Fiber-Based Masks and Respirators: Using Decontamination Methods and Antimicrobial Treatment to Improve Its Reusability during Pandemic

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Abstract: Shortage of personal protective equipment (PPE) is often projected in response to public health emergencies such as infection outbreaks and pandemics. Respiratory protective devices (RPDs), namely medical face masks and respirators, are considered the last defense for the frontline healthcare workers. Cleaning, decontamination and reuse of the disposable RPDs have been accepted by local health authorities during the pandemic period. To contribute to the mitigation of RPD shortage and ensure the safe adoption of decontamination protocols, this review discusses the regulated testing standards and the most commonly studied decontamination methods in the literature. The reuse of RPDs must fulfill three criteria: remove the microbial thread, maintain original function and structural integrity (including fitting tests) and leave no harmful residuals. Decontamination methods such as ultraviolet germicidal irradiation, moist heat and vaporized hydrogen peroxide appeared to be the most promising methods in balancing the above-mentioned criteria. However, the effectiveness of decontamination methods varies depending on the RPDs’ models, materials and design. Therefore, the adoption of protocols needs to be evidence-based with full validation in the local institutes. Additionally, new technology such as antimicrobial treated PPE that can reduce the risks of fomite during donning and doffing process with an extended lifespan should be encouraged. Overall, good training and guidance for appropriate reuse of RPDs are fundamental to ensure their efficiency in protecting front-line healthcare workers.

Keywords: respiratory protective devices; filtering facepiece respirators; masks reuse; personal protective equipment; antimicrobial masks; fiber-based material; textile

1. Introduction

Personal protective equipment, commonly referred to as PPE, is defined by the Occupational Safety and Health Administration (OSHA) as equipment worn to minimize exposure to a variety of hazards [1]. Many guidelines have been issued to advise the practical use of PPE in various work professionals [2–4]. In the Hierarchy of Hazard Controls, PPE is considered an option of the last resort, providing a barrier to prevent work injuries from potentially hazardous work environments [5]. PPE type varies and includes head and scalp protection, respiratory protection, eye protection, hearing protection, hand and arm protection, foot and leg protection, body protection, and height and access protection [6,7]. PPE has been commonly used in healthcare settings, such as at surgical sites and in infection outbreaks [8,9]. Lately, the growing pandemic caused by severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2) has placed the healthcare workers (HCWs) at great risk of coronavirus disease 2019 (COVID-19) infection [10]. The use of PPE in healthcare and community settings plays a crucial role in preventing the transmission of SARS-CoV-2 during
COVID-19 patient caring [11]. World Health Organisation (WHO) has recommended the use of PPE, including gloves, medical masks, goggles or a face shield and gowns, as well as for specific procedures, respirators (i.e., N95 or FFP2 standard or equivalent) and aprons in different activities involving contact with COVID-19 patients [11].

In the context of PPE for infection control, respiratory protective devices (RPDs) undertake as part of the package for personal protection [12]. It is also considered the last line of non-invasive defense for HCWs against respiratory infection transmission [13,14]. RPDs can be divided into medical masks and respirators according to their qualifications and functions. Medical masks are also known as surgical masks or face masks termed by U.S. Food and Drug Administration (FDA), or medical face masks termed by European Committee for Standardization (CEN) Technical Committee (TC) 205. In general, three nonwoven layers compose the most commonly used medical face masks. The middle layer is usually fabricated by the nonwoven meltblown process featuring electrostatic charging, and the two outer layers are usually fabricated by the nonwoven spunbond process [15]. Medical masks were originally designed to be used in the operation room, avoiding contaminants generated by the wearer on the wound and meanwhile preventing blood or other potentially infectious agents from reaching the wearer’s skin, mouth or mucous membranes (by splashes) [16]. Respirators, in this context, referring to filtering facepiece respirators (FFRs) or particle filtering half masks, are subject to various regulatory standards, such as N95 following the US standards NIOSH-42CFR84, FFP2 (Filtering Facepiece Particle Class 2) following the European standard EN 149-2001, KN95 following the Chinese national standard GB2626-2006, among others. FFRs are fabricated in a similar way as the medical face masks but composed of 3–5 layers of nonwovens through a range of processes such as meltblown, spunbond, drylaid, wetlaid and airlaid technologies as well as the advanced additive manufacturing method [17]. Additionally, the FFRs production involves the thermo forming and optimal fit for the final products. Along the time, new technology was introduced to produce more efficient medical masks or respirators for preventing respiratory infection, such as dimethyldioctadecylamonium bromide treated polypropylene (PP) with a positive charge to attract bacteria [18]. Not only is the effectiveness of new-generation RPDs considered, but also the comfortability and compliance of healthcare workers during their use shall be improved [19]. Both medical masks and respirators are designed to be of disposable use. However, under special circumstances (for example: if a sufficient supply of RPDs is not available during a pandemic), much local guidance considers the reuse of the RPDs [20,21]. For example, European Centers for Disease Control and Prevention has accepted the decontamination and reuse of respirators as options in case of a shortage of surgical masks and respirators since 2020 [21]. Additionally, the US Centers for Disease Control and Prevention declared the FFRs reuse, including reuse after decontamination, when there are known shortages of N95 respirators [22]. Regulatory bodies and Centres for Disease Control and Prevention of each country issued guidelines on the appreciative use of face masks under each condition thereby not covered by this review [23,24]. In view of the PPE shortage, this review specifically focuses on the RPDs (medical masks and respirators). The European testing standards of medical masks and FFP2 are presented, and testing standards of its equivalent respirators such as N95 (from the US) and KN95 (from China) are compared and discussed. Furthermore, the most commonly studied decontamination methods are summarised and discussed on the basis of their feasibility. Considering the global supply shortage of RPDs, the review has proposed possible decontamination methods for medical masks and FFRs, improving their reusability in pandemic situations such as the COVID-19.

2. Materials and Methods

There are three questions that the review attempted to answer:

1. What are the available RPDs in the market and their corresponding testing standards?
2. What are the possible decontamination methods for the reuse of the RPDs in the literature study?
3. What are the cutting-edge antimicrobial treatments on masks in research?

For the first question, the testing standards review started with EU standards issued by the European Committee for Normalization (CEN), Technical Committee 205 (TC 205). Other equivalent filtering facepiece respirators (such as from the United States, China, etc.) were compared and discussed. To answer the second and third questions, a comprehensive literature search focusing on the decontamination methods of used masks and the development of antimicrobial masks were conducted in the databases of PubMed and Google Scholar. Data were extracted by all authors based on the examination of the titles and abstracts obtained. Subsequently, full articles deemed necessary for the review were obtained and reviewed.

3. Results and Discussion

3.1. Testing Standards

The European standard for medical face masks is issued by CEN/TC 205—Non-active medical devices: EN 14683, following the directive 98/79/EC of the European Parliament and the Council concerning medical devices [25]. The standard not only details the construction, design and performance requirements of medical face masks but also indicates test methods of bacterial filtration efficiency (BFE) and breathability (differential pressure) [26]. The most important characteristics of the fiber-based masks are BFE and water resistance, according to the nature of their work. A higher BFE represents a higher level of protection for healthcare workers and patients against pathogen transmission. Water resistance indicates the resisting capacity of the material against splash or spray at various pressure. Based on their BFE, differential pressure, splash resistance (according to ISO 22609:2004) and bioburden (or microbial cleanliness in accordance with EN ISO 11737-1:2018), the medical face masks are classified into three types: Type I, Type II and Type IIR. Type I can only provide the minimum BFE protection and resist splash or spray at venous pressure, thus, it was not considered to be used for healthcare professionals. Both Type IIR and Type II can provide high BFE protection. Type IIR is more prior in use for healthcare workers than Type II due to its splash resistance feature.

The European standard for respirators is issued by CEN/TC 79—Respiratory protective devices: EN 149:2001+A1:2009 follows the directive 2016/425, 89/686/EEC of the European Parliament and the Council on personal protective equipment [6]. Standard EN 149:2001+A1:2009 has classified particle filtering half masks into FFP1, FFP2, and FFP3 based on their filtering efficiency and their maximum total inward leakage [27]. The standard specifies the minimum requirements and testing methods for particle filtering half masks, including design, leakage, breathing resistance, clogging, etc. Other test standards are necessary to refer to as the testing methods of the above-mentioned requirements, for instance: EN 143 Respiratory protective devices—Particle filters—Requirements, testing, marking; EN 13274-7, Respiratory protective devices—Methods of test—Part 7: Determination of particle filter penetration; ISO 6941, Textile fabrics—Burning behavior—Measurement of flame spread properties of vertically oriented specimens. It should be underlined that respirators are not recommended for general public use [28].

Comparing Type IIR medical face masks with respirators such as FFP2 and N95, respirators are compiled with more stringent test requirements. The highlight differences are that medical face masks do not require aerosol penetration test and face fitting test [29]. Therefore, when aerosol-generating procedures were performed, respirators instead of surgical masks were recommended [11,30]. One study with a focus on the penetration of aerosols properties of different masks found that quarantine masks fulfilled the requirements from the standard of the Korean Food and Drug Administration (KFDA) as KF 94 and the National Institute for Occupational Safety and Health (NIOSH) protocol as N95, while medical masks revealed more than 20% penetration values. These studies also indicated that using different protocols could result in a significant difference in some testing parameters (e.g., pressure drop) [31]. Other studies have compared the use performance of respirators and medical masks in HCWs against bacterial and viral infections. C. Raina
MacIntyre et al. have carried out a series of cluster randomized clinical trials. One of them compared the fit-tested and non-fit-tested N95 respirators with medical masks in terms of respiratory virus infection prevention in healthcare workers, which found that the rates of clinical respiratory illness (CRI), influenza-like illness (ILI), laboratory-confirmed respiratory virus and influenza infection were consistently lower for the N95 group compared to medical masks one [32]. Additionally, their study showed no significant difference in terms of CRI, ILI, laboratory-confirmed respiratory virus and influenza infection between the N95 arms with and without fitting test. Furthermore, another of their study revealed that continuous use of N95 respirators was more effective against CRI than medical masks [33]. It is also worth mentioning that the comparison between cloth masks and medical masks in their study revealed increased risks of infections with the use of cloth masks for healthcare workers [34]. However, in response to the masks shortages in the COVID-19 pandemic, the authors commented on the use of cloth masks as a last resort, whereas there are also other studies that found no significant difference between the use of medical masks and respirators in protecting HCWs against transmissible acute respiratory infections in clinical settings [35,36]. One randomized controlled trial performed in an Ontario tertiary care hospital showed the use of a surgical mask compared with a fit-tested N95 respirator by the nurses providing care to patients with febrile respiratory illness during the 2008–2009 influenza season resulted in noninferior rates of laboratory-confirmed influenza [37,38]. Another study revealed that N95 respirators vs. medical masks worn by participants when near patients with respiratory illness resulted in no significant difference in the incidence of laboratory-confirmed influenza [39]. It seems that the testing results vary depending on the testing methods and testing conditions. Despite all, the safe donning and doffing of respiratory protective devices is the key point for their maximum usefulness, especially in avoiding accidental self-inoculation [40].

The most commonly mentioned equivalent FFP2 respirators from other regulatory standards are N95 (United States NIOSH-42CFR84), KN95 (China GB2626-2006), P2 (Australia/New Zealand AS/NZ 1716:2012), Korea 1st class (Korea KMOEL—2017-64) and DS (Japan JMHLW-Notification 214, 2018). A technical bulletin from 3M has compared the above-mentioned respirators in terms of their filter performance, test agent, total inward leakage, pressure drop, etc. [41]. Based on their comparison and evaluation, the mentioned respirators are considered to be equivalent to each other and therefore are expected to function likewise to one another. Yet, the type of respiratory protection selection should respect the guidelines published by the CDC of each health authority and organisation [42].

3.2. Decontamination Methods

According to the standards, medical face masks and FFRs are designated as single use [43]. Manufacturers shall indicate the effective lifetime of the disposable masks or respirators. Incorporating a face shield, their lifespan may be prolonged due to reduced contaminates. Cleaning and sterilization are commonly performed for reuse of the elastomeric and powered air-purifying respirator [44–46]. However, for disposable medical masks and respirators, to mitigate the severe shortage and conserve the supplies during infection outbreaks and pandemic situations, decontamination and reuse of the disposable devices are considered and proposed by many guidelines of health authorities [20,21,47]. Additionally, the disposable masks or respirators become a water material after a single use, which is incinerated or disposed of in a landfill. The massive use of disposable fiber-based masks and respirators aggravates the problem related to their landfill management as waste. From the ecotechnological point of view, there is also an urgent need to reduce landfill disposal by recycling fiber-based masks and respirators [48]. As the life cycle assessment study of disposable and the embedded filtration layer (EFL) reusable face mask indicated that a lower emission of at least 30% in terms of CO₂-eq to climate change were contributed by using the EFL reusable face mask [49]. To reuse the disposable surgical masks and FFRs, three essential criteria need to be fulfilled: (i) remove the microbial thread; (ii) maintain the integrity of the various parts of the devices; (iii) remain no hazard residuals from the
decontamination process [50,51]. Many in vitro studies have been carried out, attempting to search for possible decontamination methods for the reuse of respiratory protective devices. Table 1 summarizes the decontamination methods proposed in the literature study. The investigated decontamination methods can be categorized into (1) energetic methods such as autoclave, dry microwave irradiation, ultraviolet germicidal irradiation (UVGI), microwave-generated steam (MGS), moist heat (MH); (2) chemical disinfection including 70% isopropyl alcohol, soap and water, bleach, ethylene oxide (EtO), hydrogen peroxide (vaporized and liquid forms), silver nanoparticles (AgNPs) and disinfecting wipes [52–55].

For example, Valdez-Salas et al. impregnated electrochemically fabricated AgNPs to the mask fibers utilizing ethanol aqueous solution (45% v/v ethyl alcohol) disinfectant formulation containing benzalkonium chloride [56]. The incorporation of the AgNPs into the surgical masks presented an outstanding antifungal activity against Candida albicans (C. albicans) and antibacterial activity against Escherichia coli (E. coli), Klebsiella pneumoniae (K. pneumoniae), Pseudomonas aeruginosa (P. aeruginosa) and Staphylococcus aureus (S. aureus). In addition, the disinfectant solution inactivated the activity of the H5N1 virus after 15 min disinfection time. In addition, this decontamination method for reusing surgical masks did not alter morphological, chemical and wetting characterization. However, aerosol filtration performance and mechanical integrities after disinfecting cycles need to be investigated. Additionally, the long-term stability and antimicrobial efficacy of AgNPs on the masks are evaluated. Nevertheless, this study supports the disinfection of medical textile masks with an economical and accessible approach to save resources and overcome the short supply of medical masks during the pandemic period. Overall, among all, UVGI, MH and vaporized hydrogen peroxide (VHP) appeared to be the most promising methods in balancing the three criteria of the removal of microbial thread, maintenance of structural integrity and function of FFRs and no harmful residuals [47]. It is noted that most research investigated only one aspect of microbial inactivation, filtration efficiency and structural integrity, including fitting tests [57]. It is difficult to obtain an evaluation of the influence of decontamination methods on all functions. Additionally, microbial inactivation assessment was mostly limited to one or two microorganisms or surrogates. It is vital to test the post-decontaminated FFR devices to see if they fulfil the required standards (EN143, NIOSH-42CFR84, etc.).

<table>
<thead>
<tr>
<th>Method</th>
<th>FFRs Type</th>
<th>Tested Virus</th>
<th>Other Tests</th>
<th>Comments</th>
<th>Ref.</th>
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<tbody>
<tr>
<td>Ultraviolet germicidal irradiation (UVGI), microwave-generated steam (MGS) and moist heat (MH)</td>
<td>N95 FFRs</td>
<td>H5N1</td>
<td>Molecular amplification assay, filter performance</td>
<td>Three methods effectively reduce viral laden on the N95 and also do not dramatically affect the filtration performance. Other tests regarding structural integrity are needed for further investigation</td>
<td>[58]</td>
</tr>
<tr>
<td>Autoclave treatment, ethylene oxide (EtO) gassing, ionized hydrogen peroxide (iHP) foggling and vaporized hydrogen peroxide (VHP)</td>
<td>4 different N95 FFR models</td>
<td>SARS-CoV-2 or vesicular stomatitis virus (as a surrogate)</td>
<td>Physical examination of structural and functional integrity, quantitative fit testing</td>
<td>All methods, which are commonly available in healthcare institutions, yielded effective decontamination performance. The response of structural and functional integrity change varies depending on the FFR models</td>
<td>[59]</td>
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<tr>
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<tr>
<td>Hydrogen peroxide vapor (HPV)</td>
<td>3M (St. Paul, MN, USA) 1860 N95</td>
<td>Biological indicator (Geobacillus stearothermophilus spores)</td>
<td>Off-gassing, odour, physical and performance degradation assessment, fitting test, facial structure check</td>
<td>HPV decontamination protocol is validated to effectively ensure the safe reuse of FFRs in real-world environments</td>
<td>[60]</td>
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<tr>
<td>Disinfecting wipes</td>
<td>Surgical N95 FFRs</td>
<td>Mucin and S. aureus</td>
<td>Particle penetration</td>
<td>3–5 log reduction after cleaning FFRs with disinfecting wipes but not considered as effectively decontamination, particle penetration fulfills the standard (&lt;5%) after cleaning despite the one with QACs wipe</td>
<td>[61]</td>
</tr>
<tr>
<td>MGS bags</td>
<td>FFR models pass the predefined quality standards</td>
<td>Bacteriophage MS2 (a surrogate for a pathogenic virus)</td>
<td>Filtration efficiency</td>
<td>99.9% effective for inactivating MS2 and filtration efficacy remain above 95% after treatment</td>
<td>[62]</td>
</tr>
<tr>
<td>MGS, warm moist heat (WMH), UVGI at 254 nm</td>
<td>6 commercially available FFR models</td>
<td>H1N1 influenza virus as aerosols or droplets</td>
<td>-</td>
<td>All three methods can reduce &gt;4 log of viable H1N1 virus, and in 93% of the experiment, virus was reduced to undetectable level; no assessment of the integrity structural change of the FFR models</td>
<td>[63]</td>
</tr>
<tr>
<td>H$_2$O$_2$ iHP</td>
<td>N95 FFRs</td>
<td>Influenza A virus (subtype H1N1)</td>
<td>-</td>
<td>iHP could kill influenza A virus at moderate to high levels of inoculum; Residual of H$_2$O$_2$ in the inner surface of N95 should be monitored; The integrity structural change of N95 was not assessed</td>
<td>[64]</td>
</tr>
<tr>
<td>HPV</td>
<td>N95</td>
<td>3 aerosolized bacteriophages (proxy for SARS-CoV-2)</td>
<td>-</td>
<td>One HPV cycle can eliminate phage to undetectable level; 5 cycles post decontamination result in no deformation.</td>
<td>[65]</td>
</tr>
<tr>
<td>UVGI</td>
<td>15 N95 FFR models</td>
<td>H1N1 influenza</td>
<td>-</td>
<td>Significant reduction in influenza viability under soiled conditions post-decontamination</td>
<td>[66]</td>
</tr>
<tr>
<td>UVGI (doses from 120–950 J cm$^{-2}$)</td>
<td>Four models of N95 FFRs</td>
<td>-</td>
<td>Flow resistance, bursting strengths of the individual respirator coupon layers and the breaking strength of the respirator straps</td>
<td>UVGI has a minor effect on filtration performance but noticeable decrease in the structural integrity; maximum limited disinfection cycles depend on the FFRs models</td>
<td>[67]</td>
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**Table 1. Cont.**

<table>
<thead>
<tr>
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</tr>
</thead>
<tbody>
<tr>
<td>UVGI, (EtO), VHP, microwave oven irradiation (MWI), and bleach</td>
<td>N95 FFRs, surgical N95 respirators, and P100 FFRs</td>
<td>Physical appearance, odour, laboratory performance (filter aerosol penetration and filter airflow resistance), dry heat laboratory oven exposures, off-gassing and FFR hydrophobicity</td>
<td>UVGI, EtO and VHP were found to be the most promising decontamination methods; the efficiency of the decontamination methods to inactivate viable microorganisms was not evaluated; infectious pathogen eradication was not assessed</td>
<td>[68]</td>
<td></td>
</tr>
<tr>
<td>Autoclave (160 °C dry heat), 70% isopropyl alcohol, soap and water (20-min soak), bleach, EtO, microwave oven, hydrogen peroxide (vaporized and liquid forms) and UV radiation</td>
<td>N95 and P100 FFRs</td>
<td>Filtration performance</td>
<td>Vaporized and liquid hydrogen peroxide and UV radiation appeared to have least effect on particle penetration performance; infectious pathogen eradication was not assessed</td>
<td>[69]</td>
<td></td>
</tr>
<tr>
<td>UVGI, MGS and moist heat incubation (MHI)</td>
<td>6 N95 FFR models</td>
<td>Fitting characteristics, odour, comfort and donning ease</td>
<td>No significant change in fitting, odour, comfort or donning ease with the six FFRs after UVGI, MHI or MGS decontamination</td>
<td>[70]</td>
<td></td>
</tr>
<tr>
<td>UVGI, EtO, hydrogen peroxide gas plasma (HPGP), HPV, MGS, bleach, liquid hydrogen peroxide (LHP) and MHI (pasteurization)</td>
<td>6 N95 FFR models</td>
<td>Physical appearance, odour and laboratory filtration performance</td>
<td>HPGP decontamination methods failed the filter penetration test requested by N95; FFR filtration efficiency of actual bioaerosols, as well as fitting tests after decontamination treatment, was not evaluated; decontamination method regarding its ability to inactivate infectious biological organisms is not tested</td>
<td>[71]</td>
<td></td>
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</table>

Unfortunately, investigation of FFRs decontamination and reuse mostly remains in the research phase apart from two studies at Nebraska Medicine and Duke University and Health System, of which the medical center of Nebraska initiated the application of the UVGI process for decontamination and reuse of the N95 FFRs in their institution, including a swell documented use guideline [72,73], yet it is indicated in the document that variation of material and condition, among others, may affect the effectiveness of the process, which should be carefully considered together with validation of the process before adopting any ones, while at Duke University and Health System, a hydrogen peroxide vapor-based FFRs decontamination protocol was evaluated, validated and further employed [60].

However, highlights from the literature summary of the decontamination procedure of disposable respiratory protective devices revealed: (i) a limited number of disinfection cycles should be performed to avoid the major loss of the FFRs function [67]; (ii) selection of decontamination methods requires careful consideration of FFR model, material type and design [74]; (iii) reusing numbers should be minimized to avoid the risks of infectious pathogen transmission during the donning and doffing of the equipment [75]; (iv) clear and
comprehensive guidelines should be developed to implement the decontamination protocol. However, as mentioned by European Centre for Disease Prevention and Control and other CDC authorities, respirators that have been visibly contaminated or are damaged or not fitting should be discarded and cannot be taken for reuse or decontamination procedures.

There are also other possible viral inactivation methods referred from other application areas, which can be considered for RPDs decontamination. For instance, Gamma irradiation has been commonly used for sterilization of medical devices and inactivation of infectious proteinaceous specimens in laboratory [76,77]; Ultraviolet C (UVC) is one of the dominant methods for surface decontamination in healthcare settings, which in combination with light and methylene blue (MB) plus visible light, has been deployed for inactivating Ebola virus (EBOV) and Middle East respiratory syndrome coronavirus (MERS-CoV) in platelet concentrates (PCs) and plasma [78], whereas evaluation of these methods on decontamination of PPE needs to be investigated. In addition, some researchers pointed out that reusing and recycling mask material are possible solutions to reduce mask waste for the need for circular economy [79]. The fiber material of masks is in the category of plastics (mostly used: polypropylene). Plastic waste recycling generally starts with shredding and sorting by composition and colour. Afterward, the separated plastics are melted and extruded into pellets for reuse [80]. This process can be applied for fiber-based medical mask recycling as well, though the financial cost is a crucial issue to consider.

3.3. Antimicrobial Masks

In addition to the decontamination of the masks after use, several studies have demonstrated strategies of incorporating functional agents (antimicrobial agents, superhydrophobic materials, electrical chargers, etc.) on masks and respirators in vitro that can reduce the risks of self-inoculation during doffing of the equipment [81–84]. To summarise, the mode of action of antimicrobial masks can be categorized into the followings: (1) superhydrophobic surface treatment avoiding pathogens’ adhesion and allowing mask self-cleaning; (2) incorporating of antimicrobial agents achieving self-disinfectant feature (chemical, photodynamic or photothermal antimicrobial actions); (3) extending the electrostatic charges of masks for sustained and improved pathogens filtration efficacy [85]. Among all, the second type of mode of action is the most employed one in the current research.

Antimicrobial agents such as quaternary ammonium compounds (QACs), metal ions (in the form of nanoparticles or nanowire, or nanorods) and natural plants extracts have been frequently utilized for the development of antimicrobial masks, exerting antibacterial or antiviral actions [86,87]. QACs are a common type of antimicrobial agent with broad-spectrum applications, which have been commonly applied in textile materials, including polyethylene terephthalate (PET), cellulose, polyamide, polypropylene (PP), etc. [88]. However, their low stability, non-adhesion or weak attachment on the substrate surface and potential toxicity result in decreased antimicrobial performance and wide application in practice [89]. In the study of Tuñón-Molina et al., a type of PET transparent mask coated with benzalkonium chloride (BAK) was prepared, which enabled to inactivate enveloped viruses (e.g., the phage phi 6 and severe acute respiratory syndrome coronavirus 2) in less than a minute of contact time [90]. The potent antimicrobial activity of the PET sample against viruses and bacteria was attributed to the positively charged nitrogen atoms of BAK, which can destroy the phospholipid bilayer, glycoprotein envelope and spike glycoprotein of the virus or destroy the bacterial membranes. The PET with BAK coating simple also showed antibacterial ability against MRSA and MRSE with inhibition zones of 0.61 ± 0.03 and 0.57 ± 0.05, respectively. It is unfortunate that the study did not investigate the safety issue regarding the use of these BAK coated masks in practice, such as the potential risks of respiratory exposure to this toxic compound. Similarly, Martí et al. developed the BAK bio-functional coating on a nonwoven mask filter by the dip-coating method, which exhibited >99% of SARS-CoV-2 particles reduction in just one-minute treatment [91]. In the study of Kumaran et al., terpyridine methylammonium chloride (TMAC) and adenine hexyl ammonium chloride (AHAC) were conjugated with lignin respectively to synthesize
lignin 2,2′,4′-terpyridine methylammonium chloride (LTMAC) and lignin adenine hexyl ammonium chloride (LAHAC) and cross-linked to form a permanent antimicrobial coating on the surface of the face masks in the form of spray or infiltration (Figure 1(Ai)) [92]. In the evaluation of the antiviral ability of LTMAC and LAHAC-coated PP face mask, human coronaviruses (alpha coronavirus: HCoV-229E and beta coronavirus: HCoV-OC43) were inactivated in 5 min and achieved 3–6 log reduction in 30 min (Figure 1(Aii–Av)). In addition, the LTMAC- and LAHAC-coated face masks significantly killed *K. pneumoniae* both in the medium composition of distilled water and artificial saliva and in the transmission modes of droplets and aerosols, which also showed a consistently time-dependent bacterial inactivation. However, the stability of the polymer coating on the mask surface needs to be verified.

Metal ions in the form of nanoparticles or nanowires or nanorods have been frequently utilized for the development of antimicrobial masks against various bacteria, fungi and viruses [86,93]. However, the weak coating adhesion between the metal particles (e.g., CuNPs, AgNPs, ZnNPs) and the textile substrate is the key issue hindering their further application in antimicrobial masks. In the study of Kwon et al., gallium liquid metal (LM) particles were used to increase the adhesion between liquid metal copper alloy (LMCu) particles and the fabric, which meanwhile achieved the reduction of Cu ions into metallic Cu by galvanic replacement (Figure 1(Bi)) [84]. After 20 min interaction between bacteria/fungi and LMCu coated fabric, significant cell death was observed (*S. aureus* (96.8 ± 4%), *E. coli* (99.7 ± 1%), and *C. albicans* (97.6 ± 4%)) (Figure 1(Bii)). Additionally, the LMCu coated fabric rapidly killed *S. aureus* in 10 s or even less. Moreover, LMCu coated fabric exhibited over 90% viral titer reduction in the antiviral test against prototype human coronavirus (HCoV 229E). Assisted with gallium LM particles, LMCu coating has a great potential for antimicrobial masks in the critical pandemic period. However, the toxicity assessment shall be performed to evaluate the potential risk of wearing these LMCu coated masks before their scaled application. In addition to the form of NPs, Cu has been produced into the shape of nanowires for a high surface-to-volume ratio. For example, in the study of Kumar et al., the surface of the blown polypropylene filtration media was dip coated with copper@ZIF-8 core-shell nanowires (Cu@ZIF-8 NWs), in which the Cu NWs were firstly passivated with the pluronic F-127 block copolymer for the following growth of ZIF-8 and meanwhile to prevent Cu NW degradation (Figure 1(Ci)) [94]. The Cu@ZIF-8 NWs exhibited lower cytotoxicity with three tested cells (A549 adenocarcinomic human alveolar basal epithelial cells, human gingival epithelial-like and primary gingival fibroblasts) in comparison to the bare CuNWs owning to the sustained and controlled release of copper ions (Figure 1(Cii)). In addition, Cu@ZIF-8 NWs showed more effective bactericidal activity against *Streptococcus mutans* (*S. mutans*) (86% inhibition) and *E. coli* (91% inhibition) than bare Cu NWs or ZIF-8, attributing to the synergistic antimicrobial effects between the Cu nucleus and the ZIF-8 shell (Figure 1(Ciii)). Additionally, Cu@ZIF-8 NWs demonstrated stronger antiviral activity in comparison with the positive control Remdesivir in the in vitro investigation (Figure 1(Civ)). However, the antiviral activity of the Cu@ZIF-8 NWs treated masks upon the exposure to virus-laden aerosols needs to be further explored as well as the general properties of masks in terms of the mechanical strength and filter efficiency.
Figure 1. (Ai). Schematic illustration of the UV cross-linked coating on the surface of the face mask. Gene expression level change of HCoV-229E (Aii,Aiii) and HCoV-OC43 (Aiv,Av) on lignin-based, LTMAC- (Aii,Aiv) and LAHAC- (Aiii,Av) coated face mask fabrics (spunbond: outer layer of SM, meltblown: middle layer of SM and spunbond-N95: outer layer of N95) (n = 3, mean ± SD) [92].

(Bi). Schematic illustration of the gallium-copper (LMCu) coatings on fabric by galvanic replacement.

(Bii). False coloured SEM images of S. aureus, E. coli and C. albicans cells on the control, Ga coated, and LMCu coated fabric (scale bars: 500 nm for S. aureus and E. coli and 2 µm for C. albicans): red arrows
indicated the physically damaged and perforated cells [84]. (Ci). Schematic illustration of the synthesis of core-shell Cu@ZIF-8 NWs. (Cii). Release profile of Cu$^{2+}$ and Zn$^{2+}$ from the Cu NWs and Cu@ZIF-8 NWs in cell media (n = 3. p < 0.05). (Ciii). Antimicrobial efficacy (percent reduction at OD 600 nm after 26 h) of Cu NWs, ZIF-8 and Cu@ZIF-8 NWs against S. mutans and E. coli at concentrations of 375 µg mL$^{-1}$. (Civ). Antiviral effects (percentage reduction) of Cu@ZIF-8 NWs and Remdesivir at 24 h and 48 h post infection [94].

Various natural plants extract exhibits strong antibacterial and antiviral properties, such as fructus arctii, sage, lycoris radiata, cinnamon and licorice, among others [95]. Son et al. prepared the AC + CO + PU antimicrobial nanofiber mat by polyurethane (PU) mixing solution activated carbon (AC) and cinnamon essential oil (CO) as the antibacterial agents via the electrospinning method, which exhibited a good inactivation effect of S. aureus and E. coli [96]. Researchers have found various compounds in licorice with antiviral and antimicrobial properties, especially 18-β glycyrrhetinic acid (GA) and glycyrrhizin (GL). In the study of Chowdhury et al., the licorice root extract that contains the antiviral substance glycyrrhetinic acid (GLR) was added to polyvinyl alcohol (PVA) solution to fabricate the bio-based filtration mask with a random porosity and orientation by electrospinning (nanofibers diameter ranging from 15 µm to 30 µm) (Figure 2(Ai)) [97]. The airflow rate of 85 L min$^{-1}$ (to maintain good breathability) can be reached with a pore size of 75 nm, which is smaller than the size of COVID-19 (Figure 2(Aii)). Additionally, the filtering efficacy was not affected when increasing the airflow rate in comparison with N95. However, the antivirus activity and the potential cytotoxicity of the licorice root-based masks need to be further evaluated. Moreover, the fluid-resistant and particulate filtration capacities need to be investigated for its final use as an antimicrobial face mask in practice.

Considering sustained and controlled antimicrobial action on face masks, researchers have proposed light-induced inactivation of pathogens upon near-infrared (NIR) light, Ultraviolet (UV) light or visible (Vis) light irradiation [98]. Photothermal and photodynamic modes of action act as a green and effective way to eliminate biological threats and generate heat or ROS to kill pathogens. In the study of Wu et al., the photodynamic BC-BPTCD-RF nanofibers (BBR-NFs) were produced by an esterification reaction between the carboxyl group of Benzophenone tetracarboxylic dianhydride (BPTCD) and the hydroxyl groups on bacterial cellulose nanofibers (BC-NFs), followed by grafting with Riboflavin (RF), which was further loaded through the high-pressure airflow onto the surface of the nonwoven fibers (Figure 2(Bii)) [99]. The BBR-NFs showed excellent antibacterial effects, achieving 99.999% and 99.9% contact-killing against E. coli and S. aureus by the sustained release of ROS owing to the forming of the intramolecular energy transfer channels and hydrogen transport after effective absorption of visible light (Figure 2(Biii)). In addition, the BBR-NFs exhibited excellent antiviral effects against a simulated virus T7 bacteriophage, achieving 5 log plaque-forming units (PFU) reduction both in 1 h under light conditions and in 90 min under dark conditions (Figure 2(Biii)). Similarly, Monmaturapoj et al. modified hydroxyapatite with anatase TiO$_2$ composite (HA/TiO$_2$ (HA50:Ti50) by solid-state reaction method, which showed an excellent antimicrobial effect [100]. HA/TiO$_2$ composite at 0.5 mg mL$^{-1}$ dose exhibited antiviral activity against the H1N1 influenza A virus, achieving more than 2 log/hour reduction of virus titer upon UV irradiation for 60 min because of the production of ROS (especially hydroxyl free radicals and peroxide) by TiO$_2$ particles upon UV light exposure, while for final application in practice, the persistence of the antiviral effects against the SARS-CoV-2 virus as well as the filtration efficacy shall be further verified. Furthermore, researchers have also attempted to combine several antimicrobial modes of action together for enhanced/synergistic antimicrobial activity. In the study of Kumar et al., shellac/copper nanoparticles (CuNPs) were coated on the PP nonwoven surgical masks by dual-channel spray method, where the bio-adhesive shellac bonded tightly to the antimicrobial CuNPs, increasing the hydrophobicity and photoactivity of the surface for self-cleaning property (Figure 2(Ci)) [101]. The nanocoated photactive antiviral masks (PAM) exhibited substantial E. coli MG1655 reduction (~4 log) under the sunlight for 5 min, attributing to
the generation of free radicals by the rapid rising of the mask surface temperature over 70 °C (Figure 2(Cii,Ciii)). Additionally, the concentration of extracellular vesicles as the model of COVID-19-virus-like particles (VLPs) decreased by 2–3 log on the PAM under sunlight for 5 min owing to the self-cleaning ability of PAM. Overall, the PAM showed excellent photocatalytic and self-cleaning activity, providing a pragmatic solution in the critical pandemic situation.

Figure 2. (Ai) Schematic illustration of the production of biobased antiviral face mask by electrospinning process. (Aii) Estimated airflow rate through the licorice membrane with a varying pore size [97]. (Bi) Schematic illustration of the fabrication of BBR-NFs and their encapsulation with the nonwoven fibers. (Bii) E Bactericidal activity of BBR@protective suit against E. coli and S. aureus under visible light irradiation (a) and dark conditions (b). (Biii) Viricidal assay against T7 phage for BBR@protective suit (a) and BBR@mask (b) under visible light irradiation and dark conditions [99]. (Ci) Schematic illustration of the virus inactivation in respiratory droplets through photothermal, photocatalytic and hydrophobic self-cleaning processes under solar irradiation. (Cii) Colony-forming unit (CFU) number of viable E. coli after solar illumination treatment on the surface of the control, PAM and raw surgical mask (data are presented as means ± SD and n = 3). (Ciii) Increase in the CAM and Mask surface temperature as a function of solar illumination time (data are presented as means ± SD and n = 3) [101].

The development of antimicrobial masks can indeed extend the lifespan of medical masks. The up-to-date developed antimicrobial masks in literature are summarized in Table 2. In combination with decontamination of the used masks, these strategies can potentially solve the global shortage of masks during the pandemic. Nevertheless, in addition to possible protocols for reuse of the medical masks or respirators, good training and guidance for proper reuse of them are fundamental to ensure their efficiency in infection prevention and control (IPC) [24,102].
Table 2. Strategies for the fabrication of antimicrobial masks.

<table>
<thead>
<tr>
<th>Coating Method</th>
<th>Antimicrobial Agents</th>
<th>Mode of Action</th>
<th>Tested Microorganisms</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polyethylene terephthalate transparent masks dip-coated with benzalkonium chloride</td>
<td>The positively charged nitrogen atoms of BAK</td>
<td>Chemically antimicrobial action</td>
<td>MRSA and MRSE</td>
<td>[90]</td>
</tr>
<tr>
<td>Cross-linked to form permanent antimicrobial coating on the surface of the face masks</td>
<td>2,2′,4′-terpyridine methylammonium chloride (LTMAC) and lignin adenine hexyl ammonium chloride (LAHAC)</td>
<td>Chemically antimicrobial action</td>
<td>Alpha coronavirus: HCoV-229E, beta coronavirus: HCoV-OC43 and K. pneumoniae</td>
<td>[92]</td>
</tr>
<tr>
<td>The deposition of liquid metal copper alloy (LMCu) particles on the fabric by spontaneous galvanic replacement reaction</td>
<td>The reduction of Cu ions into metallic Cu by galvanic replacement</td>
<td>Chemically antimicrobial action</td>
<td>S. aureus, E. coli, C. albicans and prototype human coronavirus (HCoV 229E)</td>
<td>[84]</td>
</tr>
<tr>
<td>The surface of the blown polypropylene filtration media was dip coated with copper@ZIF-8 core-shell nanowires</td>
<td>The synergistic antimicrobial effects between the Cu nucleus and the ZIF-8 shell</td>
<td>Chemically antimicrobial action</td>
<td>S. mutans and E. coli</td>
<td>[94]</td>
</tr>
<tr>
<td>Glycyrrhetinic acid was added to polyvinyl alcohol solution to fabricate the biobased filtration mask by electrospinning</td>
<td>Glycyrrhetinic acid</td>
<td>Chemically antimicrobial action</td>
<td>N.a.</td>
<td>[97]</td>
</tr>
<tr>
<td>BC-BPTCD-RF nanofiber was loaded through the high-pressure airflow onto the surface of the nonwoven fibers</td>
<td>BC-BPTCD-RF nanofibers</td>
<td>Photoactive antiviral (combined photocatalytic and photothermal properties)</td>
<td>E. coli, S. aureus and simulated virus T7 bacteriophage</td>
<td>[99]</td>
</tr>
<tr>
<td>Shellac/copper nanoparticles (CuNPs) were coated on the polypropylene masks by dual-channel spray method</td>
<td>The generation of free radicals by the rapid rising of the mask surface temperature over 70 °C under the sunlight</td>
<td>Light-induced inactivation upon irradiation with near-UV and visible light</td>
<td>E. coli and extracellular vesicles</td>
<td>[101]</td>
</tr>
</tbody>
</table>
4. Conclusions and Perspectives

The importance of using PPE to protect HCWs from pathogen exposure during a pandemic should not be underscored. When vaccination is not possible, along with other essential prevention measure such as hand hygiene, the use of PPE protects HCWs from SARS-CoV-2 exposure during patient care in the COVID-19 era. However, respiratory protective devices are likely in shortage during public health emergencies. Decontamination and reuse of disposable surgical masks and respirators are proposed by health authorities during the pandemic period. Decontamination methods such as ultraviolet germicidal irradiation (UVGI) and moist heat (MH) vaporized hydrogen peroxide (VHP) appeared to be the most promising methods in balancing the three criteria of the removal of microbial thread, maintenance of structural integrity and function of FFRs and leaving no harmful residuals. Nevertheless, several limitations were found in the current research studies regarding the decontamination of FFRs:

1. Limited microbial contaminants used in the assay (H1N1, H5N1, MS2 phage, S. aureus and biological indicators);
2. Lack of comprehensive testing of the microbial removal, FFRs structural integrity and function change after decontamination procedure;
3. Results vary based on the FFR models, materials, and designs, as well as testing protocols.

Further investigation is required regarding the decontamination and reuse of disposable surgical masks and respirators during public health emergencies. The approval of the validated protocols needs to be evidence-based and specific event oriented. The priority choice of FFRs for healthcare workers are fitting tested FFRs > non-fitting tested FFRs; extended time use of FFRs > reuse of decontaminated FFRs; non-fitting tested and post-decontaminated FFRs should be only used in a low-risk workflow. Emphasis on good training and guidance for proper reuse of RPDs are encouraged and fundamental to ensure RPDs efficiency in protecting front-line healthcare workers during patient care. In addition, new technology such as antimicrobial treated RPDs provides an encouraging outcome in reducing the risks of self-inoculation of healthcare workers during donning and doffing of the protective devices as well as extending the lifespan of the RPDs.

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