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Effect of silver ion implantation on antibacterial ability of polyethylene food packing films

Naiyan Lu^a, Zhe Chen^{a,*}, Wei Zhang^a, Guofeng Yang^a, Qingrun Liu^e, Roman Böttger^d, Shengqiang Zhou^d, Yu Liu^{b,c,d,*}

^a State Key Laboratory of Food Science and Technology, Jiangnan University, Wuxi, Jiangsu 214122, China

^b Microsoft Quantum Materials Lab Copenhagen, Lyngby 2800, Denmark

^c Center for Quantum Devices, Niels Bohr Institute, University of Copenhagen, Copenhagen 2100, Denmark

^d Helmholtz-Zentrum Dresden-Rossendorf, Institute of Ion Beam Physics and Materials Research, Dresden 01328, Germany

^e State Key Laboratory of Food Nutrition and Safety, College of Food Science and Engineering, Tianjin University of Science & Technology, Tianjin 300457, China

Abstract: Bacterial adhesion on medical instruments' and food packages' surfaces causes implanted infections, food spoilage and human disease, therefore attracts a lot of attention in the field of medical and food applications. Containing the initial adhesion of bacteria on the surface of the material plays an important role in reducing potential safety hazards. In this work, we investigate the influence of silver ion implantation with different doses on the antibacterial performance of the polyethylene (PE) films. It is found out that silver ion implantation will not color the PE films but can improve their surface hydrophilicity. The silver-implanted PE films show the ability to inhibit bacterial adhesion and have the bactericidal effect, both of which can be improved with increasing silver implantation dose. This method also proves relatively safe, because the silver ions are relatively stable. The results will introduce potential applications for ion implantation in the food packing and food accessible materials.

Keywords: polyethylene; ion implantation; surface modification; bacterial adhesion; antibacterial ability; hydrophilicity.

*Corresponding author.

E-mail addresses: jiz.chen@foxmail.com (Z. Chen), yu.liu@nbi.ku.dk (Y. Liu)

27 **1. Introduction**

28 Bacterial adhesion is the initial stage of pathogen biofilm formation and the
29 formation of biofilms is the main cause of food-borne diseases (Erdem et al., 2017).
30 During the processing of food, the food can be contaminated by microorganisms and
31 become the source of cross-infection (Trachoo, Frank, & Stern, 2002). Moreover,
32 microorganisms will be attached to food processing materials, conveying pipes, filter
33 membranes and heaters to increase work resistance and reduce work efficiency (Miao
34 et al., 2017), resulting in huge economic losses (Scharff, 2012; Wei, Helm,
35 Corner-Walker, & Hou, 2006).

36 The biofilm formation process is postulated to have several major stages,
37 including (I) initial reversible attachment, (II) a transition to irreversible attachment,
38 (III) biofilm architecture development, (IV) mature biofilm formation, and (V) cell
39 dispersion (Chao & Zhang, 2011; Fysun, Kern, Wilke, & Langowski, 2019; Renner &
40 Weibel, 2011). Because mature biofilms have strong resistance to their living
41 environment, and conventional fungicides cannot remove biofilms effectively (Bridier,
42 Briandet, Thomas, & Dubois-Brissonnet, 2011; Escribano-Viana et al., 2018; Houari
43 & Di Martino, 2007; Otter, Yezli, Salkeld, & French, 2013; Sav et al., 2018).
44 Therefore, it is very important to inhibit bacterial adhesion and to control the biofilm
45 formation in its early stage (Brecher & Hay, 2005; Lindsay & Von Holy, 2006;
46 Shorten, Pleasants, & Soboleva, 2006; Zhang, Chao, Shih, Li, & Fang, 2011).

47 Ion implantation has become a hotspot in material modification methods in
48 recent years (Li et al., 2019; Shiao et al., 2019; Xia et al., 2018; Zheng, Qian, & Liu,
49 2020). The ions generated by an ion source are shot to the surface of the materials at
50 high speed. The energetic ions enter the surface, collide with the atoms in the solid,
51 and finally stop in the materials and are embedded beneath the surface. During this
52 process, some atoms are displaced, causing changes of the surface composition,
53 structures, or properties of the materials, and thus serving purposes of modifications
54 (Hu, Liu, Gan, & Long, 2019; Li et al., 2019). Especially, the ion implantation
55 technology can adjust hydrophobicity of the surface (Price, Waters, Williams, Lewis,
56 & Stickler, 2002; Tsuji et al., 2007) and improve the tissue compatibility by changing
57 the chemical composition of the material surface (Wang et al., 2014), which can
58 influence the materials' antibacterial adhesion. Hu et al. (Hu, Liu, Gan, & Long, 2019)
59 have injected Fe³⁺ into graphene to improve the stability of the prepared material and
60 its ability of inhibiting *E. coli*. Price C. et al. (Price, Waters, Williams, Lewis, &
61 Stickler, 2002) have used argon-plasma bombardment for surface modification, which

62 has caused the incorporation of either hydrophilic or hydrophobic functional groups
63 onto the surface. It turns out that the adhesion of *Candida* has been reduced. Zahran M.
64 K. et al. (Zahran, Ahmed, & El-Rafie, 2014) have investigated antimicrobial activities
65 by applying AgNPs–alginate composite on cotton fabric. The treated fabrics show the
66 excellent antibacterial activity against *P. aeruginosa*, *S. aureus* and *E. coli*.

67 As a broad-spectrum antibacterial agent, silver is one of the most commonly used
68 antibacterial agents at present (Marchetti et al., 2016; Sahai, Gaval, & Bhat, 2020;
69 Sanchez-Valdes et al., 2018; Zheng et al., 2016). Tambur P et al. (Tambur, Bhagawan,
70 Kumari, & Kasa, 2020) have decorated silver nanoparticles on functionalized
71 multi-walled carbon nanotubes, which has proved to be effective against *Bacillus*
72 *subtilis*, *S. aureus*, *E. coli*, *P. aeruginosa*. Oses J. et al. (Oses et al., 2014) have found
73 that silver ions implanted into CrN layers have positive antibacterial effects on *S.*
74 *aureus* and *E. coli*. However, there are concerns about whether silver can be applied in
75 the food field efficiently and safely because it is a kind of heavy metal which can
76 endanger human bodies (Marambio-Jones & Hoek, 2010; Zhang, Wang, & Levanen,
77 2013). Hadrup, N. et al (Hadrup, Sharma, Loeschner, & Jacobsen, 2020) evaluated
78 silver to be genotoxic in vitro in mammalian cells. When silver ions are used in food
79 packing or contact materials, there will be potential risks. Therefore, it is important to
80 guarantee the antibacterial ability of silver ion fungicides and improve their safety at
81 the same time. In this study, ion implantation was used to introduce silver ions at
82 different doses in the food packing material – PE films (Figure 1). The treated films
83 show the ability of inhibiting bacterial adhesion and bacterial growth on the surface.
84 The safety of the implanted materials was evaluated with dissolution experiments.

85 **2. Materials and methods**

86 2.1. Materials

87 *E. coli* BL21, *S. aureus* 6538, and *P. aeruginosa* ATCC 9027 were purchased
88 from China General Microbiological Culture Collection (CGMCC, Beijing, China).
89 Luria Bertani (LB) agar and broth, M63 medium and tryptic soy broth (TSB) medium
90 were purchased from Hepobio Company, Zhengzhou, Henan, China. All other
91 chemical reagents (AR grade) were purchased from Sinopharm Chemical Reagent Co.,
92 Ltd. (Shanghai, China) and used as received.

93 2.2. Silver ion implantation

94 We used the ion implanter (High Voltage Engineering Europa B.V.,

95 Model B8385) at the Ion Beam Center, Helmholtz-Zentrum Dresden-Rossendorf,
96 Germany. It can provide all kinds of stable ions with the energy from 10 to 500 keV.
97 The fluence can be from $5e^{10}$ to $5e^{16}$ cm^{-2} . In detail, the Ag ions were produced by a
98 IHC Bernas ion source. They were electrostatically accelerated to the designed high
99 voltage. Together with a magnetic analyzer, ions can be selected according to their
100 atomic mass. To ensure the uniformity, the beam was rastered over the sample. The
101 PE films with area of $100 \text{ mm} \times 100 \text{ mm}$ were implanted with different doses of silver
102 ions. The beam energy was set to 190 keV, the temperature of PE films was kept less
103 than 50°C , and the implantation angle was 7° . The injection doses were as follows,
104 sample 0: none, sample 1: $5 \times 10^{12} \text{ cm}^{-2}$, sample 2: $1 \times 10^{13} \text{ cm}^{-2}$, sample 3: $5 \times 10^{13} \text{ cm}^{-2}$,
105 sample 4: $1 \times 10^{14} \text{ cm}^{-2}$, sample 5: $5 \times 10^{14} \text{ cm}^{-2}$, and sample 6: $1 \times 10^{15} \text{ cm}^{-2}$.

106 2.3. Contact angle measurement

107 The water contact angles of PE film were measured using a Krüss DSA100 Drop
108 Analysis System (Krüss GmbH, Hamburg, Germany) at room temperature ($\sim 20^\circ\text{C}$). 1
109 μL of DI water was added on the film surface. The contact angles were calculated
110 using the DSA Registered Version software provided with the instrument. And the
111 final result of water contact angle values was obtained by calculating the average of
112 values in triplicate at different positions of the PE film after ion implantation.

113 2.4. Bacterial strains and bacterial attachment experiments

114 *E. coli* BL21, *S. aureus* 6538, and *P. aeruginosa* ATCC 9027 were inoculated
115 into LB agar plates, and then transferred into LB broth with agitation at 37°C , 200
116 rpm for 12 h. The incubated cells were harvested in log phase, centrifuged (5000 g)
117 for 15 min at 4°C , and washed twice with 0.08 M phosphate buffer saline (PBS, pH
118 7.4). The *E. coli* suspensions were diluted with PBS until the optical density at 600
119 nm equaled 0.6 ± 0.05 (approximately 10^8 CFU/mL), and transferred into M63 medium
120 with 1% (Friedlander et al., 2013). *S. aureus* and *P. aeruginosa* were added into TSB
121 medium at the same concentration. PE films were placed at the bottom of a 6-well
122 tissue culture plate at least in triplicate; M63 medium with *E. coli*, TSB medium with
123 *S. aureus* or *P. aeruginosa* were poured into the 6-well tissue culture plate to cover the
124 film. The plates were incubated at 37°C for 5 h.

125 2.5. Determination of resistance to bacterial growth

126 *E. coli* BL21, *S. aureus* 6538, and *P. aeruginosa* ATCC 9027 were inoculated
127 into LB agar plates, and then transferred into LB broth with agitation at 37°C , 200

128 rpm for 12 h. The incubated cells were harvested in log phase. The three strains were
129 separately diluted to make the concentration of the bacterial suspension 10^5 - 10^6
130 CFU/mL.

131 Refer to "Antibacterial products-Test for antimicrobial activity and Effects"
132 (Japanese Industrial Standard JIS Z2801-2000). Each PE film after the silver ion
133 implantation was cut into samples with a size of 1 cm×1 cm, washed with 70%
134 ethanol, dried with nitrogen. 100 μ L bacterial suspension was added on the surface of
135 the film, and then it was covered with a sterile film.

136 The bacterial solution was evenly coated on the surface of the film, and the
137 relative humidity of the incubator was adjusted to be more than 90%. After incubating
138 in a 37°C incubator for 24 h, the film was taken out and rinsed with sterile
139 physiological saline. Then, the eluent was inoculated into LB solid medium to
140 cultivate in a 37°C incubator for 24 h. After cultivating, plate colony count was
141 performed and recorded as A_t . Meanwhile, the number of plate colonies in the control
142 group without silver ion implantation treatment was recorded as A_0 . The calculation
143 formula of antibacterial rate is as follows:

$$144 \quad \textit{Antibacterial rate (\%)} = (A_0 - A_t) / A_0 \times 100\%$$

145 2.6. Silver ion dissolution determination

146 After the silver ion implantation, each PE film was cut into samples with a size
147 of 1 cm × 1 cm, washed with 70% ethanol and dried with nitrogen. Then we placed
148 the material into 2 mL ultrapure water to soak it in the condition of 37°C for 24 h. In
149 the end, the water samples were collected and the content of silver ions was
150 determined by atomic absorption spectrophotometer.

151 2.7. Confocal scanning laser microscopy (CLSM)

152 *E. coli*, *S. aureus* and *P. aeruginosa* grown on PE films were rinsed and fixed
153 using the method of Friedlander et al. (Friedlander et al., 2013). Cells were
154 permeabilized with 0.1% Triton X-100 in PBS for 15 min. Samples were then stained
155 with Fluorescein isothiocyanate (FITC), 10 mg/mL for 30 min, and rinsed with PBS
156 twice. PE films were placed face-down on a glass-bottomed sample dish (Shengyou
157 Biotechnology Co., Ltd, Hangzhou, Zhejiang, China) and placed on the stage of the
158 confocal laser scanning microscope (CLSM 710, Zeiss, Oberkochen, Germany).
159 CLSM is equipped with an Ar laser at 488 nm. Images were obtained using the 63×oil
160 immersion lens and processed using Image J software (NIH, Bethesda, MD, USA).

161 Three different fields of view were randomly chosen for analysis of the percentage of
162 the images that show bacterial adhesion of the films.

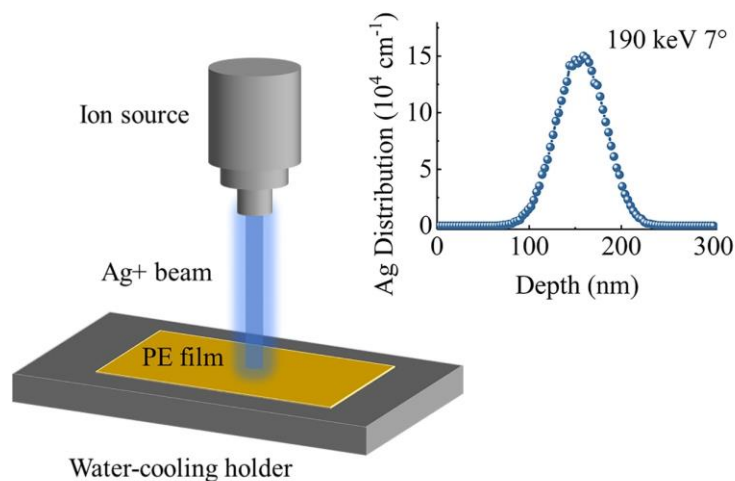
163 2.8. Statistical analysis

164 Statistical analysis was conducted using SPSS Statistics 20.0, where $p < 0.05$ was
165 used as the standard for significance. Origin 2018 and EXCEL 2016 were used to plot
166 and analyze the data.

167 3. Results and discussion

168 3.1. Characterization of surface properties of PE film after silver ion implantation

169 In the condition of 190 keV and an angle of 7° , the stretched PE film was
170 implanted with silver ions of different doses. According to the ion implantation
171 simulation with SRIM (Stopping and Range of Ions in Matter), it can be seen that the
172 silver element is mainly distributed at a depth of 75~250 nm, and the highest dose is
173 at a depth of 170 nm, as shown in the inset of Figure 1. This result illustrates that the
174 ion implantation only modifies the surface.

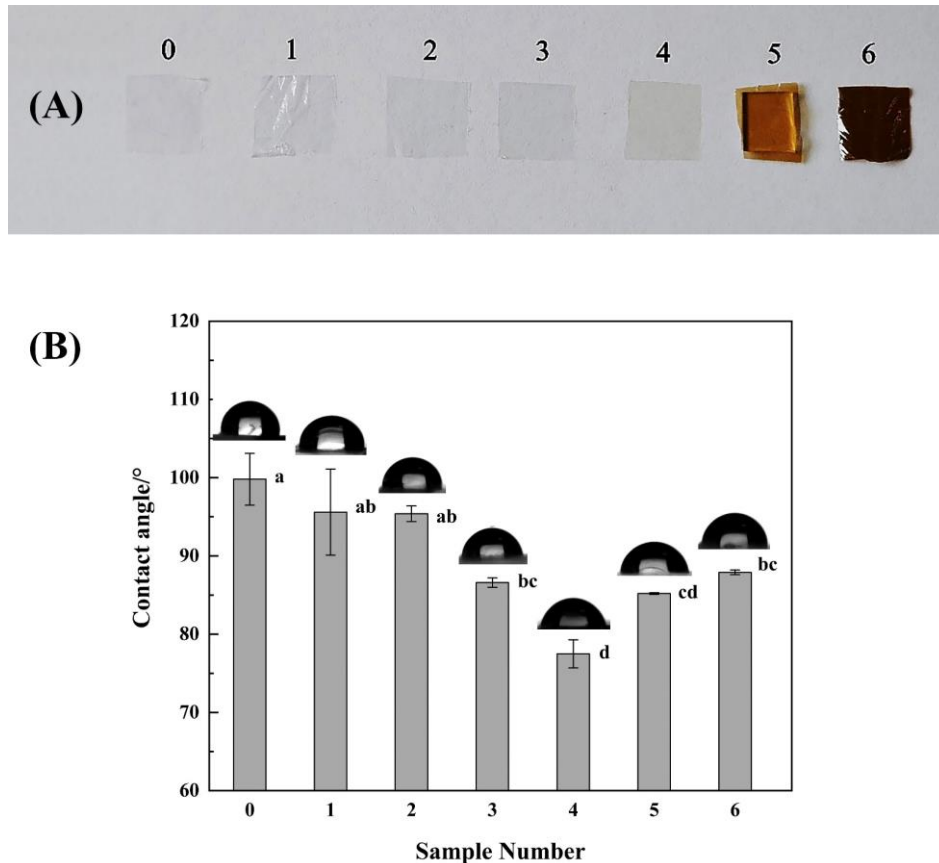


175

176 **Fig. 1.** The schematic of silver ion implantation on a PE film. Inset: Ag depth distribution on PE
177 films after implantation.

178 The appearance of the stretched film which has been ion-implanted is shown in
179 Figure 2(A). It can be found out that the color of the material was changed after the
180 silver ion implantation. Samples 1-3 is similar to the control group (sample 0), which
181 is still colorless and transparent. Sample 4 shows yellowish and sample 5 appears
182 brown, but they are still translucent. Sample 6 has almost no transparency, appearing
183 dark brown. In comparison with the control group, it can be found out that samples 5
184 and 6 become brittle and the toughness deteriorates largely. This phenomenon

185 indicates that different doses of silver ion implantation have different influence on the
186 surface of the PE film.

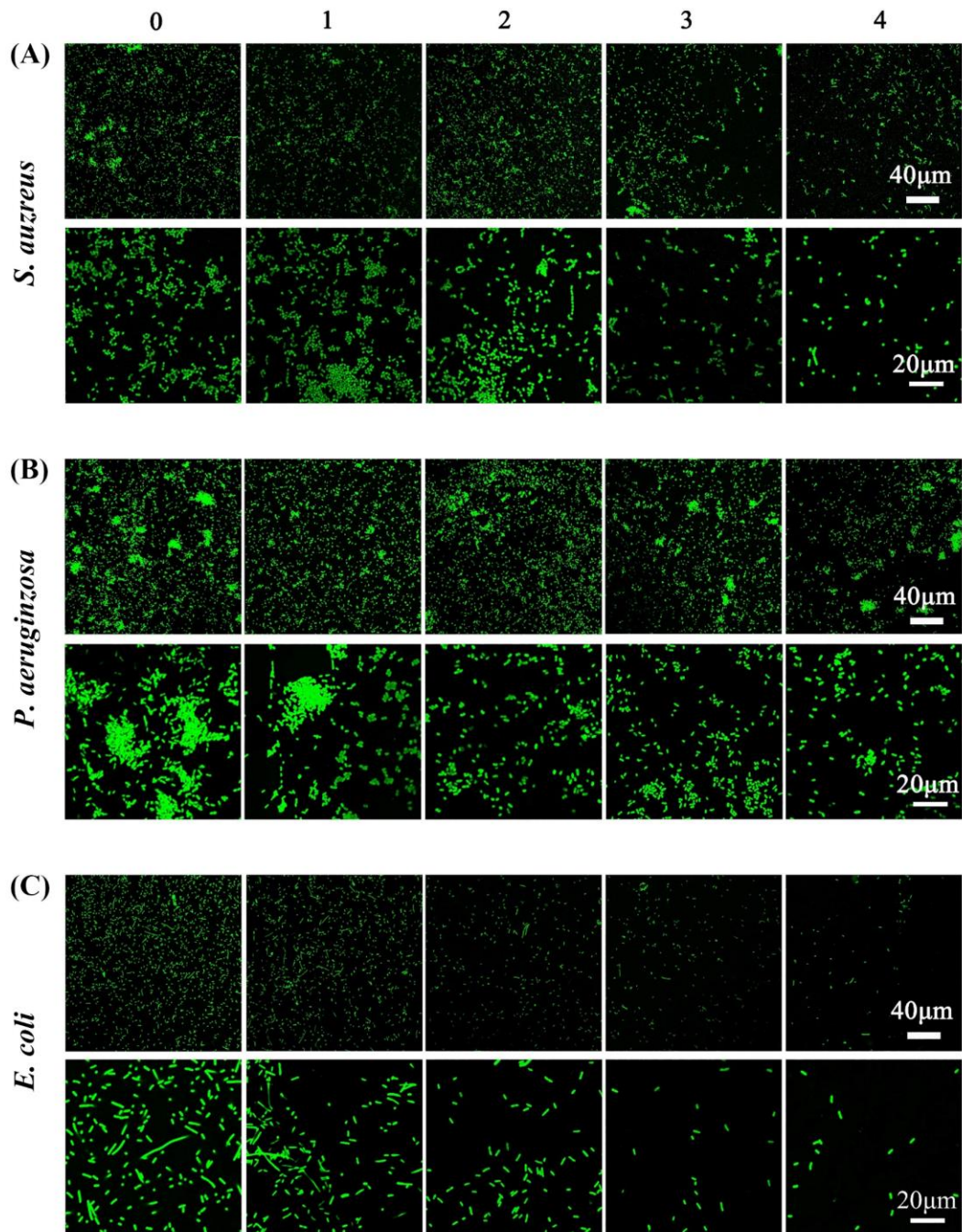


187
188 **Fig. 2.** The samples(A) and the contact angles(B) of PE films after ion implantation with different
189 doses. 0: None, 1: $5 \times 10^{12} \text{ cm}^{-2}$, 2: $1 \times 10^{13} \text{ cm}^{-2}$, 3: $5 \times 10^{13} \text{ cm}^{-2}$, 4: $1 \times 10^{14} \text{ cm}^{-2}$, 5: $5 \times 10^{14} \text{ cm}^{-2}$, 6:
190 $1 \times 10^{15} \text{ cm}^{-2}$, where $P < 0.05$.

191 The surface contact angle of samples 0 to 6 was measured in Figure 2(B).
192 Compared with the control group, the contact angle of the PE film after silver ion
193 implantation (sample 4) decreased by 24 from 100, indicating that silver ion
194 implantation has improved the surface hydrophilicity of the material. The main reason
195 is that silver ion implantation increases the surface roughness of the material. At the
196 same time, the addition of silver ion itself changes the interaction force between water
197 and the surface of the material. On the other hand, the contact angle increases for
198 samples 5 and 6, suggesting that the surface roughness decreases. It can be understood
199 as the excessive implantation doses damaged the internal molecular structure and the
200 PE films were melted and reconstituted. Therefore, the samples 5 and 6 are not
201 suitable for food packing materials or food contact materials because of their color

202 and change of structures. Based on these reasons, further measurements are only
203 performed for samples 0-4.

204 3.2. Antibacterial adhesion of films



205
206 **Fig. 3.** CLSM images of *S. aureus*(A), *P. aeruginosa*(B) and *E. coli*(C) adhesion on PE films after
207 ion implantation with different doses. 0: None, 1: $5 \times 10^{12} \text{ cm}^{-2}$, 2: $1 \times 10^{13} \text{ cm}^{-2}$, 3: $5 \times 10^{13} \text{ cm}^{-2}$, 4:
208 $1 \times 10^{14} \text{ cm}^{-2}$.

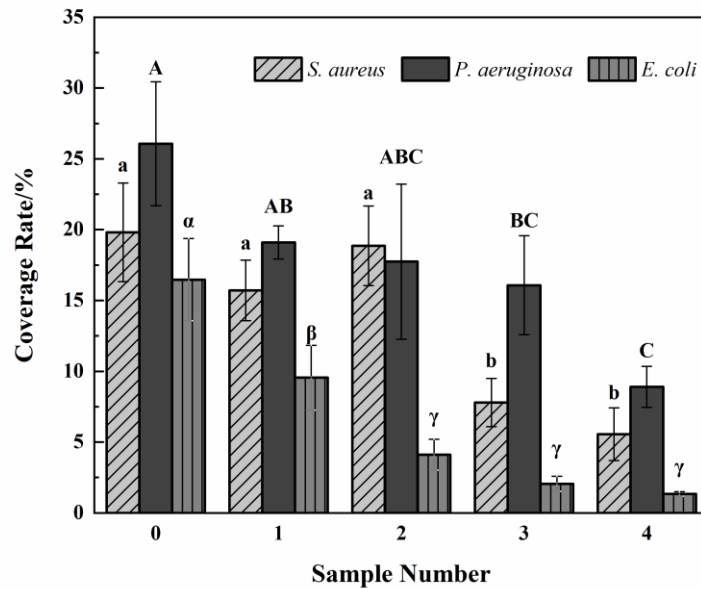


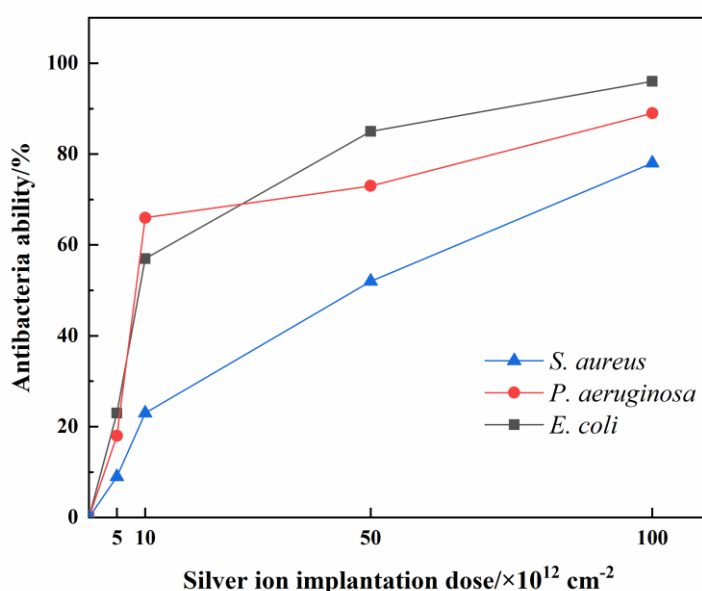
Fig. 4. The coverage rate of *S. aureus*, *P. aeruginosa* and *E. coli* counted base on the CLSM images. Histograms labeled with different letters (a-c, A-C, α-γ) represent significant differences.

Strong antibacterial adhesion can effectively hinder the formation of bacterial biofilm and slow down bacterial reproduction speed. The bacterial adhesion was identified by CLSM technology. The adhesion of *S. aureus*, *P. aeruginosa* and *E. coli* on the silver ion-implanted PE film is shown in Figure 3 and 4. For *S. aureus*, the adhesion of sample 2 was large, and samples 3 to 4 began to show a certain degree of inhibition. Sample 0-2 had a certain amount of bacterial cell agglomeration, which also indirectly illustrated that sample 3 and 4 have an inhibitory effect on the subsequent biofilm formation of *S. aureus*. For *P. aeruginosa*, samples 3 and 4 had inhibitory effects, and there were more single cell adhesion and no obvious bacterial aggregation, which indicated that the samples had a certain inhibitory effect on the subsequent biofilm formation. For *E. coli*, sample 2-4 all showed a good inhibitory effect. Comparing the adhesion of the three strains on the silver ion-implanted samples, it can be found out that, for different bacteria, the silver ion dose required to inhibit their adhesion is different. Among the three strains, the *E. coli* requires the lowest dose of silver ions, which is related to its poor adhesion ability (Lu et al., 2016); doses required against *S. aureus* and *P. aeruginosa* are similar.

The surface modification of PE film by silver ion implantation improves the ability of PE films to inhibit bacterial adhesion. According to the contact angle results, higher doses of silver ion implantation make a PE film more hydrophilic. And the

231 hydrophilic samples should have weak ability to inhibit bacterial adhesion (Lu et al.,
232 2016). Therefore, the effect of silver ions on bacterial adhesion is greater than that of
233 hydrophobicity improvement brought by ion implantation.

234 3.3. Antibacterial ability of the film



235

236

Fig. 5. Antibacterial ability of Ag ion-implanted PE films.

237 To investigate the antibacterial activity after the silver ions are implanted into the
238 PE films, the quantitative experiments were conducted by the film adhesion method.
239 As shown in Figure 5, after silver ion implantation, similar to the trend of the bacterial
240 adhesion results, the antibacterial activity of sample 1 is low. When the silver ion dose
241 increases, the antibacterial activity of the sample gradually increases. Different
242 bacteria have different tolerances to silver ions. At a dose of $1 \times 10^{13} \text{ cm}^{-2}$, more than
243 half of *P. aeruginosa* and *E. coli* can be killed. In contrast, the same activity can be
244 achieved at the dose of $5 \times 10^{13} \text{ cm}^{-2}$ for *S. aureus*. The activity difference comes from
245 the different composition and structure of the cell walls of the bacteria. The
246 peptidoglycan layer of the cell wall of gram-positive bacteria is thick and dense, with
247 phosphorus acid embedded, less or no lipid, lipopolysaccharide, lipoprotein.
248 Correspondingly, the cell wall of Gram-negative bacteria is thin and loose, and the
249 outer membrane is composed by phospholipids, lipopolysaccharides, and proteins
250 (Silhavy, Kahne, & Walker, 2010).

251 So far, Anh, D. H. et al. (Anh, Dumri, Anh, Punyodom, & Rachtanapun, 2016)
252 have found that the PE/AgNP nanocomposites restricted common pathogenic bacteria

253 (*E. coli*, *Bacillus subtilis*, *Salmonella typhimurium*) in their early developmental stage.
 254 Aalaie, J. et al. (Aalaie, Mirali, Motamedi, & Khanli, 2011) have found that PE
 255 films with as little as 1 wt% nanosilver provided absolute antibacterial performance,
 256 while generally maintaining the mechanical properties. Marchetti, F. et al.
 257 (Marchetti et al., 2015) have embedded new silver (I) acylpyrazolonato derivatives
 258 with mononuclear, polynuclear or ionic properties in a PE matrix and found that most
 259 of the composite materials have better antibacterial effects. The data in this study
 260 further shows capability of the relevant materials to break the bacterial cell membrane.
 261 Brito, S. D. et al (Brilo, Bresolin, Sivieri, & Ferreira, 2020) discovered that the
 262 packages incorporating silver nanoparticles inhibited the growth and reproduction of
 263 bacterial cells during the early stages. This is the same view as our study.

264 3.4. Determination of material safety

265 After immersing the samples in 2 mL ultrapure water for 24 h, the concentrations of
 266 silver ions in the solution were determined by atomic absorption spectroscopy. As
 267 shown in Table 1, the silver ion concentrations were all below the detection limit of
 268 the instrument, less than 0.01 mg/L. The silver ions after implantation are relatively
 269 stable on the surface of the material. This technique does not only guarantee the
 270 antibacterial activity of silver ions, but also greatly improves its safety. However,
 271 according to Commission Regulation (EU) No 10/2011 of 14 January 2011 on plastic
 272 materials and articles intended to come into contact with food, more research should
 273 be done before the material is applied directly to food and other fields.

274 **Table 1** Dissolution of silver ions from different labels in water.

Label	Implantation dose of silver ions /cm ⁻²	Dissolution
0	0	Not detected
1	5×10 ¹²	Not detected
2	1×10 ¹³	Not detected
3	5×10 ¹³	Not detected
4	1×10 ¹⁴	Not detected
5	5×10 ¹⁴	Not detected

275 4. Conclusions

276 We have explored the possibility of the Ag-implanted PE films as antibacterial
 277 food packing or food contact materials. When a dose is smaller than 1×10¹⁴ cm⁻²,
 278 silver ion implantation will not color the PE films but can improve their surface

279 hydrophilicity. Meanwhile, the PE films show the ability to inhibit bacterial adhesion
280 and have the bactericidal effect, both of which can be improved at higher doses. This
281 method is relatively safe, because the silver ions are stable and their dissolution
282 concentrations are all less than 0.01 mg/L. The results present the potential of ion
283 implantation in the food packing or food contact materials.

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