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SHORT COMMUNICATION

PVC-SiO_2-Ag composite as a powerful biocide and anti-SARS-CoV-2 material

Marcelo Assis[1,2] · Luiz Gustavo P. Simoes[3] · Guilherme C. Tremiliosi[3] · Lara Kelly Ribeiro[1] · Dyovani Coelho[1] · Daniel T. Minozzi[3] · Renato I. Santos[3] · Daiane C. B. Vilela[3] · Lucia Helena Mascaro[1] · Juan Andrés[2] · Elson Longo[1]

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Abstract

The ongoing COVID-19 pandemic has pushed scientists and technologists to find novel strategies to develop new materials to prevent the transmission, spread, and entry of pathogens into the human body. In this report, the fabrication of polyvinyl chloride (PVC)-SiO_2-Ag composite is presented, in which the percentage of Ag is 0.84% wt. Our findings render that this composite eliminates (> 99.8%) bacteria and fungus (*Staphylococcus aureus*, *Escherichia coli*, *Penicillium funiculosum*) and SARS-CoV-2, by surface contact in 2 h hours and 15 min, respectively. Specific migration analysis shown that the use of the PVC-SiO_2-Ag composite is considered safe and effective for food preservation. This research and innovation front can be considered a breakthrough for the design of biocide materials. Future directions for this exciting and highly significant research field can open the door to the development of new technologies for the fabrication of packaging films to protect consumer products (such as fruits, vegetables, and other foods).

Keywords PVC-SiO_2-Ag composite · Biocide composite · Anti-SARS-CoV-2 material · PVC

Introduction

Currently, microorganisms (including bacteria, fungi and viruses) are one of the main causes of disease in the world [1]. The public health outbreaks caused by emerging COVID-19 infectious diseases constitute the forefront of current global safety concerns and a significant burden on global economies [2–4]. While there is an urgent need for the effective treatment of these outbreaks based on antiviral and vaccines, it is essential to explore any other effective intervention strategies that may reduce the mortality and morbidity rates of the disease [5, 6]. The development of innovative materials able to prevent the transmission, spread, and entry of COVID-19 pathogens into the human body is currently in the spotlight. The synthesis of these materials is, therefore, gaining momentum, as methods are providing nontoxic and environmentally acceptable “green chemistry” procedures [7].

It has been known for over a century that silver nanoparticles (Ag NPs) are one of the most active against microorganisms and they are widely used due to their antiviral properties and the lower chance of developing resistance when compared to conventional antivirals [8–10]. Ag NPs present excellent bio-activities and have been incorporated into a wide variety of consumer products including protective equipment, clothing, food containers, packaging, and air purifiers (Project on Emerging Nanotechnologies) [11]. Nevertheless, Ag NPs are reactive and unstable in physiological conditions, and their practical applications are often hampered by oxidization, which results in aggregation and the loss of antimicrobial activity [12]. To solve this problem, Ag NPs can be dispersed on metal oxides to form metal/semiconductor composites, which are employed with success in a wide range of applications, including heterogeneous catalysis, energy conversion and environmental applications [13].

Ag NPs display a unique characteristic associated with the localized surface plasmon resonance (SPR) behavior [14, 15], which render remarkable antimicrobial activity [16–18]. This is mainly due to the following factors: (i) the SPR

✉ Juan Andrés
andres@qfa.uji.es

1 CDMF, LIEC, Federal University of São Carlos - (UFSCar), São Carlos, SP 13565-905, Brazil

2 Department of Physical and Analytical Chemistry, University Jaume I (UJI), 12071 Castellon, Spain

3 Nanox Tecnologia S/A, São Carlos, SP 13562-400, Brazil

adsorption and high electron trapping ability of Ag NPs are beneficial for promoting solar energy conversion [19, 20], water splitting [21, 22], photocatalytic [23–25] and antimicrobial [26–29] activities of the composites, (ii) the excellent conductivity of Ag NPs at the interface improve the transfer of charges from SPR metals to semiconductors, thus leading to enhanced activity[30]. Previously, we demonstrated an anti-SARS-CoV-2 activity, which efficiently hampered infection and transmission via surfaces; in particular, an antimicrobial coating, a polycotton fiber Ag based material, that could rapidly kill bacteria *Staphylococcus aureus* (*S. aureus*), *Escherichia coli* (*E. coli*), fungi (*Candida albicans*), and SARS-CoV-2 by contact was presented [31].

The amorphous semiconductor SiO_2 meets many important requirements, i.e. ease of synthesis and low cost, high biocompatibility and biodegradability, hydrophilicity, stability, optical transparency tunable size, and versatile surface chemistry [32, 33]. Due to its unique characteristics, it has been employed in a wide range of applications, such as cosmetics, food, pharmaceuticals and medicine [34, 35]. Taking advantage of SiO_2 as an efficient host for stabilizing Ag NPs and the SiO_2-Ag heterojunction has attracted considerable attention due to its excellent properties [36–38], in the following study by our research group, Ag NPs were stabilized with semiconductor amorphous SiO_2 to form a SiO_2-Ag heterojunction. This heterojunction was immobilized in a polymeric matrix (ethyl vinyl acetate), to render a highly virucide material [31]. In this study, we also confirmed, by using scavenger experiments, that hole (h^+), hydroxyl radical (●OH), and hydroperoxyl radical (●O_2H) are the reactive species along the mechanism of the virus elimination process [31]. It is assumed that the formation and separation of electrons, e^-, and h^+, are enhanced due to the synergistic interaction/effect generated at the surface of the SiO_2 semiconductor and the SPR of the Ag NPs at the interface. In the semiconductor, the h^+, at the valence band, reacts with H_2O by decomposing it into ●OH and a proton (H^+). The e^- migrates to the conduction band reacting with O_2 to form a superoxide radical (●O_2^-), which in turn interacts with H^+ forming the radical ●O_2H. These free radicals are capable of killing microorganisms by oxidizing and breaking down the cell walls and membranes of bacteria, fungi, and viruses, as demonstrated in different heterojunctions formed by Ag NPS and binary and ternary metal oxides [31].

The experimental results indicated that the as-fabricated samples exhibited high antibacterial activity towards *E. coli* and *S. aureus* as well as towards SARS-CoV-2 [31]. Furthermore, this activity can be functionalized as a new technology in the manufacture of medical devices, such as reusable masks and pharmaceutical packaging, food storage packaging materials, displaying high flexibility, biostability and easy processing [39–41]. Emerging foodborne pathogens

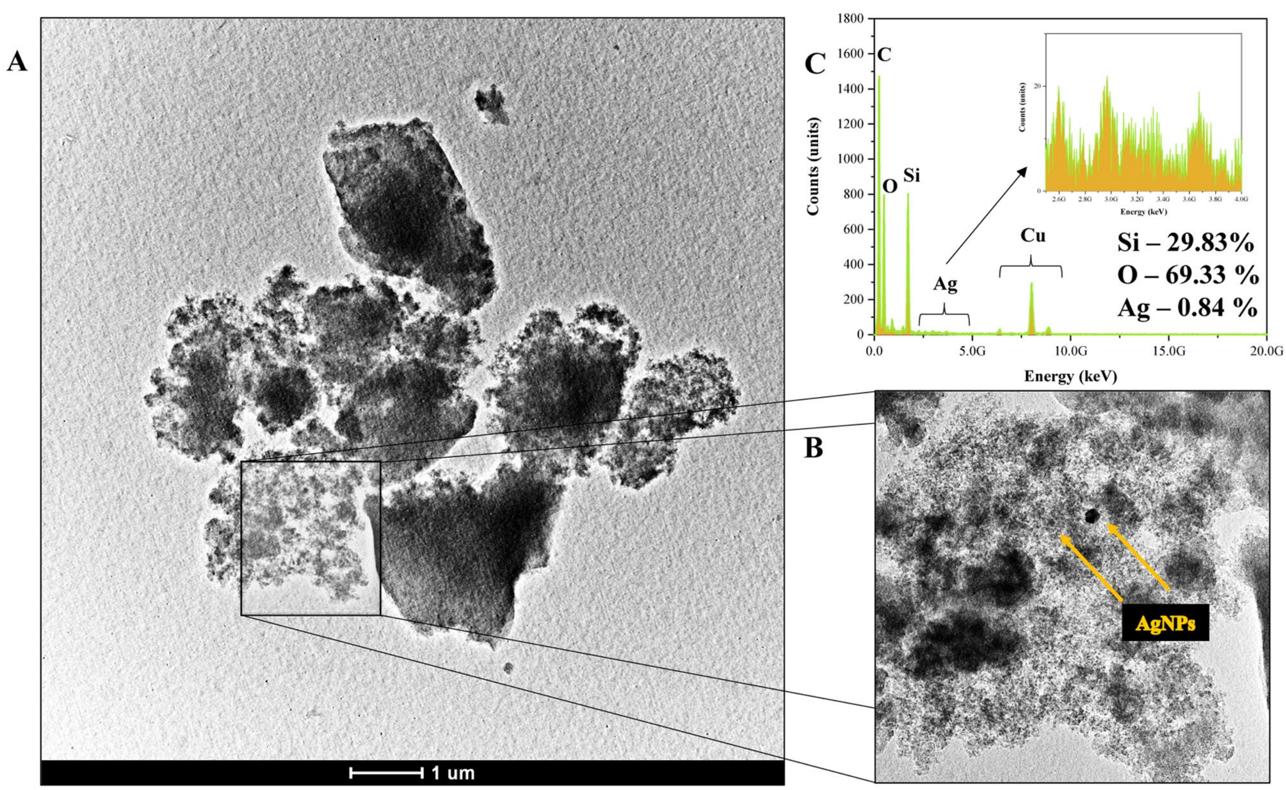

Fig. 1 **A-B)** TEM images of SiO_2-Ag heterojunction. **C)** Chemical composition from EDS analysis of the sample (weight %)

are considered a major public health and sanitary control, in addition to affecting food processing industries as well as consumers. The consumption of food contaminated by microorganisms leads to different types of foodborne illnesses, an example is the old chain associated with the re-emerging outbreaks of COVID-19 in Beijing, China [42, 43]. Furthermore, countless cases of diseases that are associated with foodborne pathogens are reported every year worldwide [44]. Biocide materials covered by polymers is an important strategy to be applied for different uses, what makes the studies of possible food storage packaging viable and promising because they can inhibit the growth of microorganisms and food spoilage, extending the product a longer shelf life [45, 46].

Inspired by these pioneering studies, in this communication we develop a greener and convenient approach for the synthesis of polyvinyl chloride (PVC)-SiO_2-Ag. PVC is one of the most used thermoplastic polymers worldwide, due to its versatility, high stability and resistance. As a hard thermoplastic it can be used in the packaging of consumer products and for biomedical applications, especially for surgical and dialysis technologies [47]. The PVC-SiO_2-Ag composite proposed in this study has the ability to kill bacteria (*S. aureus*, and *E. coli*), fungus (*Penicillium funiculosum* (*P. funiculosum*)) and SARS-CoV-2 by surface contact. These results demonstrate that this material constitutes an effective platform for simultaneously eliminating different pathogens, avoiding their transmission, protecting packaging of consumer products and increasing the shelf life of perishable foods.

Materials and methods

Experimental details on the method for obtaining the polymeric composite, microbicidal tests and specific migration tests can be found in the Supplementary Material.

Results and discussion

Figure 1A-B shows the transmission electron microscopy (TEM) and energy-dispersive x-ray spectroscopy (EDS) analysis of the SiO_2 microstructures, composed of small, and dark nanoparticles deposited on their surface. The synthesis of the SiO_2-Ag heterojunction and the PVC-SiO_2-Ag composite are shown in the Supplementary Material. When performing the EDS analysis (Fig. 1C) of this region, a ratio of almost 2:1 is observed from O (69.33%) to Si (29.83%), expected for the structure of SiO_2, in addition to a small percentage of Ag (0.84% wt) can be sensed. These results confirm that SiO_2-Ag heterojunction is obtained successfully. The stability of Ag NPs was analyzed by spectrophotometry in the UV–Vis region, as they are prone to oxidation when dried at high temperatures. There were no differences in the plasmonic absorption band of Ag NPs (573 nm) between samples (heat-treated and not heat-treated), showing that the drying process does not alter the Ag NPs (see Fig. S1 in the Supplementary Material).

Once the composite was obtained, microbiological elimination tests were performed to assess the activity of the material against infectious pathogens. This analysis is the first step to confirm the use of this material in packaging films to protect consumer products, since indirect infections can occur due to contact with contaminated surfaces [48, 49]. These infections occur because pathogenic microorganisms can remain active for long periods on different surfaces, depending on the chemical composition

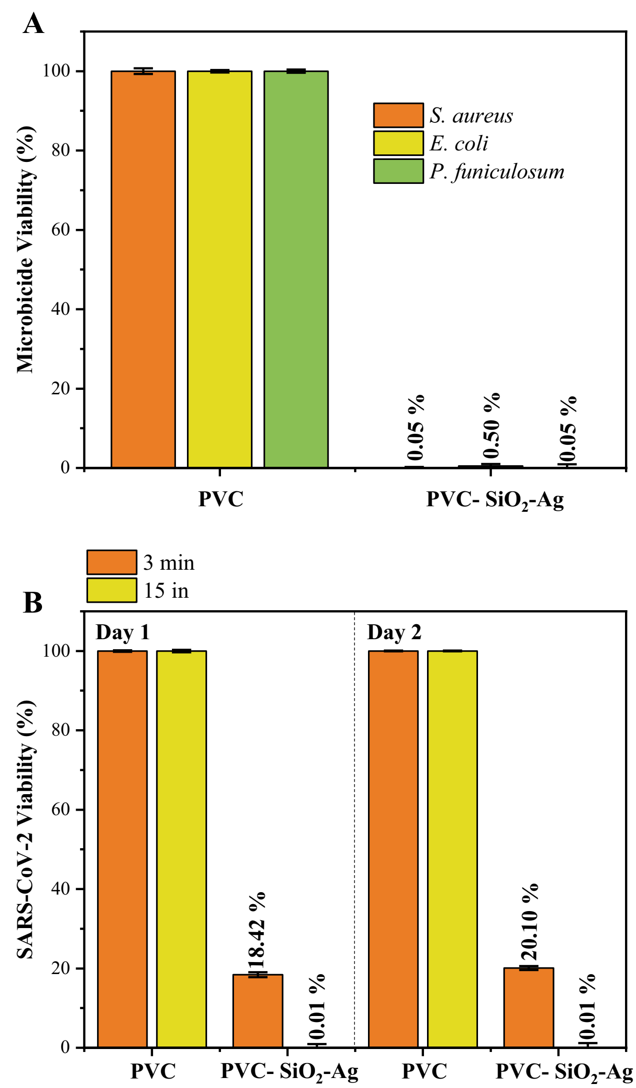

Fig. 2 (**A**) Microbicidal results against *S. aureus*, *E. coli* and *P. funiculosum* after 24 h of exposure to pure PVC and PVC-SiO_2-Ag composite. (**B**) Virucidal results against SARS-CoV-2 using PVC-SiO_2-Ag composite

and physical structure of the surface [50]. The activity of the PVC-SiO_2-Ag composite was evaluated against the bacteria *S. aureus* and *E. coli*, and the fungus *P. funiculosum* (Fig. 2A) after 24 h of contact, while activity against the virus SARS-CoV-2 was evaluated for 3 and 15 min on 2 consecutive days (Fig. 2B). The experimental procedure is described in the Supplementary Material.

An analysis of the results renders that the PVC-SiO_2-Ag composite in contact with the microorganisms *S. Aureus*, *E. Coli* and *P. funiculosum* suffer eliminations of 99.95, 99.50 and 99.95%, respectively, whereas for pure PVC no elimination is observed. As for the SARS-CoV-2 virus, the tests were carried out for two consecutive days, at the times of 3 and 15 min. At 3 min there is an elimination of 81.58% of copies of the virus, followed by 99.99% at 15 min. A similar result is observed on the second day, with eliminations of 79.90% and 99.99% at 3 and 15 min, respectively. As with bacteria and fungus, no reductions in virus copies were observed with pure PVC. Therefore, the PVC-SiO_2-Ag composite becomes an effective platform for avoiding the infection, proliferation, and transmission of bacteria, fungi, and viruses.

To illustrate the potential of PVC-SiO_2-Ag composite as a material to protect consumer products, Fig. 3 shows the results obtained in which papaya is covered by a pure PVP film and an PVC-SiO_2-Ag composite film during 14 days; for comparison purposes a papaya without a cover is also included. A time lapse video can be seen in the Supplementary Information.

It is observed that for papaya exposed to the environment, on the third day the top of the fruit begins to deteriorate, while on the fourth day, the papaya packed with pure PVC film also begins to show these signs of deterioration. Papaya packed with the PVC-SiO_2-Ag composite film begins to show signs of deterioration on the ninth day, increasing the fruit's life span by 5 or 6 days, when compared to other conditions. Thus, it is observed that in addition to preventing contact infections against bacteria (*S. aureus* and *E. coli*), fungus (*P. funiculosum*), and SARS-CoV-2, the PVC-SiO_2-Ag composite can be applied to increase the shelf life of fresh food, since contamination of microorganisms can cause food to rot.

In order to investigate the possible migration process of Ag NPs at the PVC-SiO_2-Ag composite, specific migration tests were carried out according to Commission Regulation (EU) No 10/2011 (Table 1) [51]. To this end, three different solvents have been selected: acetic acid 3% (v/v), ethanol 50% (v/v), and olive oil. Due to the acid and hydrophilic character of acetic acid, this test can be used to analyze the response of foods with these characteristics,

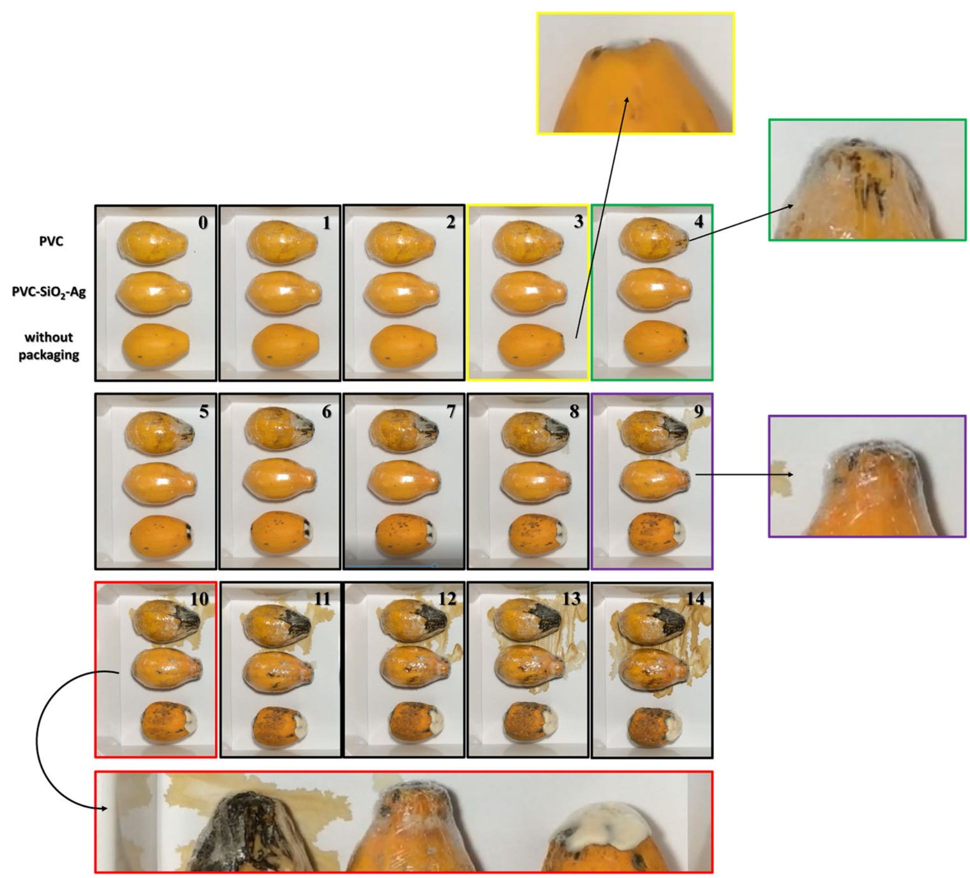

Fig. 3 Papaya degradation under ambient conditions for 14 days with pure PVC film, PVC-SiO_2-Ag composite film and without packaging

Table 1 Specific migration for Ag in PVC-SiO_2-Ag composite

Food Simulants	Ag content (μg/Kg)
Olive Oil	$< 30 \pm 0.0$
Ethanol 50%	$< 6 \pm 0.0$
Acetic Acid 3%	11 ± 1.8

while ethanol and olive oil allows us to investigate alcoholic/dairy and fatty foods, respectively [52, 53]. The specific migration analysis shown that for all cases, the Ag content was less than 30 μg/Kg, a mass much smaller than accepted by Commission Regulation, which allows limits below 500 μg/Kg for metals. The specific migration of SiO_2 was not taken into account, as it is not restricted by Commission Regulation [51]. Therefore, the use of the PVC-SiO_2-Ag composite is considered safe and effective for food preservation.

Conclusions

Finding new materials to eliminate microorganisms is clearly a high priority with the emergence of the present COVID-19 pandemic. In this study, we evaluated the efficiency of the PVC-SiO_2-Ag composite and present findings render that this material eliminates bacteria (*S. aureus*, *E. coli*), fungus (*P. funiculosum*), and SARS-CoV-2, by surface contact. Consequently, there is no doubt that materials incorporating this composite in every-day devices such as smartphones clothing, eye- and face-ware like glasses and masks constitutes will be the bases of the development of new technologies for viral disinfection in the sectors of health, processing, storage, and food transportation. In rapidly expanding bioengineering branches, this work contributes to further research to provide comfort and confidence to humans worldwide, by preventing the transmission and contamination of SARS-CoV-2 and other pathogens, and its impacts on society.

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Authors' contributions M.A.: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data Curation, Writing—Original Draft, Writing—Review & Editing, Visualization. L.K.R.; D.C.: Methodology, Validation, Formal analysis, Investigation, Data Curation, Writing—Original Draft, Writing—Review & Editing. L.G.P.S.; G.C.T.; D.T.M.; R.I.S.; D.C.B.V.: Resources, Conceptualization, Methodology, Validation, Formal analysis, Data Curation, Writing—Original Draft, Writing—Review & Editing, Visualization, Funding acquisition. L.H.M.; J.A.; E.L.: Resources, Conceptualization, Data Curation, Writing—Original Draft, Writing—Review & Editing, Visualization, Funding acquisition.

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Availability of data and material The data that support the findings of this study are available from the corresponding author upon reasonable request.

Code availability Not applicable.

Declarations

Conflicts of interest The authors declare no conflict of interest.

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