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# Investigation of the antibacterial activity of sputtered ZnO coating on borosilicate glass: a nanostructured approach for food packaging

Jyoti Pandey and Suchitra Rajput \*

In this study, nanostructured ZnO is sputter-coated on a 7059 corning borosilicate glass substrate *via* RF magnetron sputtering to explore its potential application in antimicrobial food packaging. Surface characterization *via* FE-SEM and XRD, contact angle measurement, transmittance measurement *via* UV visible spectroscopy, and antibacterial testing against *E. coli* and *S. aureus* have been conducted to evaluate the coating. Antimicrobial studies have shown excellent activity against *E. coli* (100%) and *S. aureus* (83.25%). The coating's stability was evaluated over 120 hours. The findings highlight the potential of sputtered ZnO coatings as active antimicrobial barriers for glass-based food packaging as well as healthcare packaging, offering enhanced hygiene, safety, and shelf-life extension for perishable products.

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## 1 Introduction

Maintaining food safety and prolonging shelf life are critical concerns for consumers and industries in today's fast-paced life. Foodborne illness is a worldwide public health concern. Foodborne contamination causes millions of illnesses, thousands of hospitalizations, and even fatalities worldwide.<sup>1</sup> The 1st edition of World Health Organization (WHO) in collaboration with WHO Foodborne Disease Burden Epidemiology Reference Group (FERG) estimated illness of 600 million people due to hazardous contaminants including bacteria, viruses, parasites, chemicals, and toxins by the year 2050. The reports estimate the mortality of 420 000 people due to unsafe food, out of which 125 000 deaths estimated were of children below age 5.<sup>2</sup> The estimated risk factor in 2025 would increase by 10 million due to drug-resistant microbes.<sup>3</sup> There is an urgent requirement to adopt a cutting-edge method to manage and control foodborne infections. Packaging has a key role in ensuring the quality, freshness, and safety of food products during storage and transportation. Active packaging materials, which help in prolonging the shelf life of perishable foodstuffs while maintaining the freshness and quality are of prime importance.<sup>4</sup> With the rising urbanization and industrialization, the requirement of packed food has pushed packaging materials and their quality to the research domain.<sup>5</sup> In the same line, freshness indicators and sensors actively monitor spoilage and help reduce waste.<sup>6</sup> Zhang *et al.* effectively combined antimicrobial agents with monitoring mechanisms to effectively maintain meat quality and safety.<sup>7</sup>

Though polymer and composite-based packaging are popular in healthcare as well as food industries, glass packaging still stands out due to its chemical inertness, barrier properties, and recyclability.<sup>8</sup> Plastic food packaging is highly discouraged now, owing to its sensitivity to environmental factors like oxygen, vapours, and light, resulting in facultative bacterial and/or fungal growth. In addition, the fear of microplastic leaching and carcinogens after heating/cooling the food still exists with this packaging.<sup>9</sup> Recycling of plastic-based packaging has also become one of the major concerns in the food packaging industry. Aluminium on the other hand has gained popularity in food packaging industries since the early 20th century due to its low density and malleability.<sup>10</sup> Aluminium has been used in various forms, such as foil, tubes, bottles, laminated and metallized films,<sup>11</sup> tetra packs, metal containers, and lids. However, both glass and metal (Al) containers are susceptible to microbial contamination on their surfaces, which may compromise food safety and shelf life.<sup>12,13</sup> At the same time, leaching of Al in food has been suspected to be correlated with patients having Parkinson's and Alzheimer's diseases.<sup>14</sup> Wastage of foods due to poor packaging has now become a major concern on long haul flights for airlines as studied by M. Halizahari *et al.*, indicating the need to enhance the shelf-life of food *via* better packaging options.<sup>15</sup>

The growing desire for higher food safety, shelf-life extension, and sustainability has become a pivotal priority in the global food packaging industry, leading to smart packaging or "active packaging". Modern packaging would not only protect stored stuff from contamination but increasingly function as a barrier against environmental contaminants and microbial invasion. The emergence of smart coatings, particularly those with antimicrobial properties, has opened new pathways in food preservation technology. For example, bionic pearl layer

Centre for Advanced Materials and Devices (CAMD), School of Engineering and Technology, BML Munjal University, Gurgaon, Haryana 122413, India. E-mail: rajput.suchitra@gmail.com; suchitra.rajput.chauhan@iitdalumni.com



and bionic cicada wing are getting attention due to enhanced mechanical barrier properties and physical bactericidal activity.<sup>16</sup> Despite huge progress in biomimetic materials, formidable challenges, like the reaction between these materials and the organic acids in food, expansion of these materials due to moisture adsorption from food, and food components' (salt, polysaccharides, oil) reactions with the biomimetic material, still exist.<sup>17</sup> Zein self-assembled nanoparticles (NPs) can also be used in food preservatives to protect the shelf-life of food. The zein NPs may be controlled through temperature, polarity of the solution, pH, strength of ions and proportion of polymers.<sup>17</sup> Zein NPs and  $\epsilon$ -polylysine help to fight fungal infection to extend avocado's shelf life.<sup>17,18</sup> Also, zein, lecithin, and epigallocatechin gallate keep loquat fresh.<sup>17</sup> Zein NP coating can also be used to preserve meat.<sup>17,19</sup> Nevertheless, zein NPs with controlled shapes and sizes need to be designed to enhance the stability and loading capacity in practical applications. Wang *et al.*<sup>20</sup> proposed a self-healing material (*viz.*, polyelectrolyte multilayer film (PEM)) for food packaging. PEM consists of sodium carboxymethylcellulose and chitosan. PEM film is a good anti-oxidant, exhibiting antibacterial properties along with self-healing capabilities. But these films need to be activated by water for their self-healing capabilities. Recently, essential oil-based systems, such as citral–eugenol microcapsules, have shown synergistic antifungal activities against *Aspergillus niger*, enhancing film hydrophobicity and increasing the shelf life of bread from 4 to 10 days.<sup>21</sup>

Furthermore, zinc oxide (ZnO) has gained significant attention in active packaging due to its multifunctional capabilities in food-grade coatings.<sup>22–24</sup> It has lower cytotoxicity, more biocompatibility, and better stability, and is chemically inert and transparent, with UV-blocking capability, and most importantly, it shows antibacterial<sup>25</sup> and antifungal activities.<sup>26</sup> All these properties make ZnO coatings particularly suitable for food packaging applications, where microbial control is crucial. The use of zinc oxide nanoparticles as a biomarker, biosensor<sup>27,28</sup> and drug delivery agent<sup>29–31</sup> is also extensively explored. The antibacterial action of ZnO is attributed to mechanisms such as reactive oxygen species (ROS) generation, Zn<sup>2+</sup> ion release, and direct interactions with microbial cell walls, leading to membrane disruption and eventual cell death.<sup>25</sup> These effects have been widely demonstrated against a broad spectrum of foodborne pathogens including *Staphylococcus aureus* (*S. aureus*), *Escherichia coli* (*E. coli*) and *Aspergillus niger* (*A. niger*).<sup>32,33</sup> ZnO coatings have shown encouraging antibacterial activity when applied to glass substrates without affecting the optical or structural characteristics of glass.<sup>34</sup>

While ZnO incorporation into borosilicate glass enhances functional properties such as bioactivity, antibacterial efficacy, or luminescence, it concurrently compromises optical transparency. Fathy Abdel-Wahab *et al.*<sup>35</sup> doped borosilicate glass with chromium and co-doped it with ZnO. The authors reported a clear reduction in visible light transmittance as ZnO concentration increases, which disrupts the glass network and promotes absorption and scattering. Similarly, Mohd Hafiz Mohd Zaid *et al.*<sup>36</sup> explored the effect of ZnO doping in soda-lime silicate glasses and observed that ZnO incorporation

shows band-gap narrowing<sup>37</sup> and elevated visible absorption, suggesting comparable spectral effects in soda lime glass systems. Furthermore, terbium-activated zinc borosilicate matrices form ZnO and willemite crystalline phases upon heat treatment, indicating that phase separation compromises clarity even as luminescence intensifies, as represented by Sena Dayioglugul *et al.*<sup>37</sup>

Collectively, these studies confirm that ZnO incorporation at varying concentrations tends to reduce the optical clarity of borosilicate glass, posing a trade-off between functionality and transparency of such glass-based packaging. In addition to antibacterial activity, ZnO-infused barrier coatings improve the packaging film's resistance to moisture, oxygen, and grease, all of which are essential for maximising shelf life. Nanostructured ZnO thin films have variable thickness and morphology that directly affect their antibacterial activity, particularly when deposited using methods like DC reactive sputtering or RF magnetron sputtering.<sup>38,39</sup> The ZnO nano particles (NPs) can be synthesized using various green synthesis methods and are coated onto the packaging surface.<sup>40</sup> Fabricating thin ZnO films *via* techniques such as RF magnetron sputtering in food packaging offers several advantages, including uniform coating, high adhesion, and room-temperature processing. These coatings can be deposited on various packaging substrates like borosilicate and soda-lime glass to enhance not only their antimicrobial performance but also to preserve the sensory and nutritional qualities of the food contents by inhibiting microbial growth. Despite the growing interest in ZnO coatings for biomedical<sup>41</sup> and optoelectronic applications,<sup>42,43</sup> their potential in food packaging remains limited as a coating on glass for packaging and is yet to be explored. Although direct coating on glass substrates is limited in this area, a few studies such as functionalization of silver (Ag) for Al-doped ZnO coatings on polylactic acid (PLA) substrates,<sup>44</sup> ZnO/graphene oxide/montmorillonite antibacterial coating on (polyethylene) PE substrates,<sup>35</sup> and coatings for food contact surfaces<sup>13,45</sup> highlight the wide potential and need for further research.

The current study investigates the structural, morphological, optical, and antibacterial properties of RF magnetron sputtered ZnO coating on borosilicate glass substrates. By analysing the influence of coating texture, transparency, stability, and its antibacterial activity against clinically relevant bacteria, this work aims to contribute to the development of safer, longer-lasting, and more sustainable food packaging systems.

## 2 Experimental details

### 2.1 Fabrication of ZnO thin film coating on 7059 glass substrates *via* sputtering

The glass substrate was cleaned using the standard substrate cleaning method.<sup>46</sup> The substrate was first cleaned using lobe-line solution and then sonicated using DI water and then acetone for 10 minutes. The cleaned substrates were then dried in a nitrogen environment, ensuring an ultra-clean substrate surface. A thin coating of ZnO was deposited using RF magnetron sputtering (HHV-high vacuum model) at room temperature. Fig. 1 represents the schematic diagram of RF magnetron



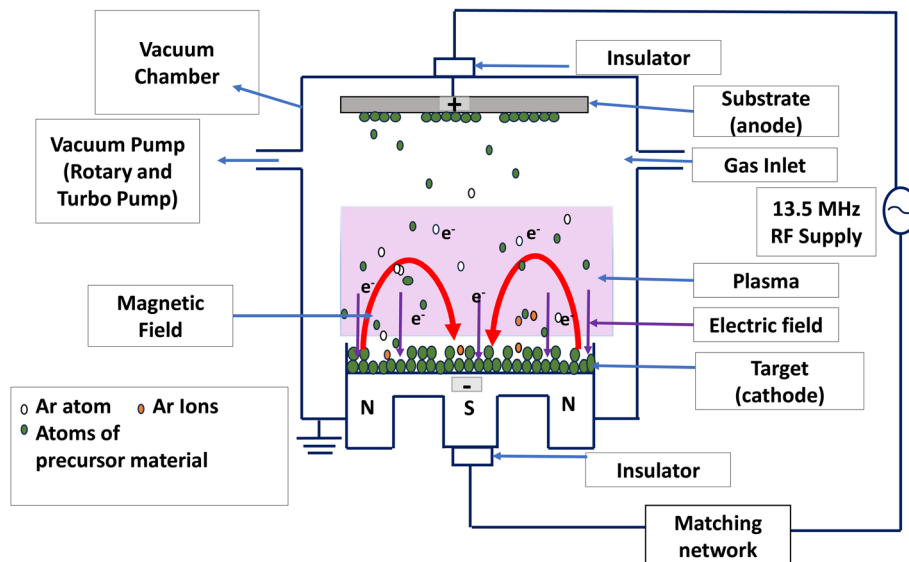


Fig. 1 Schematic representation of RF magnetron sputtering.

sputtering. It is a thin-film deposition technique that uses radio frequency power. It is widely used for depositing insulating or composite materials with excellent adhesion, density, and compositional control. ZnO (~99.99%) purity target of diameter 75 mm was used as a cathode to sputter deposit the coating. The distance between the target and substrate was kept at ~10 cm. Substrates (glass 7059) were placed on the anode with the help of copper tapes. The base pressure in the vacuum chamber of  $\sim 10^{-6}$  mbar was achieved using a turbo molecular and rotary pump combination. The argon gas was purged inside the chamber, and a constant argon pressure of  $4 \times 10^{-2}$  mbar was maintained in the chamber. The RF power of 100 W was applied between the cathode and anode for 120 minutes. Argon gas was used as a sputter gas for generating the plasma. High-energy Ar ions in the plasma bombarded the ZnO target, and ZnO was deposited on the substrate by momentum transfer. The colour of the plasma depends on incident ions and the cathode material. In this case, the colour was pink. In this study, a ZnO coating was developed on 7059 corning borosilicate glass substrates. Two types of coatings were fabricated: (a) one side coated with a step (created using Teflon tape), (b) both sides coated.

## 2.2 Characterization and testing

The thickness of the coating was determined using a depth profilometer (Bruker DektakXT). The adhesion of the film with the substrate was tested by the peel test method using Kapton tape. The tape was placed on the coated substrate and manually peeled off at an angle of  $90^\circ$ . The crystallographic structure of the film was determined with an X-ray diffractometer (Empyrean PANalytics diffractometer). The optical properties were determined with the help of a UV-visible spectrometer (PerkinElmer LAMBDA 365), and the transmittance spectra were taken between 350 and 800 nm. The structural morphology and particle size were studied using field effect scanning electron

microscopy (FE-SEM, Hitachi S-4800) and analysed through ImageJ software. The contact angle of the coatings was determined through a goniometer contact angle measurement instrument (ramè-hart Instrument Co.). The antibacterial activity of the coated thin films was determined against *Staphylococcus aureus* (*S. aureus*) and *Escherichia coli* (*E. coli*), using quantitative microbiological techniques.

## 3 Results and discussion

### 3.1 X-ray diffraction: crystallographic analysis

The structural morphology of the ZnO thin films was determined *via* the glancing angle X-ray diffraction method. Copper  $K_{\alpha 1}$  radiation with 0.154 nm was used over a scan range of  $(2\theta)$   $20^\circ$  to  $80^\circ$  with a scan rate of  $0.02^\circ \text{ s}^{-1}$ . A hexagonal phase with a wurtzite structure (JCPDS file no. 36-1451) has been identified<sup>47</sup> in the deposited zinc oxide thin film (Fig. 2). The ZnO film is textured along the (002) plane with low-intensity peaks at (102), (103), and (112) as shown in Fig. 2. The preferential orientation of the films was observed to be the (002) plane; the texture coefficient (TC) was determined using eqn (i)<sup>48</sup> and is represented in Fig. 2 (inset).

$$TC_{(hkl)} = \frac{\left( \frac{I_{o(hkl)}}{I_{s(hkl)}} \right)}{\left[ \frac{1}{n} \sum_{n=1,2,3} \left( \frac{I_{o(hkl)}}{I_{s(hkl)}} \right) \right]} \quad (i)$$

where  $I_{s(hkl)}$ , is the standard XRD intensity of the corresponding  $(hkl)$  peak,  $I_{o(hkl)}$  is the observed intensity, and  $n$  is the total number of peaks observed. The grain distribution of the coated films is determined from the texture coefficient; if  $TC_{(hkl)} < 1$ , then grain development is random; if  $TC_{(hkl)} > 1$ , then grain growth in the  $(hkl)$  plane is preferential. The obtained  $TC_{(002)}$  is 3.868 which is the highest among the observed peaks, hence it can be concluded that the (002) plane is the most preferred



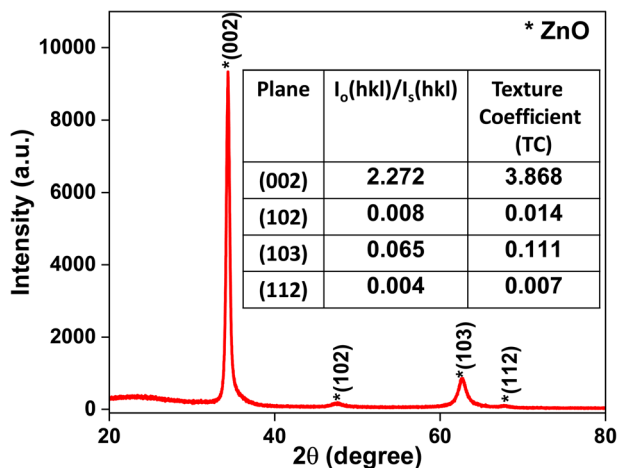


Fig. 2 X-ray diffraction pattern of sputtered coated ZnO thin films, the film is textured along the (002) plane with the ZnO hexagonal phase identified using JCPDS card no. 36-1451. The inset table represents the texture coefficient value of identified planes.

orientation of the crystal growth. The preferential texturing is related to the minimized internal stress and surface energy in the films as discussed by authors Norifumi Fujimura *et al.*,<sup>49</sup> and Dinghua Bao *et al.*<sup>50</sup> Enmao Xiang *et al.* reported a significant contribution of surface roughness to antibacterial properties in Zn metal films,<sup>51</sup> indicating the importance of surface in antibacterial properties. The average crystallite size of the particles was determined using the Scherrer equation

$$D = \frac{k\lambda}{\beta \cos \theta} \quad (\text{ii})$$

where  $\beta$  is the full width at half maxima value of the diffraction peak (in radian),  $K$  is the numerical factor (constant = 0.9) which depends on the shape of the crystallite,  $\lambda$  is the wavelength of the X-ray source, and  $\theta$  is the diffraction angle.<sup>52–54</sup> The average crystallite size of ZnO obtained by using eqn (ii) is approximately 18 nm for ZnO coating on 7059 corning glass substrates.

### 3.2 Structural morphology and elemental analysis

**3.2.1 Thickness and adhesion test.** The thickness of the deposited thin films was determined using a depth profilometer

(Bruker DektakXT model). The measured thickness of the coatings is ~50 nm. P. Carvalho *et al.*<sup>55</sup> employed the cross-sectional SEM analysis method to estimate the thickness of the DC sputtered coating of pure ZnO and Ag-doped ZnO on a silicon substrate. The authors observed that the coating thickness varies between 50 and 100 nm during a 2-hour deposition time and has a similar granular morphology to that reported in this work in the Scanning electron microscopy section.

The adhesion of the coating with the substrate was tested using Kapton tape, the tape was placed on the coating and peeled off, as represented in Fig. 3 (both 48 Mega Pixel camera and optical images (20× magnification) were taken for better representation). The coating remained intact on the substrate, thus showing strong adhesion of the ZnO coating on the glass substrate. In further sections, the stability of these coatings in an aquatic environment over a long period is also studied.

**3.2.2 Optical properties.** The optical properties were determined from the transmittance measurement of the synthesized zinc oxide coatings for both side coated substrates in the range of 280–800 nm *via* a UV-visible spectrometer (PerkinElmer LAMBDA 365), as shown in Fig. 4 and the inset.

The UV-visible transmittance spectrum (Fig. 4) of the ZnO coating reveals a sharp edge around 375–400 nm and transmittance above 80%. This indicates strong absorption in the UV region, effectively blocking harmful ultraviolet radiation through the sputtered film. The transmittance increases steeply to 85% and then gradually approaches nearly 100% around 700 nm in the visible range. This high optical transparency confirms the suitability of the ZnO coating for applications in UV-shielding films as well. A. Ismail *et al.*<sup>56</sup> studied the optical properties of ZnO coating, which was RF sputtered at different powers (150–250 W) and obtained an optical transmittance of 70–80%. Alireza Samavati *et al.*<sup>57</sup> also studied the effect of thickness on the optical properties of RF-deposited ZnO films and got similar results. The minimal absorption and scattering in this region also suggest good film uniformity and surface smoothness. These findings are consistent with previous reports<sup>57</sup> that highlight the excellent optical properties of ZnO thin films, including their wide bandgap and high transmittance in the visible spectrum.

**3.2.3 Scanning electron microscopy.** The structural morphology of the coated film on the 7059-borosilicate

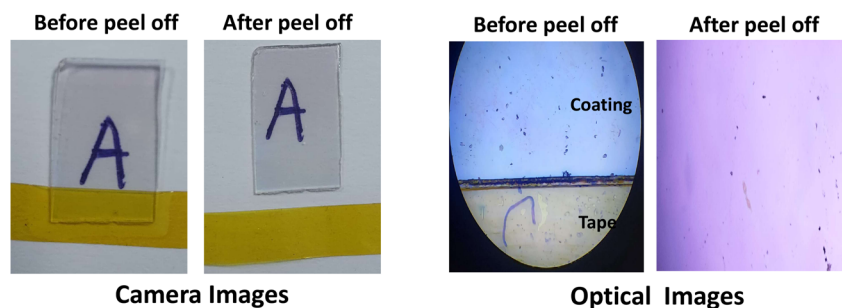


Fig. 3 Visual inspection images of ZnO coatings taken using a camera (48 Mega Pixel) and optical images (20× magnification) before and after the peel-off test, showing no change indicating the highly adhesive ZnO coating deposited on the glass substrate.



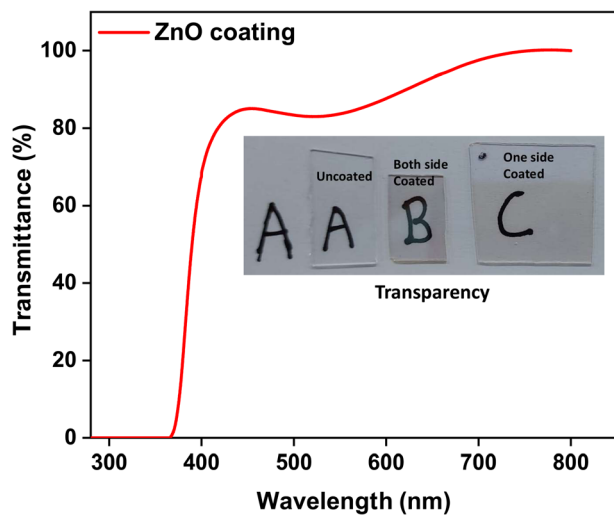


Fig. 4 UV-visible transmittance spectra of both side ZnO coated 7059 glass substrates; the inset shows the images of the transparent coatings.

substrate was observed using field emission scanning electron microscopy (FE-SEM). The sputtered ZnO coating exhibits compact grain morphologies (Fig. 5a), indicating the formation of densely packed nano crystallites. The EDX spectrum (Fig. 5b) confirms the presence of Zn and O as the primary constituents of the coated films, with no significant impurities detected. The

particle size distribution was determined *via* FE-SEM analysis (ImageJ software);<sup>58</sup> the particle size distribution analysis (Fig. 5c) revealed an average particle diameter of  $21.52 \pm 1.27$  nm. The Gaussian fit curve indicates a narrow size distribution, suggesting controlled growth conditions during the deposition, resulting in uniform particle size. The borosilicate substrate acts as a nucleation site at room temperature and 100 W applied RF power. P. Carvalho<sup>55</sup> observed similar granular morphologies of ZnO NPs grown at room temperature *via* DC reactive magnetron sputtering. This nanoscale grain structure was reported to be favourable for improving surface-dependent functionalities, such as antimicrobial and wettability behaviour.<sup>40,59,60</sup>

### 3.3 Antibacterial testing

The coated ZnO thin films were tested for the desired potential use as an antibacterial coating. The antimicrobial efficacy of ZnO coating was tested against two microorganisms, Gram-positive *Staphylococcus aureus* and Gram-negative *Escherichia coli* bacterial strains for a 24-hour incubation period. The nutrient broth and Muller Hinton Agar were procured from Hi-media. To prepare the bacterial cultures, 0.13 g of nutrient broth powder was dissolved in 10 mL of distilled water and sterilized. The bacterial inoculum was introduced into the broth and incubated at  $36 \pm 1$  °C for 24 hours under aerobic conditions to ensure active bacterial growth.

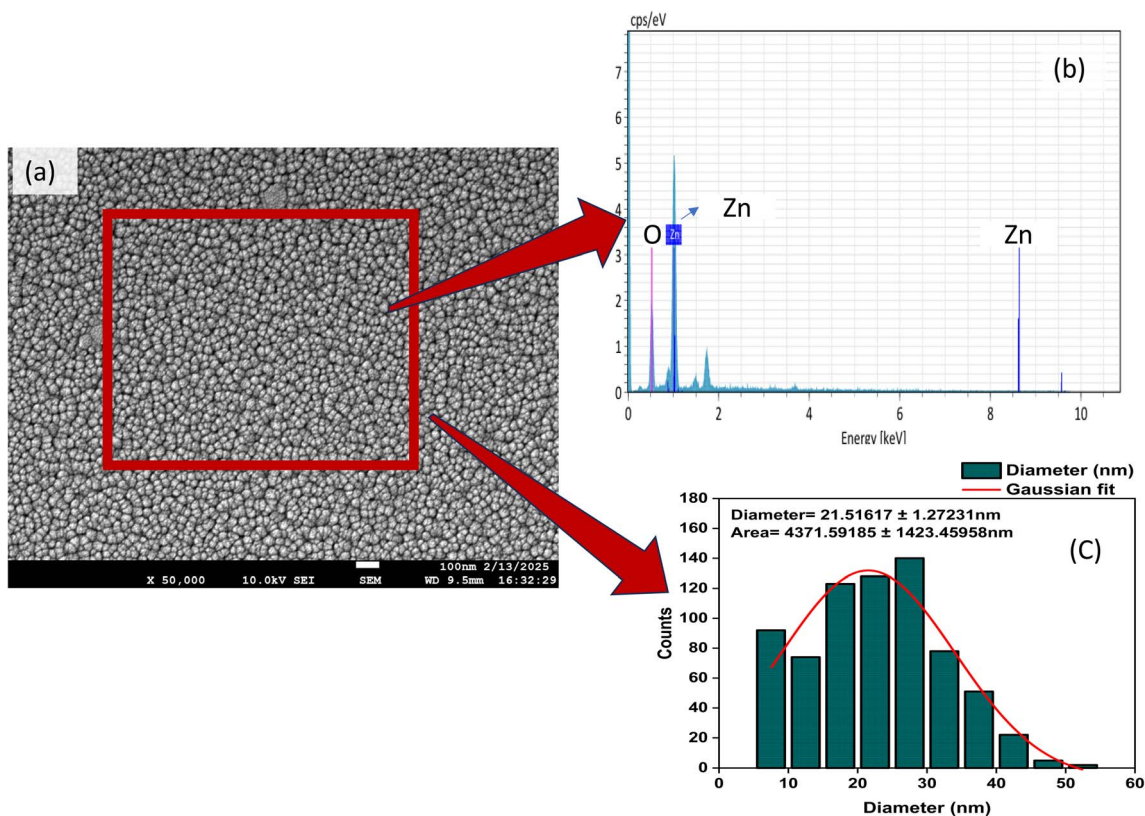


Fig. 5 FE-SEM images of ZnO coated film at 100 nm scale (a–c): (a) FE-SEM image of coated 7059 substrates at 50 000 $\times$  magnification; (b) EDX spectrum confirms the presence of Zn and O; (c) particle size distribution as observed *via* FE-SEM analysis.



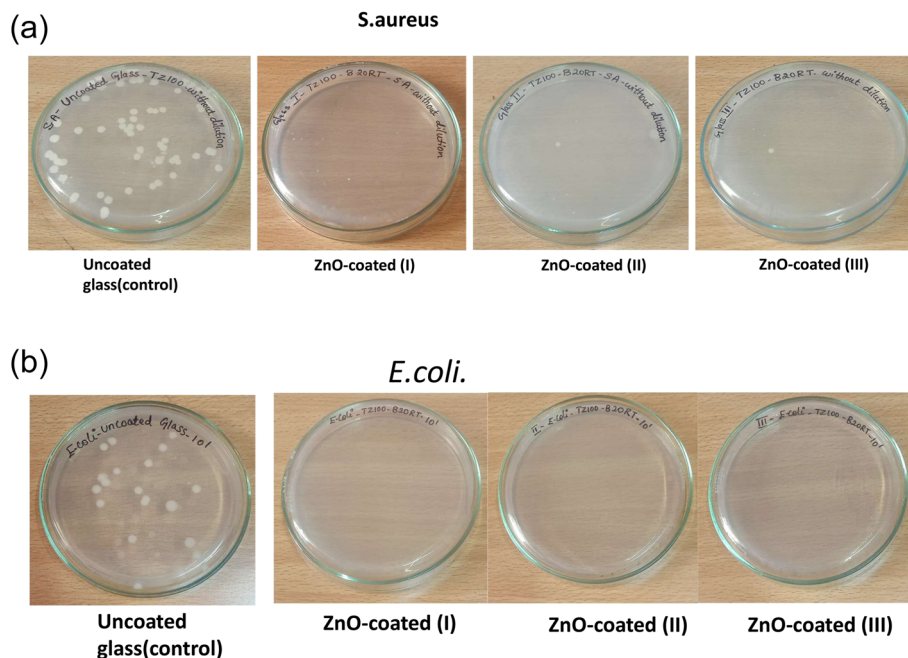


Fig. 6 The antibacterial activity on the agar plate after an incubation period of 24 hours, for (a) *S. aureus* and (b) *E. coli* for ZnO-coated glass thin films. The uncoated glass substrate is the control.

Following incubation, 1 mL of the bacterial culture was diluted in 9 mL of sterile distilled water to achieve a  $10^5$ -fold dilution. Then, 100  $\mu$ L of this diluted bacterial suspension was pipetted onto the surface of the test films. The samples were incubated in a humidified chamber at 37 °C for 24 hours to facilitate bacterial adhesion and interaction with the coated surface. Post-incubation, the bacterial inoculum was recovered by gently resuspending the remaining liquid on the film surface. The recovered suspension underwent a series of tenfold serial dilutions ( $10^{-1}$  to  $10^{-4}$ ). From each dilution, 50  $\mu$ L was plated onto Mueller Hinton Agar plates, which were subsequently incubated at 37 °C for 24 hours. The experiment was performed in triplets represented here as ZnO coated (I), (II), (III) for each bacterium, as shown in Fig. 6a and b.

Colony-forming units (CFUs) were calculated from the agar plates corresponding to each dilution. The CFU concentration in the recovered suspension was calculated using the following formula:

$$\text{CFU} = \frac{\text{number of colonies} \times \text{dilution factor}}{\text{volume plated (mL)}}$$

Fig. 7(a) represents the percentage killing in the coated film in comparison to the control (uncoated 7059 glass) substrate. The results revealed a highly potent bactericidal effect against *E. coli*, with 100% killing, followed by 83.25% against *S. aureus*. The particle size and morphology play an important role in enhancing the antibacterial activity of ZnO nanomaterials

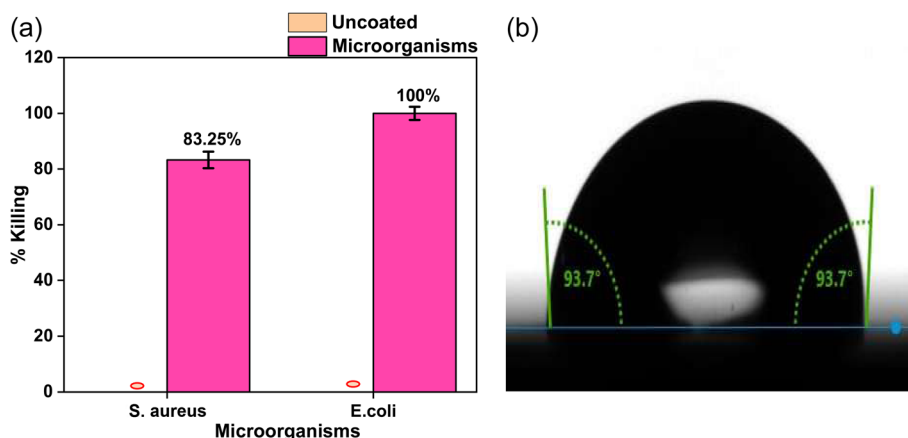


Fig. 7 (a) Antibacterial activity of the coated sample compared to the uncoated sample. (b) Contact angle image of ZnO coating on the 7059 substrate.



against Gram-negative and Gram-positive bacteria.<sup>61</sup> Surface morphology is known to vary with deposition parameters for ZnO NPs.<sup>62</sup> The deposited ZnO nanoparticles grown by RF magnetron sputtering at room temperature have shown the island growth of spheroid-like morphologies (Fig. 5a). The grown ZnO nanoparticles have a high specific surface area, due to a smaller particle size of 18–21 nm, as confirmed on the basis of XRD and FE-SEM results, with a slightly hydrophobic surface having a contact angle of 93.7° as shown in Fig. 7b. As the observed hydrophobicity of the coating is very small according to the contact angle, there is high probability of bacterial colonies on the coating. The enhanced *S. aureus* and *E. coli* killing with respect to control indicates an active killing.

The NPs in the coating have a smaller particle size which may easily rupture the cell wall of Gram-negative bacteria with the thickness of the cell wall varying between 7 and 8 nm, as demonstrated by Rafel Alvarez Chimal *et al.* in their study. The authors have demonstrated the influence of particle size on the antibacterial activity of green-synthesized ZnO and also confirmed the interaction between ZnO NPs and bacterial protein through molecular docking analysis.<sup>60</sup> The fabricated ZnO film having a smaller particle size may be able to rupture the cell wall of bacteria, resulting in reduced bacterial activity. The molecular docking analysis experiment may be undertaken in the future to confirm the bacterial killing mechanism for the RF sputtered ZnO coated glass. Krishna R Raghupathi *et al.*<sup>63</sup> studied the size dependent bacterial inhibition of ZnO NPs and their mechanism against *S. aureus*, and concluded that the killing was inversely proportional to the size of NPs. Nataliya Babayevska *et al.*<sup>64</sup> have also shown similar results for lower particle size nanoparticles and nanorods of ZnO, having comparable killing of bacteria against uncoated samples for both *S. aureus* and *E. coli*. The killing or inhibition of bacteria in the literature has been summarized and represented in Table 1.

A scalable and affordable option for glass container applications in the food and pharmaceutical industries is soda-lime glass doped with 5% ZnO, which demonstrated 99.82% inactivation of *E. coli*.<sup>8</sup> The same group doped 2% Ag<sub>2</sub>O and showed 99.99% killing of *E. coli* in antibacterial soda lime glass in comparison with undoped glass.<sup>65</sup> Furthermore, they reported that 5% ZnO incorporation in borosilicate glass for pharmaceutical packaging leads to the antibacterial inactivation of 99.29% *E. coli*.<sup>66</sup> The study further concludes that the leaching of ZnO may occur, which is below the toxicity level. In comparison to the literature, *E. coli* and *S. aureus* killing is greater in our results (see Table 1). The smaller particle size of

ZnO crystallite in the deposited coating along with the strong texturing along (002) have probably contributed to 100% and 83.25% killing of *E. coli* and *S. aureus*, respectively.

### 3.4 Stability evaluation of ZnO coating under simulated moisture conditions

Hajer Rokbani *et al.*<sup>67</sup> evaluated the long- and short-term antibacterial properties of low-density PE (polyethylene) films coated with ZnO NPs, for food packaging. ZnO-coated LDPE-g-AM (Bynel) films exhibited strong and long-lasting antibacterial activity, especially against *E. coli*, due to uniform nanoparticle distribution and strong surface adhesion. These films on PE remained effective even after eight months and ultrasonic washing, unlike standard LDPE films. This highlights their potential as durable, passive antibacterial coatings for food packaging applications.

To evaluate the long-term stability of the ZnO coated films under simulated aqueous environment conditions for food packaging, both side coated substrates without a stepped edge were completely immersed in 20 ml of deionized (DI) water in a glass beaker and kept in an isolated environment for over 120 hours. Before the stability test, the coating was kept under ambient conditions for 5 months after deposition. The coatings were evaluated across the edge and in the middle region of the surface to distinguish any variation, allowing visual tracking of the degradation (Fig. 8a–d). Microscopic images of the coating were taken to understand the variation in thickness/surface morphology at defined time intervals (0, 24, 72 and 120 hours) at 20× magnification.

In the initial hour (zeroth hour, Fig. 8a), the coated surface appeared uniform with a clearly defined interface and no evidence of defects, pits or delamination. The coating adhered well to the substrate, indicating satisfactory film integrity even after being exposed to an ambient environment for more than 5 months of deposition. Upon 24 hours of immersion in DI water (extreme exposure to moisture and oxidising conditions) (Fig. 7b), a subtle variation can be observed near the edge, potentially due to the initial hydration effect. Since the coating used exhibits a water contact angle of 93.7°, with a slightly hydrophobic surface, it gives a barrier, though weak, to water retention in the initial hours, without substantial mechanical failure. The continuous exposure to moisture did not show much surface transformation in 72 hours (Fig. 8c). After 120 hours (Fig. 8d), the subtle beginning of delamination was observed at the coating edge, whereas the middle region is observed to be stable. Álvaro Garrido-López *et al.*,<sup>11</sup> studied the

Table 1 Comparison of bacterial killing in active packaging

Bacteria	Material and technique	% Killing	Particle size	References
<i>E. coli</i>	ZnO doped soda lime glass	99.82%	—	8
<i>E. coli</i>	Ag <sub>2</sub> O doped soda lime glass	99.99%	—	65
<i>E. coli</i>	5% ZnO doped borosilicate glass	99.29%	—	66
<i>E. coli</i>	Sputter coated ZnO	100%	18–21 nm	This paper
<i>S. aureus</i>	Green-synthesized ZnO	Higher zone of inhibition	(~12 nm)	63
<i>S. aureus</i>	Green-synthesized ZnO	Higher zone of inhibition	(~4–10 nm)	60
<i>S. aureus</i>	Sputter coated ZnO	83.25%	18–21 nm	This paper



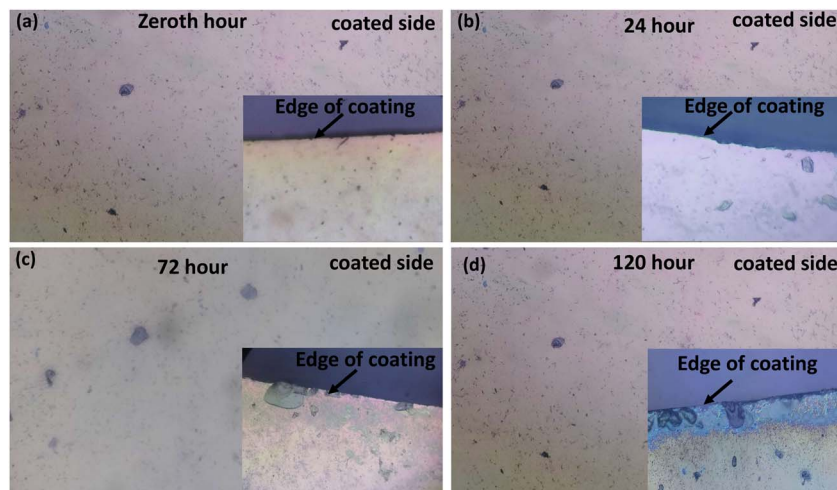


Fig. 8 Optical images of ZnO coatings after immersion in DI water (a) 20 $\times$  magnification coating before immersion, (b) after 24 hours of immersion, (c) after 72 hours of immersion (d) after 120 hours of immersion. The inset images represent the edges of the coating.

delamination mechanisms of multilayer packaging of aluminium foil and polyethylene (PE) *via* XPS, FTIR and ToF SIMS analysis. Their results indicated that chemical delamination was mostly caused by the reaction of migrated molecules with adhesive-aluminium bonds, whereas the breaking of van der Waals bonds leads to delamination of extruded materials. The authors reported similar degradation pathways in oxide and polymer hybrid coatings, where water uptake leads to hydrolytic cleavage or loss of adhesion. Similarly, Ghislain Fotie *et al.*<sup>68</sup> evaluated the effect of moisture on cellulose nanocrystals (CNCs) used in food packaging for high gas barrier protection. They coated a PET substrate with CNCs, and the solubility results indicated that the integration of hydrophobic coatings would lead to the reduction of the delamination process.

This study highlights the coatings' stability for food packaging used for a shorter duration. Additionally, the promising adhesion even after 5 months in the ambient environment strongly suggests the possibility of this coated glass in packaging to store dry food products. Enhancing the moisture resistance of the coating through improved cross-linking, hydrophobic modification, or incorporation of barrier layers would further improve the coating stability over a longer duration. Moreover, the time-based imaging approach provides a simple yet effective technique for evaluating coating reliability in the presence of environmental stimulants. These antibacterial, transmittance and stability results are highly encouraging to consider the ZnO sputter coating on borosilicate as sustainable, multifunctional materials for active food packaging applications. This work shall be extended in future to other wrapping films used for packaging.

## 4 Conclusion

ZnO nanofilms were successfully coated on 7059 corning glass *via* RF sputtered deposition. The deposited film is highly dense, exhibits homogeneous granular morphology and textured (TC = 3.87) along the 002 plane having a crystallite size of 18 nm. FE-SEM revealed a grain size of  $\sim$ 21 nm diameter. Antimicrobial

studies showed excellent activity against *E. coli* (100%) and *S. aureus* (83.25%). The preferential film orientation and smaller particle size could be the reason for the observed improved antibacterial activity of the ZnO material. The stability study indicated strong adhesion even after exposure to the ambient environment for approximately 5 months, as well as moisture conditions for a longer duration of 120 hours. The fabricated ZnO coatings are transparent and highly adhesive. The RF-sputtered ZnO coatings can be proposed as a potential coating on glass packaging used in both healthcare as well as food industries.

## Author contributions

JP performed the experiments, data analysis, and conceptualization, and wrote the original draft. SR reviewed, edited, evaluated and documented the results, and supervised the work. All authors have read and agreed to the published version of the manuscript.

## Conflicts of interest

We wish to confirm that there are no known conflicts of interest associated with this publication that could have influenced its outcome.

## Data availability

The datasets generated during the current study and supporting the findings of this study are included in this paper.

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