Since January 2020 Elsevier has created a COVID-19 resource centre with free information in English and Mandarin on the novel coronavirus COVID-19. The COVID-19 resource centre is hosted on Elsevier Connect, the company's public news and information website.

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Current perspective in metal oxide based photocatalysts for virus disinfection: A review

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ABSTRACT

Nanotechnology holds huge potential for the prevention of various viral outbreaks that have increased at a disquieting rate over the past decades. Metal oxide nanomaterials with oxidative capability are the effective materials that provide platforms as well as tools for the well understanding of the mechanism, its detection, and treatment of various viral diseases like measles, influenza, herpes, ebola, current COVID-19 etc. In this inclusive review, we survey various previous research articles on different notable photoactive transition metal oxides that possess enough potential to act as antiviral agents for the deactivation of harmful viruses. We investigated and highlighted the plausible photocatalytic oxidative mechanism of photoactive transition metal oxides in degrading viral coatings, genomic RNA using suitable free radical generation. The key finding of the present review article including the discovery of a vision on the suitable photocatalytic transition metal oxides that have been proven to be excellent against harmful viruses and consequently combatting deadly CoV-2 in the environment. This review intends to provide conclusive remarks and a realistic outlook on other advanced photocatalytic metal oxides as a potential solution in battling other similar upcoming pandemics.

1. Introduction

Different microbial pathogens like viruses, bacteria, protozoa, etc. present in the environment may sometimes threaten human health, and cause dangerous infectious illnesses (Budlayan et al., 2020; Gall et al., 2015). Specifically, among various kinds of pathogenic contaminations viruses have always been regarded as increasing hazards owing to their small-sized particles, challenging inactivation, and extraordinary environmental endurance (Habibi-Yangjeh et al., 2020; Qiu et al., 2017). A virus is a small intracellular parasite having either ribose or deoxyribose nucleic acid as genetic material that is surrounded by a protective protein layer (Pemmada et al., 2020; Lee et al., 2014). Viruses are ubiquitous that fail to survive outside the host organism and their contact with a human may impair health and can lead to extreme illnesses and other ailments. Contaminants associated with water, air, and food have become a notable threat owing to their adverse effects on the environment and health (Howell and D'Souza, 2013; Anderson et al., 1990). Food, water, and air-borne viruses enter the body of the host through various modes of infection and cause millions of deaths all over the world annually. Serious human pandemic has a great impact on the dynamics of rules and human's lifestyle that cause negative social effects, and massive global scale economic damage (Lampertico et al., 2005). Air and waterborne outbreaks correlated with environmental changes as well as disturbances in the ecosystem.

From the acute illness up to the novel contagious disease, scientists and researchers worked together to provide means of containing, detecting, preventing and curing these biological hazards. Among the known diseases, infections triggered by viruses have been labelled as one of the most strenuous and difficult to handle due to their infectious

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Abbreviations

AOPs	Advance oxidation processes
COVID-19	CoV-2 disease
HAdV40	Human adenovirus-40
LED	Light emitting diode
MERS	Middle east respiratory syndrome
RNA	Ribose nucleic acid
SEM	Scanning electron microscopy
SARS-CoV-2	Severe acute respiratory syndrome of coronavirus
SDSP	Sodium dodecyl sulfate polyacrylamide electrophoresis
TEM	Transmission electron microscopy
UV–Vis	Ultraviolet–visible spectroscopy
WHO	World health organization

and proliferative nature (Brunelle et al., 2005; Hasija et al., 2021; Lee et al., 2006; Wong et al., 2004). More importantly, viruses like herpes, hepatitis, dengue, influenza, measles, human immunodeficiency virus, gastroenteritis virus, etc. replicate and multiply in a huge number by infecting a host cell, and cause serious illness. Spiraling worries about different pandemics like dengue haemorrhagic fever in tropical, and subtropical countries, Spanish flu outbreak in 1918, influenza pandemics *via*. H1N1 and H5N1 (every 10–15 years), severe acute respiratory syndrome and middle east respiratory syndrome-related coronavirus i.e. SARS-CoV, MERS-CoV, in 2003, and 2014 respectively drawn attention of many researchers for viral treatment (Anderson et al., 1990; Zhong et al., 2003; Hasija et al., 2019; Schiff et al., 2007). Since 2019 (December), the whole world is facing a first-time medical emergency due to the arrival of a novel deadly new strain i.e. severe acute respiratory syndrome (SARS) of coronavirus CoV-2 (SARS-CoV-2) (Fauci et al., 2020).

Enveloped SARS-CoV-2 belongs to the largest genome of 26–32 kb size as shown in Fig. 1a. Under an electron microscope, the morphological structure of CoV-2 is observed as crown-shaped, containing single-stranded RNA as genetic material inside helix capsid (Fig. 1b) (Nasir et al., 2021). Fig. 1 c and d depicting the magnified structures of SARA-CoV-2, whereas e and f are the scanning and transmission electron microscopic images (SEM and TEM), respectively. Various essential and non-essential structural proteins encoding RNA genomes like a spike (S), membrane (M), nucleocapsid (N), envelope (E) are vital for constructing complete virus particles (Nasir et al., 2021; Benameur et al., 2021; Tabibzadeh et al., 2021). Spike protein facilitates the host-virus connection and subsequent fusion of their cellular membranes to accelerate the viral entry into the host. Nucleocapsid in SARS-CoV-2 is the only protein that primarily binds to the RNA genome and thereby forming a whole nucleocapsid unit. SARS-CoV-2 rapidly multiply inside the host cytoplasm, endoplasmic reticulum, and Golgi-apparatus, indicating that the host membranes and organelles also equally contributing for its replication.

The presence of harmful viruses in wastewater confirmed its waterborne transmission in the areas where there is no adequate water treatment or sanitation facilities. As these enveloped viruses are highly stabilized in the favorable environmental conditions (Fig. 2), and may be transmitted through direct contact with exposed contaminated surfaces (doors, handles, mobile phones, other household items), inhalation of respiratory droplets (sneezes, coughs, exhale), through aerosols and faecal matter (faecal-oral transmission) of an infected patient (Teymoorian et al., 2021). Deadly CoV-2 is rapidly transmitted among human societies (public transportations, factories, prisons, nursing homes, etc.) as compared to SARS-CoV and MERS-CoV (Cevik et al., 2020; Meyerowitz et al., 2021). Research reports reported that even the CoV-2 negative (respiratory tract) patients contain virus in their stools for a long period. Consequently, SARS-CoV-2 can be found in the outer environment, mainly in the urban H_2O cycle, which affects the existing

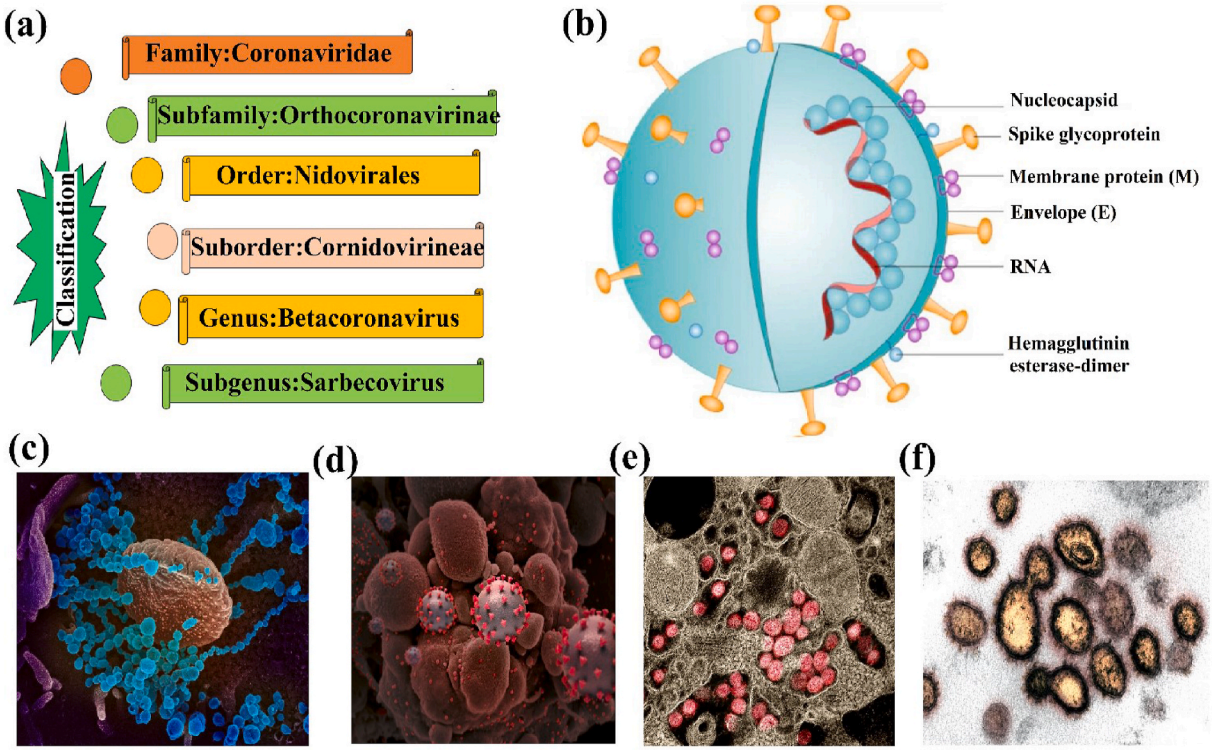

Fig. 1. (a) Classification of SARS-CoV-2; **(b)** Schematic diagram of SARS-CoV-2 structure (Nasir et al., 2021), Adapted with permission from Elsevier (license No. 5121780134344); **(c** and **d)** The magnified structures of SARA-CoV-2, Adapted with permission (License no. CC BY 2.0); **(e** and **f)** SEM and TEM images of SARS-CoV-2, Adapted with permission (License no. CC BY 2.0).

Fig. 2. The possible water contamination passageways during the pandemic like COVID-19, Reprinted with permission from Elsevier (license No. 5121830840322) (Teymoorian et al., 2021).

cycles in the environment as well as the life of aquatic organisms (La Marca et al., 2020).

Antiviral drugs, passive immunization, heat treatment, several inactivation agents (antiseptic solutions), ultraviolet irradiation, various vaccines, chemical compounds, and medicines, are different preventive measures to reduce the transmission and infections of viruses including CoV-2 (Teymoorian et al., 2021; Huang et al., 2020; Yang et al., 2020; Kumar et al., 2020a). UV radiations and toxic chemical compounds cannot be applied in the presence of human beings, and thus most of the physicochemical methods are associated with certain drawbacks. In order to overcome the inherent shortcomings associated with standard techniques, several effective nanotechnology-based methods have evolved for virus inactivation like H1N1, AIDS virus (human immune virus), CoV-2, etc. (Teymoorian et al., 2021; Hasani et al., 2021). Advanced oxidation processes (AOPs) are highly advanced technology that generates oxidative and reductive agents like hydroxyl radicles to treat toxic pathogens (Nasir et al., 2021; Kokkinos et al., 2021). Generated reactive radicles directly and effectively deteriorate the viruses. Ozonation (O_3 in water), chlorination, chlorine gas, chloramines, ultrasound, Fenton process, photocatalysis, etc. are the various known AOPs that are efficiently used for treating wastewaters containing viruses (Patel et al., 2020; Snell, 2020; Ji et al., 2020). These AOPs are associated with certain limitations like the formation of –CHO, –COOH, Br, etc. by-products (ozonation), high energy consumption and instrumentation (ultrasound), explosions, fires, gas leak, unpleasant tastes, and odour's (chlorination, chloramines), high consumption of H_2O_2, formation in sludge (in Fenton process) (Nasir et al., 2021; Teymoorian et al., 2021; Mattioli et al., 2020). Among AOPs, photocatalytic disinfection is the most efficient technique for wastewater treatment without forming any harmful by-products (Fig. 3a) (Truong et al., 2021). As solar light is a freely abundant, and obtainable source of energy, an effectual photocatalytic material should be capacitated with harnessing an extensive range of the solar spectrum ranging from ultra-violet to near-infrared light (Truong et al., 2021). The utilization of

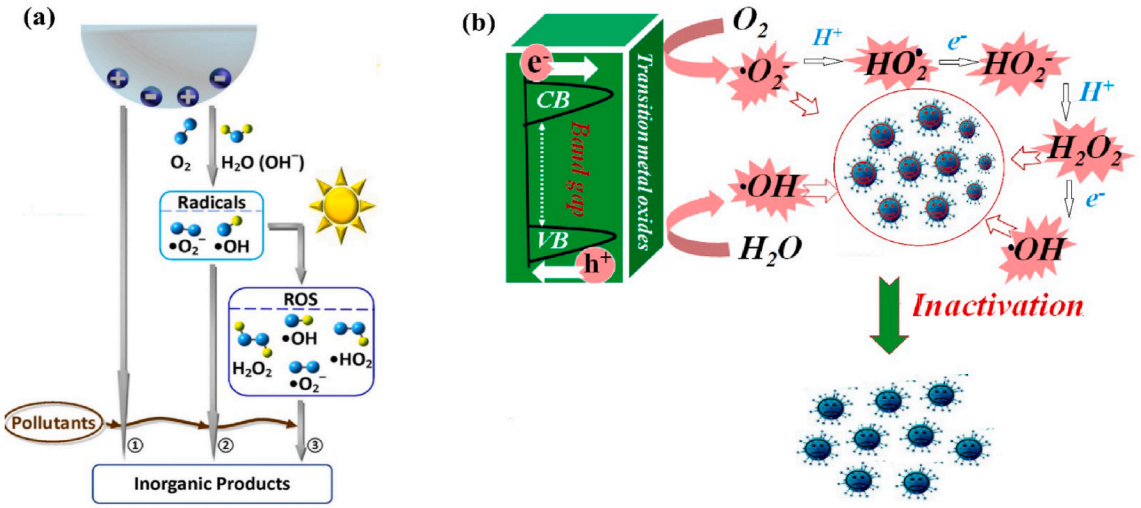

Fig. 3. (a) Schematic diagram of radical generation mechanisms for the photocatalytic degradation of harmful pathogens, Adapted with permission from Elsevier (license No. 5121780790083) (Truong et al., 2021); **(b)** Mechanism of inactivating virus process through photocatalysis, Adapted with permission from Elsevier, (license No. 5121780134344) (Nasir et al., 2021).

photocatalysis with bare or promoted photoactive materials has been suggested as an operative tool for surface self-sterilization under mild conditions.

Photocatalytic materials that require light as an energy source either from fluorescent or solar irradiation are considered as the most suitable way to inactivate enveloped viruses (Fig. 3b) (Nasir et al., 2021; Lu et al., 2019a, 2022; Li et al., 2021; He et al., 2019). The photocatalytic reaction takes place under the combined action of solar light with suitable frequency and the photoactive material that absorbs a wide range of solar spectra (Soni et al., 2022a, 2022b). Typically, by acquiring an excited state after the absorption, single e^- migration event triggers to produce active e^-/h^+ pairs in the conduction band (CB) and valence band (VB) of photoactive semiconductor, respectively (Soni et al., 2021a; Lu et al., 2019b, 2020). Isolated e^- in the CB can reduce dissolved O_2 and produce H_2O_2, or $\bullet O_2^-$ along with H^+ ions, whereas, remaining h^+ in the VB can oxidize the OH^- ions from water to generate unstable •OH radicals (Soni et al., 2021b, 2022a; Wang et al., 2021). These generated reactive oxygen species (ROS; •OH, H_2O_2, H^+, and $\bullet O_2^-$) during photocatalysis having redox potential participates in photocatalytic disinfection and degradation processes (Soni et al., 2021c; Kumar et al., 2021a). The photocatalytic performance mainly depends upon the type of potential photocatalyst used, their physical and chemical properties, and the light source (Kumar et al., 2021a, 2021b; Raizada et al., 2020).

Viruses show a significant heterogeneity in term of size, morphological structure, biologically properties, and in their interaction behaviour with environment. Generally, nanoscopic antiviral agents biochemically interacted with the nano-viruses, are mainly categorized into two types; virostatic and virucidal (Akhavan et al., 2012; Guillard et al., 2008). The virostatic agents are accelerated in the very early stages of virus infection to reduce the RNA replication rate and their functioning is centered on the binding mechanism of surface proteins. However, virucidal agents are the agents that have the potential to deactivate the nanovirus permanently and almost completely (Rabiee et al., 2020; Zan et al., 2007; Nakano et al., 2012). Photoactive metal oxides are much potentially lethal for various viruses by disrupting proteins, genomic RNA, cell membrane etc. *via.* generating unstable radicals even at low concentration (Zhang et al., 2019). In the last years, the attention was focused on the development of nanostructured photoactive metal oxides with the development of various physicochemical and eco-friendly approaches to obtain photoactive metal oxides nanoparticles and heterocomposite (Giannakis et al., 2017; Sudhaik et al., 2018). Co-Precipitation, microemulsion, hydrothermal, sonochemical, and sol-gel techniques are generally used for the preparation of transition metal oxides (Mondal and Sharma, 2016; Kumar et al., 2020b; Kato et al., 2019; Mondal, 2017; Munawar et al., 2020; Subhan et al., 2015; Guo et al., 2011). Fabricated photocatalytic transition metal oxides nanoparticles with different morphologies exhibit remarkable growth inhibition of harmful viral species. In this regard, among various reported photocatalytic materials certain reports assessed the ability of transition metal oxides like TiO_2, WO_3, CuO, ZnO, SnO_2 etc. against enveloped and non-enveloped viruses including CoV-2 (Habibi-Yangjeh et al., 2020; Yemmireddy and Hung, 2017; Karimi-Maleh et al., 2020; Sharma et al., 2019; Erkan et al., 2006; Ditta et al., 2008). Also, various metal oxides (TiO_2, ZnO) act as an excellent packing materials in various photoreactors for bioaerosol disinfection with high filtration capacity, and reusability. It is valuable that photoactive transition metal oxides exhibited superior antiviral activity owing to their remarkable surface characteristics, microstructural features, high surface area and other superior physio-chemical properties (Djurišić et al., 2014). Noting that transition metal oxides are considered as potential photocatalytic material owing to their apt physicochemical properties, cost-effectiveness, stable nature, and thermodynamically favorable band edge potentials for various photocatalytic applications. Moreover, they have remarkable recycling rate and good stability rate (Danish et al., 2020).

Subsequently, here we impose the importance of nanomaterial-based photocatalytic materials in various possible applications of the fight against toxic viruses. There are certain review papers already published on the photocatalytic inactivation of virus but there is no review report depicting the virus inactivation using photoactive transition metal oxides. Scopus data shown the total number of research articles published from the year 2012–2021. These research reports were used for the literature review through the Elsevier, Springer, Wiley, RSC and many more publishers. This perspective review aimed to explore the antiviral activity of various photoactive transition metal oxides against toxic viruses, including SARS-CoV-2 using visible light in order to prevent viral infection. The properties like high surface area and other superior physico-chemical properties of photoactive metal oxides are taken advantage to interfere with the activity of the toxic virus which may bring about the mitigation of viral infestation. It will start shedding light on the primary potential mechanistic studies known to date that are accountable for virucidal deactivation. Finally, the transition metal oxides which are widely explored for their antiviral nature would be necessarily highlighted. The published research reports demonstrated

that transition metal oxide-based heterojunctions can be capably used as a virucidal agent in the liquid or gas-phase, that successfully deactivated up to 20 different viral strains out of the 25 tested, including deadly CoV-2 (both on surface and air), as proved in latest below mentioned 4 research reports. With all the gathered results, we expect that this present review paper will inspire the development of different antiviral photoactive nanomaterials to control the wastewater and aerosol transmission of various viruses. To the best of our knowledge, this systemic review paper is first to provide insights in viral inactivation using photoactive metal oxides semiconductor for sustainable water or air purification.

2. Photocatalytic performance evaluation on virus disinfection

Virus deactivation using photocatalytic material could possibly overcome the limitations of existing conventional methods. In principle, disturbing the outer cell membrane, destroying functional as well as structural proteins, and genetic RNA by the generation of reactive oxidation species, inhibited the growth of virus and may lead to cell lysis. Potential photocatalytic materials as antiviral agents are the best alternative solution to battle the pandemic like COVID-19 (Nasir et al., 2021; Truong et al., 2021). Among photoactive transition metal oxides, non-toxic, economical, and profusely available TiO_2 photocatalyst has exposed the high potential for deactivation of phage MS2, bacteriophage, phage f2, HIV, norovirus, etc. (Bayarri et al., 2021; Bishai, 2021). UV active commercial TiO_2 photocatalyst was the first among transition metal oxides to perceive the virus inactivation owing to its disinfection potential towards genetically similar SARS-CoV-2 and HCoV. Destruction of the shell or the capsid of viruses resulting from the release of RNA, minerals, structural proteins inside the viruses and cause their inactivation. Photocatalytic semiconductor material in the presence of solar light produced e^-/h^+ pairs to form unstable radicals that rapidly undergo oxidation and reduction reactions (Ornstein et al., 2020; Yoshizawa et al., 2020).

Photoactive metal oxides (ZnO, CuO, TiO_2, Fe_2O_3, MgO, etc.) being excellent semiconductors are prominently studied as antimicrobial and antiviral agents having outstanding results (Habibi-Yangjeh et al., 2020; Zhang et al., 2019; Ditta et al., 2008). Anti-pathogenic potential of metal oxides is extended to harmful viruses like hepatitis C, polio, H1N1 influenza, herpes, SARS-CoV, SARS-CoV-2, etc. For example, spherical Cu_2O nanoparticles with 40 nm size were effectively reported to inhibit the growth of herpes simplex virus up to 83.3% at 100 μg/mL concentration. Mechanistically, release of Cu ions from the Cu_2O catalysed the generation of reactive radicals that induced damage on the outer capsid layer of virus and ultimately degraded its genome at non-cytotoxic dosage. It was assumed that interfering of photocatalytic metal oxides

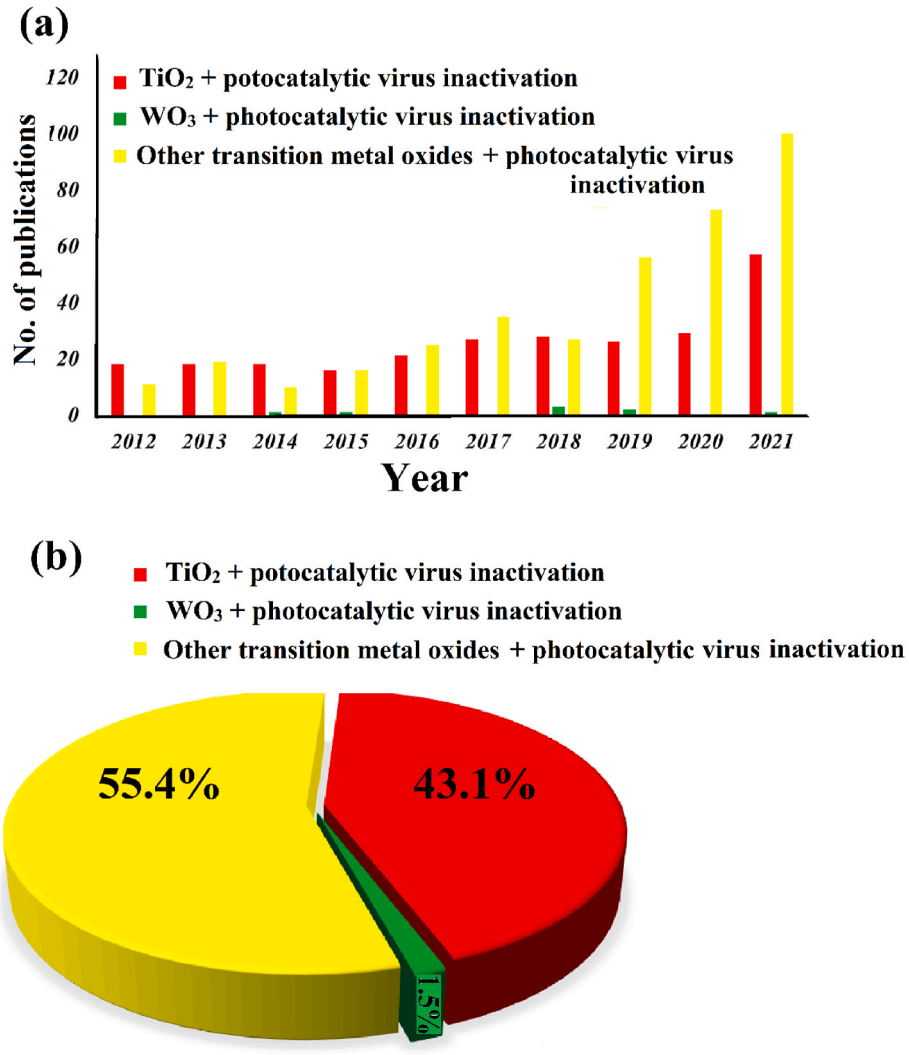

Fig. 4. (a) Bar graph representing Scopus data analysis results for a number of publications from the year 2012–2021, using keywords "TiO_2 + photocatalytic virus inactivation; WO_3 + photocatalytic virus inactivation; Other transition meta oxides + photocatalytic virus inactivation," **(b–d)** pie chart shows the proportion of active TiO_2, WO_3, and other transition metal oxides photocatalyst for virus inactivation, respectively.

in one or two steps during viral replication process generally inactivates the viral particles (Tavakoli and Hashemzadeh, 2020). In another report, spherical cuprous oxide nanoparticles degraded hepatitis C virus by blocking viral attachment to the cell which mitigated further infection on the hepatic cells (Hang et al., 2015). Similarly, Mazurkova et al. studied the interaction of prominent photoactive TiO_2 nanoparticles (4–10 nm) with H3N2 influenza virus that required almost 30 min for its degradation. TiO_2 may degraded viral lipids, glycoproteins spike present in the envelop to destroy virus-host attachment mechanism (Mazurkova et al., 2010). Although there was no such detailed elaboration on lipids, glycoproteins degradation. Therefore, from all these reports it is indicated that potential metal oxides effectively used as antiviral agents against various toxic viruses.

Herein, the photocatalytic inactivation mechanism of the deadly viruses occurred on the outer surface of the film and can be described by degradation of the capsid as well as viral RNA with the help of unstable free radicals (Fig. 5a) (Truong et al., 2021). Catalyst generates $\bullet O_2^-$, •OOH, and •OH active ROS in the presence of solar light and actively degrade the components of viruses present on its surface (Truong et al., 2021; Bayarri et al., 2021). These generated active radical oxidation species degrade the components of the virus through oxidation damage under solar light illumination (Nasir et al., 2021; Rodriguez-Morales et al., 2020). Mechanistically, the radicals first attack the outer layer comprise of phospholipids, proteins (host), and viral glycoproteins (to identify receptors site on the host membrane). As a result, damaged cell membrane (coenzyme A) disturbed the cellular respiration, other biochemical systems, and ultimately cause cell lysis (Ghotekar et al., 2021; Bandala et al., 2021). Depending upon the complexity of the virus structure, the comprehensive degradation mechanism involves; (a) the peroxidation of proteins, lipids by radicles that depresses the periplasmic enzyme activity and damage the genetic RNA material; (b) release of metal ions (photocatalyst) into the cell that interacts with COOH, –NH, –SH like functional groups to damage nucleic acids; and (c) disturbing the cellular metabolism without the induction of oxidative stress (Li et al., 2020a; Ghaemi et al., 2021). Thus, understanding the deactivation mechanism of viruses might be helpful to construct more superior potential photocatalytic materials with apt morphology and practical importance.

3. Various potential reported transition metal oxides as antiviral agents

Researchers analyzed that viruses are more rapidly transmitted through air rather than direct contact or water droplets. Thereby, it is crucial to understand the mode of aerosols and surface contaminations by viruses in order to plan operative preventive processes to curb the

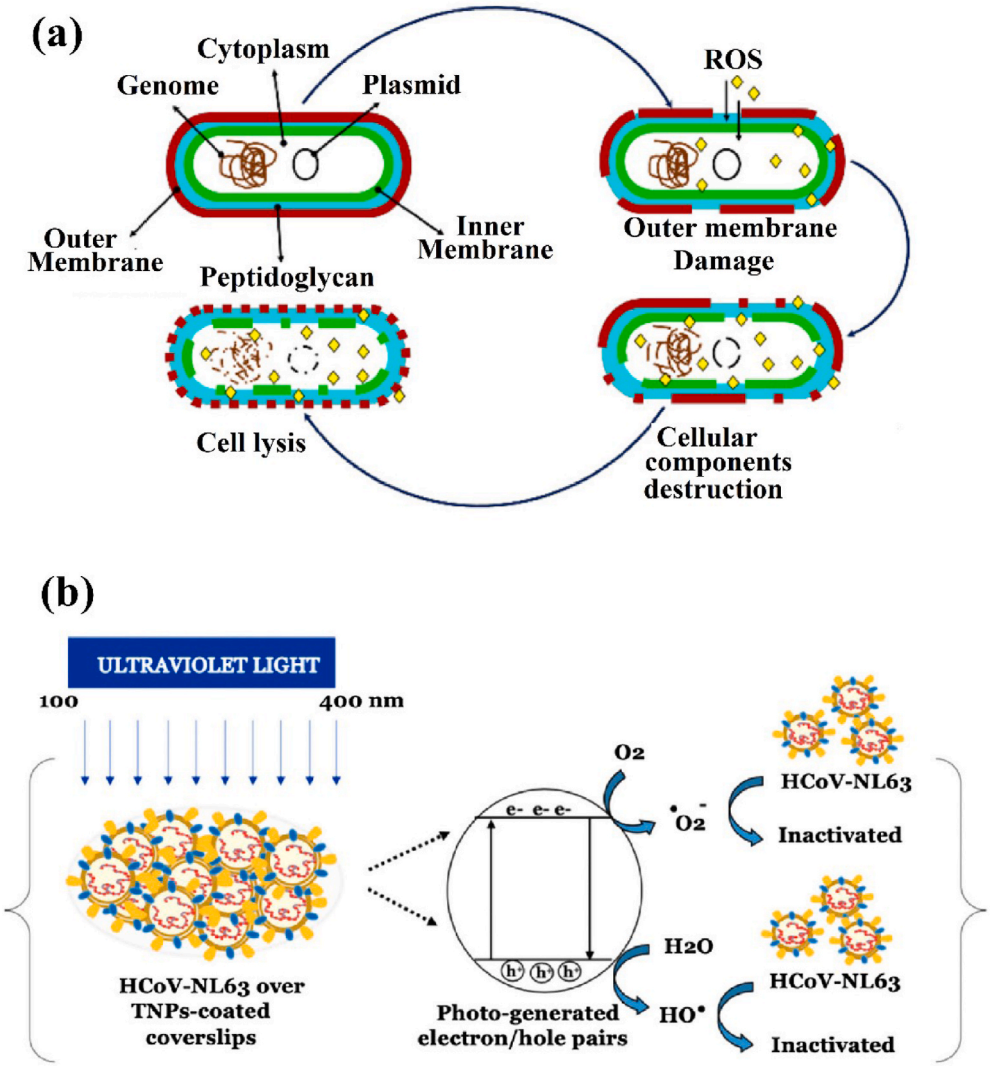

Fig. 5. (a) The mechanism of photocatalytic disinfection (Truong et al., 2021), **(b)** The mechanism of virus inactivation, Reprinted with permission from Elsevier (license No. 5121780790083) (Khaiboullina et al., 2021) (Truong et al., 2021).

increase in viral infections (Nasir et al., 2021; Barouki et al., 2021). Research studies analyzed that the stability of viruses like SARS-CoV-2 is 3 h approximately in aerosols, 72 h in surfaces like plastics, stainless steel, cardboard. Inactivation of viruses present in the environment is vital for preventing its transmission rate. In this section, study of various several transition metal oxides-based photocatalytic heterojunctions are reported to investigate their antiviral characteristics (Li et al., 2020b; Miyauchi et al., 2020). And, it is scrutinized that the photocatalytic performance of catalyst is mainly dependent on the properties like surface area, structural morphology, bandgap potentials, and crystal structure.

3.1. TiO_2 based photocatalysts

UV and visible light irradiation activate the photocatalytic TiO_2 to produced •OH and $•O_2^-$ like ROS on its surface having strong oxidizing power and that help disinfect the air, water, and other solid surfaces (Miyauchi et al., 2020). For instance, in a report by Matsuura et al., the inactivation of deadly SARS-CoV-2 using photoactive TiO_2-coated sheets was reported using a light emitting diode (LED) source. Oxidation of the lipid bilayer significantly increased the ions and H_2O permeability, which ultimately enlarges the virions particle size and altered its morphology. Transmission electron microscopy (TEM) images and western blotting results confirmed the enlarged viral diameter, spike, and other viral proteins degradation, RNA damage, and decreased level of nucleocapsid protein count. LED-TiO_2 photosystem ($\lambda = 405$ nm) mainly reduced the RNA levels and thus, disturbed the SARS-CoV-2 infectivity in both liquid and aerosols. Photoactive LED-TiO_2 affected the whole CoV-2 particles and incapacitated the virus by destroying structure, as well as genome (Matsuura et al., 2021). Another report illustrating the virucidal efficacy of small-sized (10 μm), photoactive TiO_2 (TNPs) placed on glass coverslips. Excited e^- in nanosized TiO_2/TNPs composite generated •OH and $•O_2^-$ (ROS) that rapidly damaged the HCoV-NL63 RNA genome through oxidative damage using low-intensity UV light as shown in Fig. 5b. Photoactive TiO_2 nanoparticles were virucidal efficient in a relatively humid environment (85%), signifying its use for wide-scale applications (Khaiboullina et al., 2021).

To further explore photoactive TiO_2 as an antiviral agent, Guo et al. studied the inactivation mechanism of ubiquitous human adenovirus-40 (HAdV40) that belongs to the family Adenoviridae using UV light source in the presence of TiO_2 photocatalyst. They investigated the photocatalytic disinfection rate kinetics of HAdV40 in H_2O using natural organic matter and the inactivation of HAdV40 in a batch photoreactor. Mechanistically, UV-induced inactivation of HAdV40 was *via.,* dimerization instead of generated free radicals (•OH) in the photocatalytic process. Also, the virus inactivity linearly depends on ϕ_{254}, suggesting that water quality mainly impacts the genetic DNA of HAdV40 (Guo et al., 2018). In another report, 4 log inactivation rate of MS2 (ssRNA) and PhiX174 (ssDNA) was achieved in sterilized DI-water using UV/TiO_2 within 20 min (Zuo et al., 2015). It was scrutinized that complete inactivation was mainly depended on the photoactive TiO_2 to virus mass ratio. In another study, 100% antiviral efficiency was demonstrated against Influenza H1N1 virus using TiO_2-coated porous ceramic substrate at $\lambda = 365$ nm (UV-A) (Daikoku et al., 2015). Owing to its large bandgap, bare TiO_2 photocatalyst often displayed visible inactivity and rapid reassembly rate of photoexcited e^- with remaining h^+. These associated fundamentals bottlenecks ultimately reduced the redox potentials of the resulting TiO_2 based photosystem.

V, Cr, Cu, Ag, Pt etc. like metal dopants in its lattice probably not only decreased the repairing rate of e^- and h^+ but also extend the solar light response somewhere in between the visible and NIR region. Doped TiO_2 provides higher photocatalytic disinfection and mineralization rates. Higher influenza A (H_3N_2) and vaccinia virus inactivation rate from 90% to 99.8% was likely due to better charge carrier isolation in Pt/TiO_2 as compared to bare photoactive TiO_2 semiconductor (Kozlova et al., 2010). Also, it was scrutinized that electrostatic forces play an important role in virus inactivation. In a report antiviral efficiency of N-doped TiO_2 was investigated by combining filtration and photocatalytic inactivation. MS2 exposure to photoactive N-doped TiO_2--coated Al_2O_3 membrane reactor displayed 3.9 ± 0.3 log and 4.9 ± 0.1 log inactivation in dark as well light respectively. Free •OH radicals near the surface were exposed to photocatalytic oxidation and the surface active sites. MS2 inactivation is facilitated by the oxidation of outer capsid or adsorption mediated by electrostatic forces. Experimental results demonstrated that salt bridge effect is the most likely route to reduce the electrostatic repulsions existed between the MS2 and photocatalytic membrane (Horovitz et al., 2018).

In a report, Liu et al. successfully reported the grafting of Cu (II) onto the surface of TiO_2 *via.* impregnation method to enhance the disinfection rate. Annealing under 950 °C notably changed the color from white to brown demonstrating the generation of oxygen defects or Ti^{3+} species as analyzed through SEM images. Also, the ultraviolet visible (UV–Vis) spectrum depicted that the visible light absorptivity is mainly owing to d-d transitions or interfacial charge transfer. Thereby, visible active Cu doped TiO_2 (420–800 nm) inactivated Qβ bacteriophage by 99.9% within 2 h and illustrated its reduction by log10 (6.5) after 30 min in light (Fig. 6a). Also, there are results that depicted the antiviral activity of as-prepared Cu–TiO_2 under dark conditions (Liu et al., 2015). Similarly, electrospun Cu doped TiO_2 nanofibers were fabricated for the selective degradation of bacteriophage f2 residing on host *Escherichia coli* using humic acid (HA). UV–Vis absorption peak displayed the absorption of Cu-doped TiO_2 in the visible region. The electrostatic force of attraction in between positively charged Cu–TiO_2 and negatively charged bacteriophage f2 strengthened the antiviral performance of the doped photocatalyst. The removal efficiency remained almost the same with and without HA in the first 3 successive cycles, however, the degradation affinity sharply decreased (1.9 log) after the fourth cycle (Fig. 6 b and c). Thus, photodegradation results inferred that increasing HA concentration in the solution relatedly decreased the bacteriophage f2 removal efficiency (Cheng et al., 2019).

Another electrospinning method was utilized to fabricate antiviral Cu-doped TiO_2 that displayed visible-light-driven inactivation of bacteriophage f2. Cu–TiO_2 photocatalyst provided a higher surface area to generate free unstable •OH radicles and offer more active sites for enhancing more f2 virus particles adsorption. It was analyzed that f2 removal rate increases with the increase in the number of photocatalysts (75 mg/L), higher solar light intensity (40–130 Mw/cm^2) as well as reaction temperature (25 °C–35 °C), and the fastest removal rate of f2 was observed at the pH = 6 and 7 (Zheng et al., 2018).

Dispersed nobel metal over the photoactive metal oxide semiconductor possesses outstanding properties owing to surface plasmonic effect. The individual capability of absorbing and scattering light particles mainly depends upon the type of nobel metal and the semiconductor used. For example, to avail the advantages of plasmonic Ag, Moongraksathum et al. studied the antiviral effect of Ag doped photoactive TiO_2 fabricated *via.* peroxo sol-gel route. Highest photoinactivation rate was observed at Ag: TiO_2 (1:100) under the illumination of ultraviolet A light source. As-fabricated multifunctional rhomboidal Ag-doped TiO_2 sols (1–3 nm) displayed 100% virucidal efficiency against H1N1 and enterovirus type 71 (Moongraksathum et al., 2019). Similarly, in another report by Liga et al. bare P25 TiO_2 attained a 1.6 log MS2 inactivation rate while UV-A achieved negligible removal within 2 min. From Fig. 7a and b, it was analyzed that Ag dopant in TiO_2 lattice enhanced the MS2 disinfection rate by 5 folds and rate constant to 584% owing to the increased photocatalytic oxidation (•OH generation) in the solution (Liga et al., 2011).

As compared to mono-doping, binary doping in TiO_2 lattice more effectively reduced the energy bandgap, shifted the spectral response well into the visible light region, and can more effectually improve its stability, solubility, magnetic as well as electronic properties as compared to monodopant. Venieri et al. used the co-precipitation route

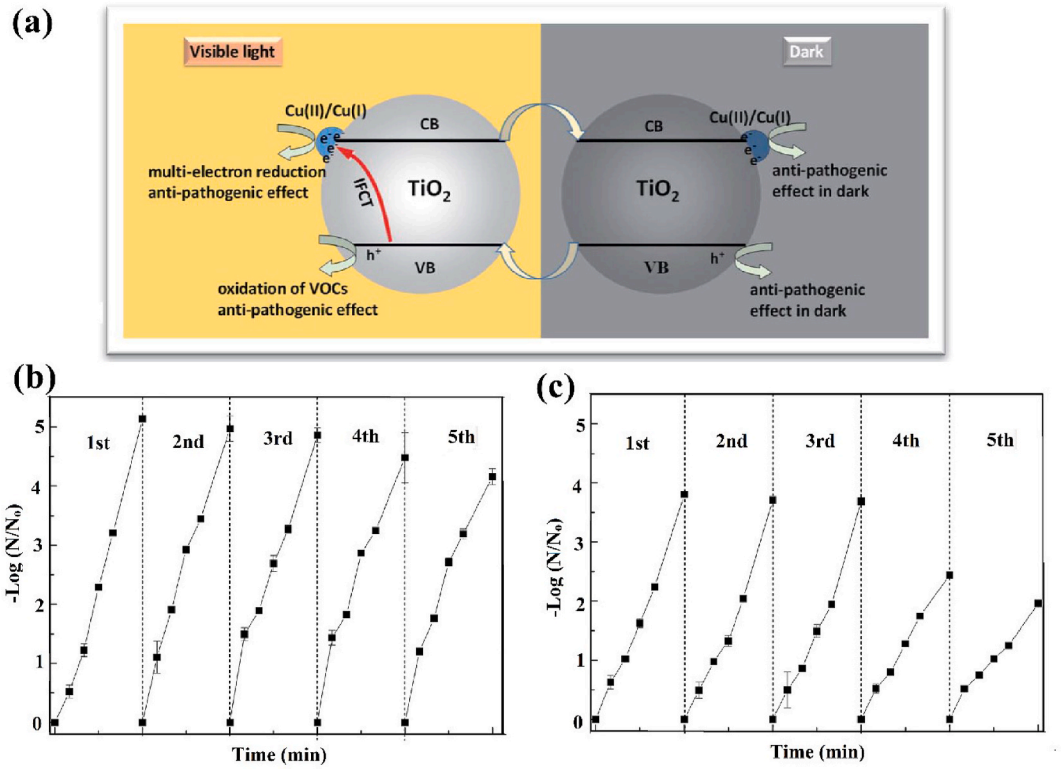

Fig. 6. (a) Proposed mechanism for the photocatalytic inactivation of Qβ bacteriophage viruses under visible-light and dark conditions, Adapted with permission from RSC (Liu et al., 2015); The degradation efficiency of f2 by as-fabricated recycled Cu–TiO_2 nanofibers in the **(b)** absence, **(c)** presence of HA, reprinted with permission from Elsevier, (license No. 5121831287775) (Cheng et al., 2019).

to fabricate Mn/Co-doped TiO_2 photocatalyst to incapacitate MS2 in a slurry reactor. Disinfection kinetic rate of bacteriophage MS2 using Mn/Co–TiO_2 followed pseudo-first-order reaction. Owing to the synergistic action of 0.1 wt % Mn/Co dopants in TiO_2, it was analyzed that less than 60 min were required to reduce 99.9% of the MS2 bacteriophage population (Venieri et al., 2015).

Silver and copper loaded TiO_2 effectively showed higher bacteriophage MS2 inactivation rate than bare or single atom doped TiO_2. Synergistic effect of both the dopants significantly improved the membrane photocatalytic activity to generate highly active •OH radicals. Very few amount of 0.16% and 0.02% of Ag and Cu ions, respectively were released indicating the stability of loaded ions. According to the results, synergistic effects of Ag and Cu on TiO_2 showed 4.06 ± 0.27 log removal of the bacteriophage (Rao et al., 2016).

3.2. WO_3 based photocatalysts

Instead of using UV-active TiO_2 as photocatalysts, WO_3 is another visible active transition metal oxide that could be potentially exploited for the removal of infectious CoV-2. Photocatalytic WO_3 semiconductor generally uses visible light radiations to generate a large number of reactive oxygen intermediate species that affect spike proteins, lipids, genomic RNA and nucleic acids. In this regard, very recently, Ghezzi et al. used strong oxidative WO_3 as a disinfectant to increase the effectiveness of the photocatalytic process. The photocatalytic reaction was carried out in infected liquid solution using WO_3 catalyst, antiviral tissue, and photocatalytic filter. Aliquotes were collected every 10 min to check the infectivity level and genomic RNA content. Results depicted that WO_3 reduced the infectious SARS-CoV-2 content by 98.2% as measured by quantitative polymerase chain reaction after 10 min and reached approximately 1.5 log10 after 30 min of the reaction. Kinetic studies of CoV-2 indicated that visible active WO_3 photocatalyst rapidly inactivated the viral particles almost 100% after 30 min of reaction by reducing detectable viral genomic RNA amount (Ghezzi et al., 2020a).

This team (Ghezzi. et al.) further reported a photosystem comprised of metallic filter coated with WO_3 and cotton fabric treated with Cu metal particles in the fight against the COVID-19 pandemic. The whole reaction was carried out in a photoreactor containing SARS-CoV-2 suspension and light-activated WO_3 coated grid. Fluids aliquots were similarly collected for 60 min and tested by using plaque assay and quantitative polymerase chain reaction to check its activity. Similar results were obtained after 30 min of the photoreaction (Ghezzi et al., 2020b). Similarly, visibly active (470 nm) Pt-loaded WO_3 photocatalytic material inactivated the influenza virus H1N1 (>5.3 log) within 2 h, and was effectively used to coat walls, and other solid surfaces to reduce virus infectivity (Takehara et al., 2010). Ultra-high surface area, tunable energy band gap, controllable defects density of graphene usually promotes reactivity, carrier mobility, and the rate of redox reactions that happen on the surface of photocatalyst. In order to exploit the properties of graphene, Akhavan et al. used graphene sheets to disinfect surfaces infected with MS2 using a visible light source. Chemically exfoliated graphene oxide (GO) sheets were fabricated to incorporate in the lattice of thin-film WO_3 photocatalyst. Annealing at 450 °C resulting in the formation of W–C and W–O–C bonding that helped break the protein units present on the enveloped MS2 as confirmed by sodium dodecyl sulfate-polyacrylamide electrophoresis (SDSP). As-fabricated graphene-WO_3 reduced the MS2 count to less than 5 PFU/mL i.e. more than 99.99% within 3 h (Akhavan et al., 2012). Thus, progressively as well as significantly, visible active WO_3 catalyst inactivated the infectious CoV-2 and other family viruses both for liquid and air decontamination in indoor environments.

3.3. Other metal oxide-based photocatalysts

Chandra et al. designed three non-toxic multi-core-shell SnO_2NPs@zeolitic imidazole framework (ZIF-8) composites through in-

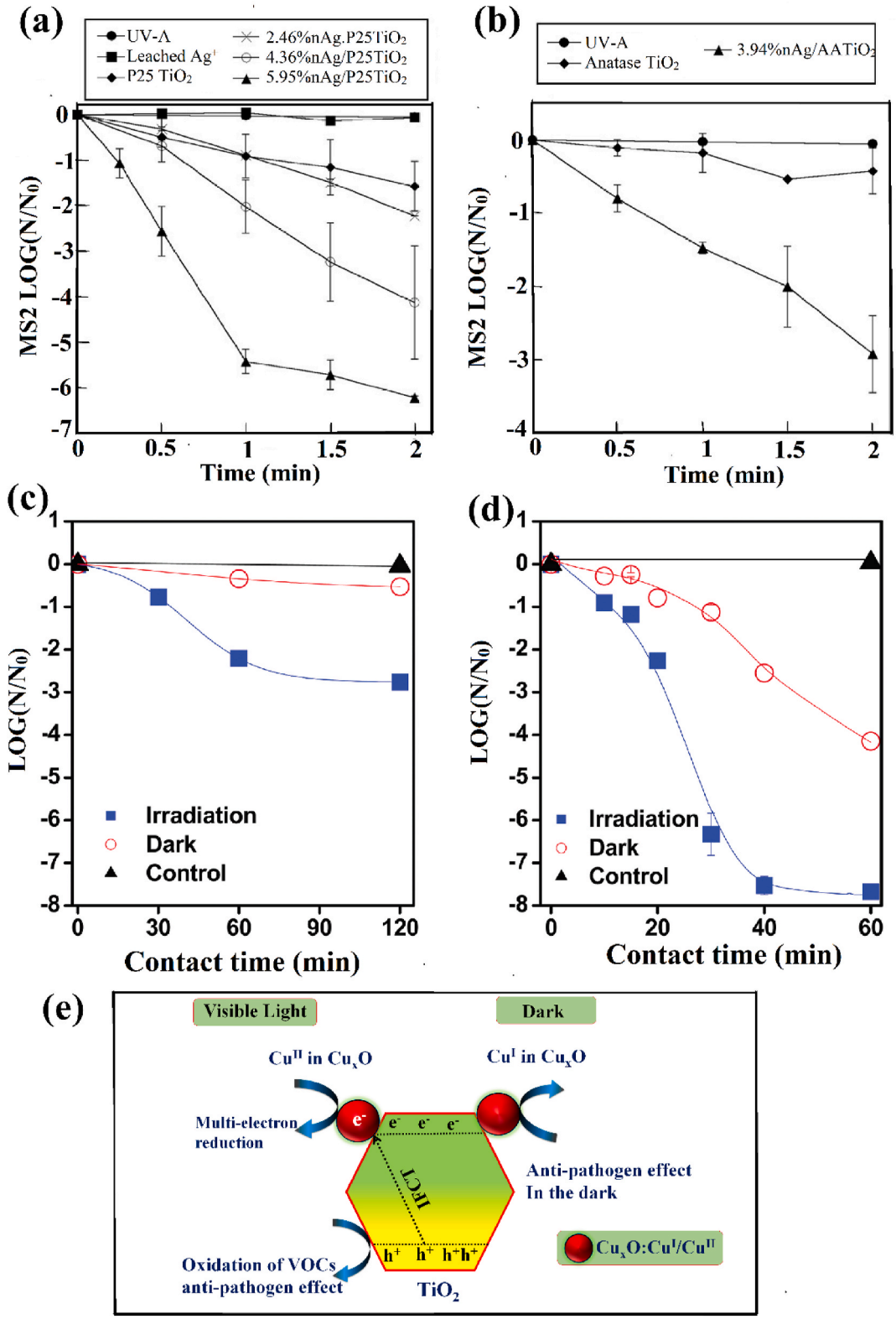

Fig. 7. MS2 virus inactivation by using **(a)** UV-A alone and Ag^{+} ion, P25 TiO_2, 2.46%,4.36%, 5.95% nAg/P25 TiO_2, **(b)** UV-A alone, Anatase TiO_2, and 3.94% nAg/anatase TiO_2 using UV-A irradiation, Adapted with permission from Elsevier (license No. 5121840259967) (Liga et al., 2011); Qβ bacteriophage inactivation by using various as-fabricated samples under dark and light irradiation, **(c)** 0.25% Cu(II)/TiO_2, **(d)** 0.25% Cu_xO/TiO_2 (Cu^{I}/Cu^{II} = 1.3), Reprinted with permission from ACS; **(e)** Proposed mechanism of photo-inactivation of viruses under both visible-light irradiation and dark (Qiu et al., 2012).

situ encapsulation of SnO_2 over ZIF-8 at 25 °C against chikungunya virus. Chikungunya virus is a re-emerging alphavirus transmitted by mosquitoes and having single-stranded RNA as genetic material. Up to 0.04 mg mL^{-1} bare SnO_2, ZIF-8, and SnO_2@ZIF-8 did not affect the viability of the cell. The antiviral property of as-fabricated three composites namely NC1, NC2, and NC3 was investigated, and the viral plaques were observed after 48 h of incubation. Composite NC1 (orange colored) displayed the highest antiviral activity i.e. >80% at the post and >50% at the pre-treatment stage as compared to other NC2 and NC3 nanocomposites. Also, bare SnO_2 nanoparticles showed 15% (co-treatment stage) and <15% (pre and post-treatment stage) antiviral activity, and resulted graphs exemplified that no significant changes in the antiviral property were reported when the samples were irradiated with light (Chandra et al., 2019).

Another stable Cu_xO grafted TiO_2 heterojunction was reported by Qui et al. to display its antiviral properties both in light as well as in dark conditions. Inactivation rate of Qβ bacteriophage under dark and visible light irradiation is shown in Fig. 7 c and d. Fig. 7 e depicted the denaturalizing of Qβ bacteriophage structural and non-structural proteins by Cu(I) ions under dark, and the oxidation of viral organic components under light irradiation using VB_{h+}. In the presence of light, Cu (I) ions were continuously formed so as to inhibit the self-oxidation rate of the Cu_xO semiconductor photocatalyst (Qiu et al., 2012).

In 2020, Domagala et al. reported the antiviral potential of Cu_2O nanoparticles coated with multi-walled carbon nanotubes effectively against MS2. Cu ion attachment, $Cu(OH)_2$ precipitation and $[Cu(NH_3)_4]^{2+}$ complex attachment were the three selected routes to fabricate three different composites (1–3), respectively. Fig. 8a and b depicted the MS2 bacteriophages removal tests performed for the first time prior to (day 1) and second time (day 2) after conditioning filters with water at pH 5 and 7 (24 h). The efficient removal results depicted that the design process is influential in virus removal. The effectiveness of disinfection in MWCNTs coated with copper oxide and the possibility of removal of the virus due to not only maintaining viral electrostatic

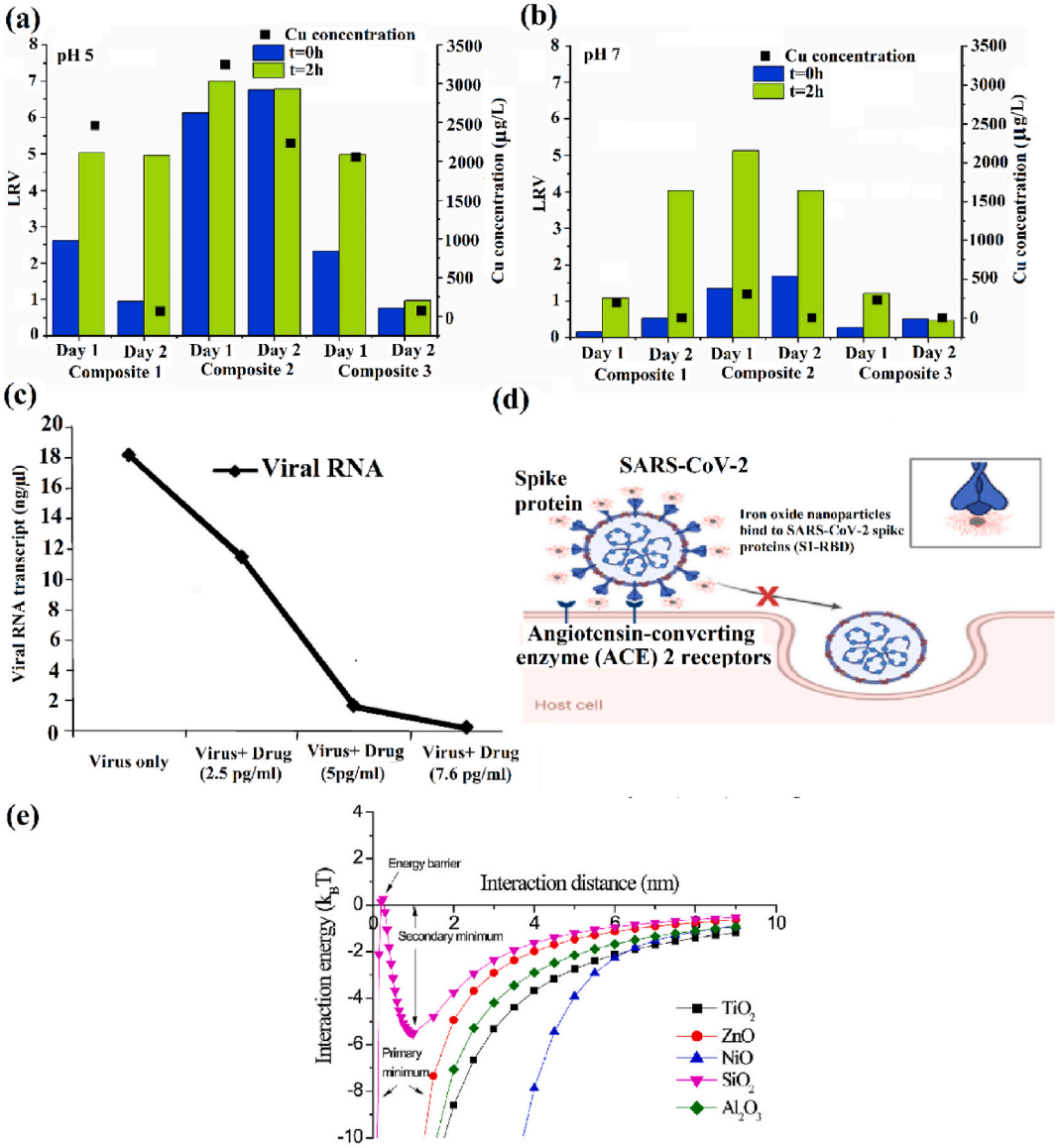

Fig. 8. MS2 bacteriophage removal test results for composite 1, composite 2 and composite 3 at **(a)** pH 5 and **(b)** pH 7, and the associated copper concentrations detected in the permeate, Adapted with permission from Elsevier (license No. 5230790562991) (Domagała et al., 2020), **(c)** An approximately 100-fold reduction in viral NP transcript was observed in the presence of Iron oxides nanoparticles (07 pg), Reprinted with permission from Elsevier (license No. 5230790739753) (Kumar et al., 2019), **(d)** Molecular docking study repurposes FDA approved iron oxide nanoparticles to treat and control COVID-19 infection, Reproduced with permission from Elsevier (license No. 5230781385241) (Abo-Zeid et al., 2020), **(e)** Typical repulsive, attractive, and total interaction energy curves between MS2 and different metal oxide NPs, Reproduced with permission from Elsevier, (License No. 5230800779974) (Zhang and Zhang, 2015).

adsorption in MWCNTs, but also in antimicrobial properties of copper that continued to act in this area (Domagała et al., 2020). Similarly, another report reported the synergistic action of photocatalysis and toxic Copper to inactivate bacteriophage T4. The experimental results demonstrated that inactivation rate increases with increased chemical oxidizing activity, as a result, as-fabricated TiO_2/CuO with chemical vapor deposition route effectively inactivated bacteriophage T4 within 3 h. An enhanced disinfection rate with antiviral TiO_2/CuO photocatalyst was achieved owing to the inclusion of Cu^{+}/Cu^{2+} ions in solution and photocatalysis (Ditta et al., 2008). Another study demonstrated the inactivation of human CoV-2 and rhinovirus using Ag metal mediated nanoscale cerium oxide particles. Two formulations of cerium oxide were produced with Ag modified surfaces. The physical interaction of cerium oxide nanoparticles with emerging pathogens (SARS-CoV-2, rhinovirus) leads to the disruption of lipid membrane integrity, denaturation of structural viral proteins, and virus encapsidation (Neal et al., 2021).

Another green synthesis was also reported by Navid and his group to fabricate ZnO NPs using Salvia hispanica leaves. As-fabricated ZnO NPs with both photochemical and biological properties had enough potential to act as an antiviral agent that inhibits the H1N1 virus up to 40% in 48 h and also had a wide range of medical applications (Rabiee et al., 2020). Similarly, phytofabricated 25-38 nm-sized Cr_2O_3 nanoparticle extracted from the fruit extracts of Hyphaene thebaica (reductant) have much potential to act as an antiviral agent against sabin like poliovirus owing to its stability and oxidation resistance ability. Mohamed et al.'s data analysis reported the slight inhibition of polio virus when co-incubated with Cr_2O_3 nanoparticles in L20B cells, suggested that Cr_2O_3 nanoparticles well interacted with virus particles and inhibit their activity (Ahmed Mohamed et al., 2020). Another 100-folds enhancement was observed using strong magnetic Fe_2O_3 (10–15 nm) nanoparticles to reduce HINI virus at the dosage of 7.0 pg as depicted in Fig. 8c. Mechanistically, the interaction of surface –SH groups with the proteins resulting into the complete inactivation of HINI virus (Kumar et al., 2019). Also, Fe_2O_3 and Fe_3O_4 were successfully interacted with SARS-CoV-2 receptor binding domain to form a stable complex which may lead to CoV-2 inactivation (Fig. 8d) (Abo-Zeid et al., 2020). The photoreactive iron-oxide surface provides an exclusive opportunity for improving the inactivation rate of adsorbed viruses by generating oxidants under sunlight.

Other metal oxides like As_2O_3 and Sb_2O_3 also showed improved deactivation rate against various harmful viruses. The antiviral photoactivity of As_2O_3 and Sb_2O_3 were studied against viral strain bacteriophage. Diffusion of the disinfecting •OH radical species into the viral cells suppresses the viral cell growth and viability. Studies depicted that Sb_2O_3 drastically reduced the growth whereas, As_2O_3 completely inhibited viral multiplication at 10 and 12 ppm. Comparative analysis demonstrated that As_2O_3 was much more effective to inactivate virus at lower dose of 4–8 ppm as compared to Sb_2O_3 at 8–12 ppm (Neal et al., 2021). Perovskite La_xMnO_3 version (x = 1, 0.95, and 0.9) is well-known for their catalytic properties involving oxidization reactions. By utilizing superb oxidative ability of $La_{0.9}MnO_3$, amino acid residues in viral envelope proteins are oxidized, thereby stimulating renewable protein production large-scale input into the process of making this description is not required. Best oxidative $La_{0.9}MnO_3$ (20 μg mm^{-2}) effectively degraded 76% influenza virus in 15 min and owing to its oxidative mechanism it is capable to disinfect wide range of other harmful pathogens (Weng et al., 2015).

Thus, virus inactivation requires a strong interaction between virus particle and photoactive metal oxide where adsorption and attachment of virus onto nanoparticles might occur in the region of primary minima. All metal oxide nanoparticles exhibited strong adsorption and inactivation of MS2 to different extent. Zhang et al. demonstrated the colloidal interactions of NiO, TiO_2, Al_2O_3, ZnO, SiO_2 nanoparticles with bacteriophage and MS2 (Fig. 8e). Small particle sizes, protein surface, and charged colloidal particles of bacteriophage and MS2 on to the surface of TiO_2>Al_2O_3>ZnO > NiO > SiO_2. Similar surface charges (negative) of MS2 and SiO_2 lead to less repulsions owing to small energy barrier of 0.3 k_B T. The electrostatic repulsion between virus particle and metal oxide nanoparticles was completely overcome by the attractive van der Waals and acid-base forces (Zhang and Zhang, 2015). The multi-antiviral effects of metal oxides-mediated photocatalytic reaction implies universal disinfection potential for different viruses. Thus, metal oxides semiconductor based photocatalysis has indicated a potential alternative technology with significant advantages. ROS, especially hydroxyl radicals (•OH) would accumulate near the cell membrane and their interaction disturbs the metabolism as well as replication rate of viruses. The outcomes demonstrated that the fabricated above-mentioned photo--activated metal oxides and their nanocomposites could proficiently inactivate human viral pathogens.

Conclusively, from the above results, it is observed that viruses are more resistant to disinfection than other pathogens in both mono and hetero photosystems under given reaction conditions. Herein, there are only four reports where transition metal oxides directly worked with CoV-2, and rest of the explored work focused on their effective study with other enveloped and very few with non-enveloped viruses that are relatively closed to SARS-CoV-2. It is first to discover that photoactive transition metal oxides inactivate the enveloped CoV-2 by destroying its outer shell or the capsid protein with the help of unstable reactive species. Induced oxidative stress owing to the formation of reactive $•O_2^-$, •OOH, •OH radicals, basically rupture the CoV-2 shell, oxidize the C–H bonds, disturb the functioning of intracellular components and exposed the genetic RNA. Researchers strongly agreed that reactive oxygenated species generated on the surface of transition metal oxides plays a significant role in CoV-2 disinfection. Taken together, the above studies demonstrated that transition metal oxides mediated photocatalytic reaction can be effectively used to control the spread of CoV-2 and thus helpful in mitigating the ongoing pandemic. Moreover, transition metal oxides based photocatalysts can also potentially be useful against other newly evolving toxic, and infectious pathogens.

4. Conclusion and outlook

The properties like abundant active reaction sites, low toxicity, superior physicochemical, biocompatibility, electronic properties, greater surface to volume ratio, high recyclability, and durability of transition metal oxides make them effective materials for photocatalysis. This review paper reports a succinct overview of the use of potential photoactive transition metal oxides as antiviral agents. Apt design and preparation of transition metal oxides photocatalyst with improved exciton communication would accelerate robust disinfection of viral particles at a low level of radiation energy. Recent analyzed studies affirm that TiO_2 among transition metal oxides have been exploited as superior antiviral material to inactivate viruses including CoV-2 in fomites or air.

Viruses' inactivation route by using solar light and catalyst has been broadly studied, though the mechanistic studies remain unclear for naked viruses as compared to the enveloped. Sympathetic the virus-host interaction is key to vaccines, treatments, and diagnoses. Reported data analysis clarified that the inactivation mechanism of non-enveloped viruses includes the damage caused by ROS in the capsid protein or either in the genetic RNA, but still the mechanism is not well experimentally clarified. Also, few reports demonstrated the inactivation mechanism of enveloped viruses where the inactivation is mainly dependent on the generation of unstable •OH species. Although thorough eradication of SARS-CoV-2 requires more research to reduce the prevalence of COVID-19.

Commercialized use of photocatalysis on a wide scale will definitely clean up the contamination from the environment and consequently recognized as a significant future research way worthy of consideration. Metal oxide nanomaterials are effectively used in numerous applications like biosensor detection, diagnostic kits preparation, as antiviral agents,

in filters, facemasks, as drug delivery agents, and in vaccines against various harmful viruses. Many research groups have dedicated their efforts in developing new metal oxides based photoreactors to improve existed designs. In brief, m-RNA and DNA-based vaccines are not much effective without the nanoparticles, hence by using nanoparticles ingredients, not only the transmittance of H1N1, SARS-CoV, SARS-CoV-2 like deadly viruses can be better and sooner controlled but it also makes the protecting masks, textiles, and filters reusable in the society as well. Further studies are still required to analyse the antiviral nature of transition metal oxides to reduce viral infections and to combat various pandemic. Therefore, it can be assumed that these exclusive transition metal oxides photocatalyst will surely pave the effective way to completely combat the deadly CoV-2 in near future.

However, the complexes are little expensive and have stability issues due to d-d transitions. Therefore, developing accessible, relatively cheap, metal free photocatalysts are promising strategies to meet the increasing demand for more sustainable green antiviral agents. Moreover, the toxicity of using nanomaterials in vivo is still a serious concern in this direction, hence the use of least toxic, standard nanomaterials with antiviral properties is a big challenge that needs further exploration. Further, research reports are needed to explicate various factors (viral, and host) in the pathogenesis of various deadly infections. Also, other following aspects for instance; detailed understanding of virus disinfection mechanisms, the required optimal reaction conditions, the effect of reaction parameters, should be examined. The use of antiviral nanomaterial agents is an active research arena and we believe that more research should be done to prepare more active metal oxides with higher superior properties for environmental mitigation and air purification. Thus, further research should be necessarily carried out to take the benefits of other heterogeneous transition metal oxides photocatalytic semiconductors to disinfectant deadly viruses and their diseases. Moreover, the activity of photoactive nanomaterials is mainly dependent on a light source to generate unstable free radicals, which may increase their cost. Therefore, the suggestion is to focus on the development of room temperature-based catalytic materials within the nano range that can similarly produce radical species but without using any additional energy output.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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