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Nanocoatings: Universal antiviral surface solution against COVID-19

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1. Introduction

Viral pandemics raise serious threat to human life. The coronavirus disease 2019 (COVID-19) caused by severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2) has emerged as a global pandemic since its outbreak in Wuhan, China during December 2019. SARS-CoV-2 through its varied mutants, still persistently maintains its ubiquitous presence. As of 9th June 2021, the global death due to COVID-19 has reached 3,742,653 with confirmed cases of 173,609,772. Latest reports revealed the sustainability of SARS-CoV-2 for about three days on a plastic surface as well as on stainless steel surface, one day on cardboard and about four hours on a copper surface. World health organization (WHO) does not rule out the chance of ‘airborne transmission’ (where smaller droplet nuclei or dust particles containing the pathogen can remain suspended in air) of COVID-19 like its predecessor, SARS-CoV-1. While the respiratory droplets are >5–10 μm in diameter, droplet nuclei have diameter <5 μm and their mode of transmission is schematically shown in Fig. 1.
As of now, in most countries, the accessibility of vaccination against SARS-CoV-2 is limited to healthy workers and more vulnerable people. Hence, the percentage of prioritized people is very low, the global community should be vigilant and continue the current lifestyle to some extent. Moreover, the efficacy of existing vaccine against several variants of SARS-CoV-2 is still a question among the experts. In this scenario, a sustainable solution for the prevention of COVID-19 is very crucial [10]. With this unprecedented current pandemic situation, the demand over antiviral protection strategies has been massively increased. The use of personal protection equipment (PPE) including facemasks is considered as the most trusted way of protection against droplet transmission of COVID-19. However, these viral filtering media allows only filtering of virus and they are not meant to destroy or avoid virus proliferation. This may cause severe viral transmission via used and discarded PPEs. Moreover, the contaminated surfaces in healthcare environment, public places and even in domestic environments, are the major transmission sites for SARS-CoV-1. The disinfection of daily handling surfaces is critical for healthy life. The surfaces which itself have the anti-virucidal activity, would be a promising solution for this issue [11]. The antiviral coatings over these surfaces kill the virus and avoid the transmission of COVID-19 through the respiratory droplets deposited on the surfaces [12,13].

Nanomaterial can offer technologies to develop universal antiviral protection coatings. The better understanding of the structure of the virus would help to design nanomaterials to act against [14]. Coronavirus are enveloped positive-sense single-stranded RNA virus [15–17]. The size of the coronavirus is 65–125 nm in diameter and is characterized by glycoprotein spikes on the outer surface (Fig. 2). These crown like spikes of coronaviruses are responsible for the attachment and entry of the virus to host cells [18]. This morphological feature highlights the possibility of interaction between the coronavirus and nanomaterial, which will be the target of the nanotechnologist to act against coronavirus. Latest review articles detailed possible mechanism of inhibition of SARS-CoV-2 entry to host cell by the nanomaterials [19,20].

The current review discusses the scope of using nanomaterials as antiviral coatings for PPEs including masks and daily handling surfaces in domestic and public places to protect the humankind from COVID-19. Antiviral surface coatings offer pathogen free surfaces in healthcare environment and daily life [22]. In the current scenario, such coatings are needed in textile materials, personal protection equipment (PPE), portable electronics, medical devices, domestic home furnishings, automotive, food packaging and many more. Based on the previous reports on the use of nanomaterials against viruses having similar morphology, size and mode of transmission as that of SARS-COV-2, this review suggest a pathway to redirect the research in nanomaterials to change our life style from the lessons learnt from COVID-19 pandemic.

![Fig. 1. Larger droplets with viral content deposit close to the emission point (droplet transmission), while smaller droplets can travel several meters long in the air indoors (airborne transmission) [9].](image1)

![Fig. 2. The structure of corona virus [21].](image2)

2. The antiviral action of nanomaterials against corona virus models

The review starts with the existing reports on the use of nanomaterials against corona virus models. Various studies have been conducted worldwide to explore the antiviral action of nanoparticles against SARS-CoV-1, MERS-CoV and other corona virus models. The nanostructures of metals and metal oxides have been evolved as popular antiviral agents. The anti-viral activities of nanostructured silver (Ag) [23,24], titanium dioxide (TiO2) [25], copper oxide (CuO) [26], zinc oxide (ZnO) [27] etc. have been well proved. The inherent sterilizing effect of TiO2 due to its photocatalytic decomposition of the microbial matter, make them suitable to fabricate antiviral films [28]. Lv et al. [29] studied the antiviral effects of silver (Ag) nanomaterials against coronaviruses. They have tested the inhibitory effect of Ag nanostructures on transmissible gastroenteritis virus (TGEV), a porcine coronavirus. Spherical Ag nanoparticles (diameter < 20 nm) and silver nanowires having two different dimensions (diameter – 60 nm, length – 60–80 μm and diameter 400 nm, length 20–30 μm) were proposed to establish direct interaction with TGEV surface glycoprotein to inhibit the initiation of TGEV. According to the authors, the silver nanoparticle and nanowires act against coronavirus by inhibiting TGEV-induced apoptotic signaling cascades. In the case of copper nanoparticles, it was reported that the released copper ions generates reactive oxygen species (ROS), which completely kill the virus [30]. Biosynthesis of nanoparticles in green environment helps to reduce their cytotoxicity. For instance, the copper nanoparticles can be generated using L-vitamin...
C, which avoids the toxicity of copper nanoparticle [31].

As the cytotoxicity associated with metal nanoparticles is a major challenge, nanomaterials other than metals and metal oxides are getting more attention. In this scenario, traditional natural herbs have been investigated for their potential to resist several viral infections, including those caused by SARS-CoV1 [32]. Turmeric, a vital component of India’s traditional medicine system, is known to have antiviral activity. Curcumin, a polyphenol compound obtained from turmeric roots, have the ability to inhibit human pathogenic virus like the influenza virus, hepatitis C virus and HIV [34]. Curcumin was shown to inhibit SARS-CoV1 in the dose range of 3–10 μm [35]. However, insolubility in aqueous medium limits its clinical application. Researchers reported that the use of nanoscale forms of curcumin could help to overcome the disadvantage of low aqueous solubility and poor bioavailability of curcumin [36]. Carbon dots (CDs) obtained from curcumin is an effective way to enhance the bioavailability of curcumin. In such an attempt, Du et al. [37] prepared carbon dots from curcumin (CCM-CDs) by a simple hydrothermal method and demonstrated their inhibitory effect against porcine epidemic diarrhea virus (PEDV) as coronavirus model. Less cytotoxic cationic carbon dots with ultra-small size (diameter ca. 1.5 nm), rich hydrophilic groups and a positive potential (+15.6 mV) were prepared from curcumin. The survival rate of Vero cells exposed to different concentrations of CCM-CDs is shown in Fig. 3 and it shows cell viability above 90% after 24 and 48 h indicating low cytotoxicity of CCM-CDs. This cationic carbon dots from curcumin was found to inhibit all stage of viral infection via viral entry, the synthesis of negative-strand RNA in the virus, the budding of virus and the accumulation of reactive oxygen species (ROS) by PEDV. The positively charged CCM-CD undergoes strong electrostatic interactions with PEDV and thereby competing to bind the virus to cells. CCM-CDs showed superior antiviral activity than common CDs (ethylene diamine (EDA)-CDs) as indicated by PEDV titer values when exposed to CCM-CDs and EDA-CDs (Fig. 3b).

Another promising non-toxic nanoparticle which would expect to have potential antiviral action against corona virus is chitosan nanoparticles. Chitosan is a cationic polysaccharide. N-(2-hydroxypropyl)-3-trimethylammonium chitosan chloride (HTCC) has reported inhibitor activity against coronavirus entry [38]. There are preliminary reports on the action of HTCC against SARS-CoV-2 and MERS-CoV [39]. The electrostatic interaction between the cationic polymer and the spike protein of coronaviruses is the key factor in inhibiting the coronavirus entry into the host cell. The plausible mechanisms of antiviral activity of chitosan have been reviewed elsewhere [40].

In addition to the inherent antiviral effect of nanomaterials, the hierarchical nano-surfaces possess superhydrophobicity which prevent the adhesion of virus. It was reported that superhydrophobic surfaces exhibit a significant reduction in SARS-CoV-2 attachment of up to 99.99995% compared to bare surfaces [41].

3. The nanocoatings: the future of antiviral surfaces

The frequent and regular use of disinfectants is not a sustainable solution to control the transmission of corona virus. The use of disinfectants is not ecofriendly and long lasting. In this scenario, the develop self-sanitizing surfaces is a wiser solution for this problem. Herein, the nanotechnology could offer permanent coating over the daily handling materials (fabrics, glass, metals, plastics etc.), thereby make virus free surrounding. Since early 2020, scientist has been extensively involved in research and discussions to find an antiviral coating solution by making use of inherent antiviral property, size and shape effect of nanomaterials and nanostructures. The following sections will detail both reported and proposed research on developing antiviral nanocoating against SARS-CoV-2 over various substrates.

3.1. The nanocoating over commercial masks to prevent transmission

The commercial medical masks are made of polypropylene (PP) non-woven fabric with three different layers. While the outer layer filters large particles, the middle layer is a filter media for fine particles and the third layer is a direct contact skin layer. The different layers in commercial mask and their filtering capacity of different particles is shown in Fig. 4. The filtered microbes and dust should remain on the surface of these layers.

In certain circumstances, the commercially available medical masks and respirators have very low protection level. In 2006, Li et al. [42] studied the performance of commercial medical mask and N95 respirators. They found that both medical mask and respirators allowed the penetration of viruses smaller than 100 nm. In the same year, Balazy et al. [43] critically analyzed the filtering efficiency of both N95 respirators and surgical mask against airborne viruses. They tested the collection efficiency of these masks using bacteriophage MS2 virus (a non-harmful simulant of several pathogens) in the particle size range of 10 to 80 nm. The study revealed that N95-respirators might not necessarily block penetrating particles of size smaller than 300 nm. So, N95 respirators sometimes failed to provide 95% protection level against airborne viral agents and some surgical masks allowed the penetration of significant fraction of airborne viruses. The lack of performance of medical masks forced the global researchers to think about high performance, reusable medical masks for the safety of both public and medical workers. Even though the triple layered medical mask is adequate to block respiratory droplets, they are in some cases unable to filter fine droplet nuclei.

Fig. 3. a. The effects of different concentrations of CCM-CDs on Vero cells viability were detected by CCK-8 assay; b) the titer of PEDV when exposed or unexposed to 125 μg/mL EDA-CDs or CCM-CDs [37].
Due to the reported drawbacks of commercially available medical mask in filtering, killing the pathogens and risk associated with their disposal, researchers have been engaged in devising effective medical masks in economical and simple means. Two main strategies were employed to achieve this goal. The first one is to find an alternative material for fabrics used in commercial face mask such as the use of electrospun polymer nanofibers [44,45,46]. Yang et al. [45] came up with a user comfortable mask made up of nanofiber on nanoporous polyethylene (fiber/nanoPE) for a proper thermal management (Fig. 5). This type of nanofiber masks would effectively eliminate difficulty in breathing especially for elderly person and those with lung diseases. Further, the Ag coating on fiber/nanoPE has warming effect.

The second strategy is to either modify surface of the fabric or apply a protective coating over the fabrics used in commercial face mask. Among such attempts, the use of anti-microbial protective coatings for the fabric layers has been emerged as a promising approach to control the infection. Some suitable antimicrobial coatings over commercial medical mask and N95 respirators have been reported without affecting its basic performances. Some metals and metal oxides have effective anti-microbial properties. Borkow et al. [47] impregnated copper oxide into disposable N95 respiratory mask layers. They showed that mask layers with copper oxide have anti-viral activity against human influenza A virus (H1N1) and avian influenza virus (H9N2). By integrating anti-viral activity, the mask could reduce the risk of environmental contamination with virus. Recently, Balagna et al. [5] showed that silver nanocluster/silica composite sputtered coating over commercial face-mask has excellent virucidal activity against SARS-CoV-2. Another milestone in this area is the generation of a photo sterile mask by the utilization of photo-thermal effects of nanomaterials [48]. The self-sterile properties along with super hydrophobic nature of these surfaces make the mask more effective to stop the spreading of COVID-19 [49]. Lin et al. [50] developed such a photo sterile mask based on graphene nanosheet-embedded carbon (GNEC) film which can effectively reused after solar illumination. GNEC films developed by ultra-sonic extrusion were uniformly and evenly distributed on the melt blown polypropylene fibers (Fig. 6). These GNEC films coated mask have surface aggregated charges which make the mask hydrophobic. The temperature sensitive spike proteins in Corona virus can be inactivated by high temperatures. Upon solar illumination, the temperature of the GNEC coated mask rises rapidly and most virus are inactivated over 80°C. Pal et al. [51] also developed a sunlight sterile mask by depositing few layer ultra-thin graphene coating onto commercial surgical mask by a dual mode laser induced mechanism.

Apart from graphene nanosheets, copper nanoparticles (CuNPs) are also used to develop photo sterile coating for commercial masks. Kumar et al. [52] developed a nanocomposite coating of natural bioadhesive polymer (shellac) with CuNPs over nonwoven surgical mask. In this coating shellacs act as a binder for CuNPs into the mask fabric. The mask was developed via a scalable technique, where the surface of nonwoven fibers deposited with nanocomposite particles using spray technology.
In this case, the combined photocatalytic and photothermal effect of CuNPs are responsible for the self-sterile ability of the mask (Fig. 7a). This photoactive antiviral mask (PAM) could break the plasma membrane of virus-like particles (VLPs) under sunlight illumination as seen in transmission electron microscope (TEM) images (Fig. 7b).

In order to reduce the risk of copper toxicity, Kumar et al. [30] coated the metal nanowires with a metal-organic framework (MOFs) and polymer. Uniform core-shell structures of Cu@ZIF-8 NWs were obtained by the growth of zeolitic imidazolate framework 8 (ZIF-8) over Cu nanowires stabilized using pluronic F-127 block copolymer. Thus prepared Cu@ZIF-8 NWs were used to coat the polypropylene filter material in medical grade face mask using a simple dip-coating technique where nanowires were attached to the fibers of the filter media uniformly. They have proved antiviral efficiency of Cu@ZIF-8 NWs even at low concentration associated with sustained release of both copper and zinc ions.

Exploring various nanostructured morphology in metals and metal oxide would help to entrap the corona virus. The morphological features such as nanocactus, nanoflower, nanostar etc. would expect to facilitate the easy entrapping of this crowned virus. A promising antiviral coating has been proposed for cotton mask using non-toxic ZnO with nanoflower morphology [53]. The self-assembled petal like structures of ZnO nanoflower over cotton cloth is shown in Fig. 8. Computational study revealed that the petals of ZnO nanoflowers are able to trap SARS-CoV-2 spike proteins and denaturate the spike proteins on the nanosurface.

3.2. Nanocoating over other fabrics

In the current pandemic situation, it is critically important to integrate self-disinfection ability to fabrics used in clothing, surgical dressings, beddings, wipes etc. Hamouda et al. [54] developed a cellulose-based wipe treated with silver nanoparticles (Fig. 9a). The antiviral
activity of AgNPs imparted disinfectant property to the wipe and was successfully tested against MERS-CoV with 48.3% viral inhibition at 0.0625 μL. They have proposed the future use of this wipes in hospitals and healthcare centers against human pathogenic viruses especially coronavirus. They have also developed low cost antimicrobial and antiviral winter sweater using cotton yarns treated with AgNPs (Fig. 9b).

A multifunctional fabric was developed for protective clothing application where the electrospun nanofibers of poly(methyl methacrylate) (PMMA) decorated with nanoparticles [55]. ZnO nanorods and Ag nanoparticles were decorated over the nanofibers to impart antiviral activity to the nanofiber mat. In the pandemic period, scientists put forward the suggestion of using the electrospun chitosan nanofibers with positive zeta-potential in personal protective cloths. The electrostatic interaction between the chitosan nanofibers and the viral spike reduce the virus infection [56].

3.3. Nanocoating over other solid surfaces (metals, plastics, glass etc.)

Apart from masks and fabrics, there are some other materials used in hospital and other public environment which need to protect from the virus. They include display glass for consumer electronics and smartphones [57,58], contactable glass surfaces and metal surfaces in hospitals, public offices, schools and public transport etc. A universal transparent coating systems has been developed which would be suitable for face shield, screen guard for cell phone, door knobs, lift buttons, medical instruments etc. without affecting the appearance or function of the surface [59]. They make use of synergistic antiviral effect of copper nanoparticles and graphene nanoparticle (Cu-Gr hybrid) in poly(vinyl alcohol) (PVA)- based nanocomposite coating. Here, the compositing of nanoparticles helps to inhibit their agglomeration and stabilizes them by altering their surface architecture [60] and their antiviral effect remains unaltered upon blending with PVA to make the coating completely transparent (Fig. 10). They have proposed a possible mechanism on antiviral activity of Cu-Gr hybrid based on their study against influenza A virus. Due to the close resemblance between SARS-CoV-2 and influenza A virus, in terms of particle size and mode of transmission, same protective measures can be employed to prevent disease transmission. Both copper oxide and graphene in their composite affect the structural integrity of the viral envelope and the HA protein to interfere the entry of the virus particles into the host cells.

The creation of nanostructures on metal surfaces is an effective way to use them as antiviral surfaces. Aluminum metal (Al) which is a widely used metal in hospital and medical equipment have been etched with nanostructures topography and studied against SARS-CoV-2. Morphological studies revealed that wet etched surface of the Al have nanostructures which are grouped randomly in the form of parallel ridges.
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A universal coating solution against SARS-CoV-2. In Fig. 12, the nanoparticles embedded PEM would be another viable representation of fabrication and mechanism of action of PMSs is shown induced contact killing and anti-adhesion [68-70].

Polyanions and polycations on any type of substrates including textiles, glass, metal etc. would facilitate the easy entrapping of this crowned virus and interaction of spike protein with the nanostructures would be the expected mechanism.

4. Opportunities and major challenges

Material scientists and nanotechnologists are proposing and investigating several nanomaterials as antiviral protective coatings against SARS-CoV-2 over easily vulnerable substrates. This includes reusable masks, textiles, electronic gadgets, doorknobs, medical instruments etc. There are still numerous opportunities in nanocoating technology which would be explored and some of them are listing herewith. Like nanomaterials, polymer has also been designed for effective virus inhibition [62]. Antiviral polymers [63] including dextran sulfate [64], sulfated polysaccharides [65], heparin [66], poly(acrylic acid) and poly(methacrylic acid) are potential viral inhibitors and strategies to associate these polymers with nanomaterials would expected to augment the efficiency of antiviral nanocoatings against SARS-CoV-2. Those strategies may include chemically grafting of antiviral polymers on the nanomaterials and the generation of nanocomposites by incorporating nanomaterials in to antiviral polymers.

Polyelectrolyte multilayers (PEMs) are known antimicrobial coating solutions which deactivate the microbes via electrostatic interaction induced contact killing and anti-adhesion [68-70]. PEMs can be developed via. alternating deposition of polyanions and polycations on any type of substrates including textiles, glass, metal etc. The schematic representation of fabrication and mechanism of action of PMSs is shown in Fig. 12. The nanoparticles embedded PEM would be another viable universal coating solution against SARS-CoV-2.

Even though there are no scientific reports on viability of SARS-CoV-2 on food surfaces, it is indeed to eliminate the risk of contamination on food and food packaging [71]. Yekta et al. [72] does not rule out the feasibility of viral transmission through food stuffs. Nanotechnology can offer edible coatings for fruits, vegetables and other food stuffs. There are some fascinating bio-based nanomaterials which have proven antiviral activity, which could make use in protecting the food items from viral contamination, as their nanocomposites with biodegradable polymers. Curcumin, known as a golden spice, have the ability to inhibit human pathogenic virus like the influenza virus, hepatitis C virus and HIV [73]. In the wake of COVID-19, the scientists have been thinking of nanocurcumin therapy against SARS-CoV-2 [74]. Curcumin incorporated biodegradable polymer coatings will be better solution for the food safety during the pandemic. Similarly, chitosan will be suitable choice as biodegradable polymer matrix for developing edible nanocomposite coatings for food stuff with COVID-19 safeguard. It is proposed that the cellulose nanomaterials isolated from agricultural wastes decorated with antiviral nanoparticles will serve as economic antiviral food coatings (Fig. 13).

Carrageenan, a sulfated polysaccharides extracted from red seaweeds is an antiviral polymer applicable as antiviral edible coatings for fruits [75]. Carrageenan nanocomposites designed with potent antiviral nanomaterials would be expected to have synergistic viral inhibition. Carrageenan nanocomposites coatings would be sustainable solution for antiviral edible coatings in this current pandemic scenario.

A major challenge in this area is the lack of testing possibility of the nanomaterials against SARS-CoV-2, as it required biosafety level 3 (BSL-3) laboratories due to its highly transmissible and coronavirus models [76]. This prevents the scientific community from exploring the actual antiviral response of the developed nanomaterials against SARS-CoV-2.

5. Conclusions and future outlook

In brief, nanomaterials could offer a sustainable solution in minimizing the spread of COVID-19 in the form of nanocoatings over vulnerable surfaces. The review highlights the potential of various nanomaterials including metal and metal oxide nanoparticles, graphene nanosheets, chitosan nanoparticles, curcumin nanoparticles, electrospun polymer nanofibers in developing antiviral coatings for various substrates. Hybrid of two or three above mentioned nanomaterials could impart synergistic antiviral action toward this deadly spike virus. The hierarchical nanostructures such as nanocactus, nanoflower, nanostars etc. would facilitate the easy entrapping of this crowned virus and denaturate its spike protein. The combination of nanoparticles with polymers would make functional coating materials to keep the surfaces away from SARS-CoV-2. Textiles finished with antiviral polyvinyl alcohol (PVA) nanocomposites will be studied for their action against SARS-CoV-2 and its variants. The practice of isolation of nanoparticles from agricultural wastes and its use in large scale development of antiviral coatings will be entertained. The research in this area will need to explore more with respect to experimental evidence on the action of various nanomaterials against SARS-CoV-2. Moreover, the use of same antimicrobical coatings against different kinds of emerging pathogens needs to study. The nanotechnology research prospect more sustainable and scale-up anti-COVID surface coating techniques for better healthy future.

CRediT authorship contribution statement

Poornima Vijayan P: Conceptualization, Methodology, Writing-Original draft preparation, Supervision.
Chithra P.G, Pinky Abraham, Jesiya Susan George, Hanna J Maria & Sreedevi T: Writing-Reviewing and Editing, Sabu Thomas: Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence
Fig. 11. Scanning electron micrographs of the (A) smooth control Al (scale bar = 1 μm) and (B) etched Al (scale bar = 200 nm) with nanostructured topography (C). Viability of SARS-CoV-2 on the surfaces of the etched (nanostructured) Al 6063 alloy, control Al 6063 alloy and nonmetal surface tissue culture plates (TCP) at different time intervals of 1, 3, 6, 24, and 48 h. The titers of viable viruses are expressed as TCID$_{50}$/mL in logarithmic scale [61].

Fig. 12. (a) Schematic representation of a polyelectrolyte multilayer (PEM) film buildup by successive adsorption steps of polycations and polyanions followed by rinsing steps using the dipping method. (b) Three main strategies were followed to design antimicrobial PEM: antiadhesive films inhibiting the close approach of pathogens, contact killing films by exposing antimicrobial agents on the surface, and release-killing films delivering antimicrobial agents in the supernatant, with the last two strategies leading to the death of pathogens [68].
the work reported in this paper.

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