) or resin composite, allowing reduction of environmental contaminations in hospitals. These are few examples adopting nanotechnological concepts besides

Table 8 Examples of proposed product or equipment with nanotechnological strategies in combating COVID-19 or other environmental problem for healthcare workers

Product/equipment	Purpose	Nanotechnological strategy (in the market* or proposed**)	Reference
Nano-based PPE (face mask)	To increase comfortability and ease breathing capability besides filtering virus particles, self-killing pathogens (hybrid composite)	Nanomaterial hybrid composites	(Campos et al., 2020)
Antiviral coating on respiratory mask	To create hydrophobicity layer against virus	**Atomic layer deposition (ALD) method	(Shirvanimoghaddam et al., 2021)
NanoHack mask	To protect from airborne particles	*Copper nanocomposite	(Zuniga & Cortes, 2020)
Antimicrobial filament for 3D printing	To be used as antimicrobial additive in rapid prototyping medical device manufacturing	Copper nanocomposite additive mixture	(Zuniga & Cortes, 2020)
Plastic waiting room chairs	To reduce environmental contamination in hospitals	*Copper nanoparticle-polymer matrices' composite	(Palza et al., 2018)
Toilet seat-pure copper/resin composite (70% copper)	To reduce environmental contamination in hospitals	–	(Palza et al., 2018)
Wound dressing	To create wound dressing hybrid polymers that are highly biocompatible, non-toxic, biodegradable, and antibacterial for chronic/non-chronic wounds	*Nanofiber-based electrospun wound dressing via electrospinning (with antimicrobial agents)	(Jatoi, 2020; Sylvester et al., 2019)

being as hybrid or a single function product available in healthcare worker environment.

Conclusion

The agricultural wastes which are abundantly available have high potential to offer for world's benefit on recent COVID-19 strike, via their cellulosic fiber formulation with antimicrobial agents. Metal oxides and nanocellulose in hybrid as nanocomposites serve effectively in various industries. Natural lignocellulosic feedstocks and the related wastes need to be pretreated for CF production together with the synthesis of metal oxides, prior to undergoing the fabrication of hybrid nanocomposites as the future opt for COVID-19 hybrid product to a safer resistant environmental offer to human. Metal oxides or zinc oxides serve as antimicrobial agent as reviewed indicated by solid or suspension microbial inhibition technique. Zinc oxide exemplary synthesis results presented pH effectiveness via functional group identifications and review on various effective precursors laid down open possibilities to effective methods for antimicrobial agent synthesis. The possibilities of zinc oxides alone in combating COVID-19 virus via its current instances of antimicrobial coating, nano-spray product, as personal protective equipment (PPE), air filters, as in textile product and drug delivery can become future emerging solution alternative for a better safety and precautions which is stated in the title of this article as a highly potential option. This is supported

by great analytical characterization available as reported in this article with astounding scientific proofs together with variety methods of antimicrobial agent effectiveness with the formulated nanocomposites. With this knowledge, the search for COVID-19 options with the usage of cellulosic fiber can be speeded up.

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Author contribution AA wrote the article and investigated on COVID-19 proposal solutions for CF intervention.

AAS experimented on the ZnO antimicrobial synthesis.

MBSB investigated on the precursors of ZnO synthesis.

MHD searched the idea of the nanocomposite.

NRR supervised on MBSB work.

NFAB advised the nanoparticle synthesis work.

MA gave the input on the metal oxide chemistry link.

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