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Yang, Z, Li, C, Wang, T et al. (2023) Novel gellan gum-based probiotic film with enhanced biological activity and probiotic viability: Application for fresh-cut apples and potatoes. *International Journal of Biological Macromolecules*, 239. 124128. ISSN: 0141-8130

<https://doi.org/10.1016/j.ijbiomac.2023.124128>

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1 Novel probiotic film with enhanced biological activity and probiotic viability in 2 active packaging: application for fresh-cut apples and potatoes

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11 Abstract

12 A novel probiotic film based on gellan gum (GN), cranberry extract (CE), and *Lactococcus lactis* (LA)
13 was developed in this work. The SEM and fluorescence image results revealed that GN/CE film
14 containing LA was successfully fabricated. The addition of LA significantly improved the antibacterial
15 activity of the film. The presence of CE strengthens the antioxidant activity and LA survivability in the
16 film. The combination of LA (0~1.0%) and CE (0.5~1.0%) improved the mechanical property of the film
17 through the formation of density structure. The colored writing patterns were successfully printed on the
18 GN/LA/CE film. The best comprehensive properties were obtained with the film containing 2.0%LA and
19 0.5%CE. The GN/2.0%LA/0.5%CE film also showed the optimal preservation effect on fresh-cut
20 potatoes and apples. The GN/2.0%LA/0.5%CE film with colored patterns was expected to be a novel
21 probiotic film with improved biological property in active packaging.

22 **Keywords:** Cranberry extract; Green label; Gellan gum; *Lactococcus lactis*; Probiotic film;

23 Introduction

24 Nowadays, natural edible polymers packaging materials are a promising alternative to petroleum-based
25 ones due to the former being environmentally friendly and biodegradable. Various natural edible
26 polymers have been explored [1]. To broaden the application of edible film in food preservation, various
27 natural agents have been intentionally added to the edible film for developing active packaging materials
28 [2].

29 Recently, the incorporation of probiotics into the edible polymer to develop probiotic film has
30 attracted more attention in the active packaging field. Because probiotics as gut-friendly live

1 31 microorganisms that not only confer healthful effects on the host in adequate numbers but also reveal
2 32 high antimicrobial activities through the production of bioactive substances or competition for nutrition
3
4 33 [3]. Different probiotics with excellent antimicrobial activity have been used as effective antibacterial
5
6 34 agents [4-7]. Among them, *Lactococcus lactis* (LA), a popular probiotic, has been listed as a generally
7
8 35 safe (GRAS) food additive due to its excellent antibacterial activity and health benefits [8]. Some
9
10 36 research revealed that the addition of LA could strengthen the antimicrobial property of the film, such as
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12 37 PVOH film [9], and starch/carboxymethyl cellulose film [10]. Hence, the LA could be considered to add
13
14 38 into the edible film as a natural antimicrobial agent.

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17 39 However, the antioxidant property of the probiotic film was not adequate, which could limit its
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19 40 application on easily oxidized food [7, 11, 12]. Settler-Ramírez et al. [9] also pointed out that the LA did
20
21 41 not obviously affect the antioxidant property of the polyvinyl alcohol film because *L. lactis* produces
22
23 42 limited antioxidants in the film. In addition, a big concern of the probiotic film is the survivability of
24
25 43 probiotic embed in the film during storage period. Because the poor probiotic viability could significantly
26
27 44 decrease its biological activity and health benefits [7]. Fortunately, various studies have demonstrated
28
29 45 that the co-encapsulation of probiotic and bioactive compounds in a single matrix could increase the
30
31 46 probiotic viability [13, 14]. Some bioactive compounds have been explored to improve the viability of
32
33 47 probiotics, such as omega-3 oil, green tea extract, and resveratrol. Besides, the addition of bioactive
34
35 48 compounds positively affected the antioxidant property of the film due to their scavenging ability on the
36
37 49 free radicals [14, 15].

38
39 50 Cranberry extract (CE) was a natural bioactive compound-rich extract obtained from dried cranberry
40
41 51 (*Vaccinium macrocarpon* Ait.). CE contains a large amount of bioactive compounds, such as polyphenols,
42
43 52 flavonoids, anthocyanins, and VC [16]. It has been widely used as a natural antioxidant in food due to it
44
45 53 is safe and rich nutrition [17]. Previous studies also reported that the CE could improve the viability of
46
47 54 LAB-related probiotics [18, 19]. Thus, the BE could be considered a bioactive compounds-rich substance
48
49 55 to improve the probiotic viability and antioxidant activity of the film.

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51
52 56 In addition, to broaden the application scope of the anthocyanin-rich film, our previous work has explored
53
54 57 the possibility of printing different patterns on the nanocomposite film containing anthocyanin by using
55
56 58 electrochemical writing [20]. These developed films with printed letters or patterns could be used as a
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58 59 promising alternative to petroleum-based labels to provide some essential product information to
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60 60 consumers. However, there are only few works regarding the development of printing patterns on
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61 anthocyanin/probiotic composite films.
62 Accordingly, the objective of this study was to develop a novel probiotic film with improved biological
63 activity and probiotic viability. The gellan gum (GN), an important natural extracellular polysaccharide
64 obtained from *Sphingomonas paucimobilis* fermentation, was used as the edible film-forming matrix in
65 this work due to its low prices and good film-forming property. The effect of LA concentration and CE
66 concentration on the structure, biological and mechanical properties of the film, and viability of *L. lactis*
67 was explored. Furthermore, the colored writing patterns were printed on the probiotic film. Finally, the
68 preservation effect of the probiotic film on easily oxidized and perishable food, including fresh-cut apples
69 and potatoes, was also studied. This study would not only benefit the development of novel and effective
70 packaging systems but also reduce white pollution.

71 **2. Materials and methods**

72 **2.1. Materials**

73 Dried cranberry (cultivar “Yageda”) was obtained from Nanjing, China. Chemical agents, including
74 gelatin (CP, water content $\leq 12\%$, pH=5.0~7.0), methanol (GR, purity $>99.8\%$), Fluorescein Diacetate
75 (FDA) (purity $>99.99\%$), and Man, Rogosa, and Sharpe broth (MRS) (pH = 6.2 ± 0.2), and Tryptone Soy
76 Agar (TSA) medium (pH = 7.3 ± 0.2) were purchased from Shanghai Aladdin Biochemical Technology
77 Co., Ltd. (Shanghai, China). Probiotic *Lactococcus lactis* (CICC 21028), and pathogenic microbe
78 *Listeria monocytogenes* (ATCC 19114) were obtained from Shanghai yiyuan bio-technology Co., Ltd.
79 (Shanghai, China).

80 **2.2. Preparation of cranberry extract (CE)**

81 The extraction method of CE was measured following our previous method [21]. The dried cranberry
82 was ground into powder and soaked in an alcohol (80%)-HCl (40 mM) mixture. The mixture was reacted
83 in the dark for 1 h and centrifuged at 10000 rpm for 15 min. After that, the crude CE extraction was
84 filtered four times with filter paper (Whatman No. 1) and then freeze-dried to achieve the CE powder.
85 The obtained powder was stored at 4 ± 1 °C for further experiment.

86 **2.3. Preparation of *L. lactis* culture**

87 The *L. lactis* was cultured in MRS broth. The prepared *L. lactis* cultures were packed in Erlenmeyer flask
88 and incubated at 30 °C for 24 h. The *L. lactis* culture was centrifuged at 6000 rpm for 15 min and then
89 washed with sterile normal saline. Finally, the *L. lactis* suspension was centrifuged at 5000 rpm for 20
90 min to obtain the *L. lactis* pellet.

91 2.4. The fabrication of films

92 The pure GN solutions were prepared by dissolving 2 g of GN powder in 100 mL of distilled water at
93 98 °C, and expressed as GN/0%LA/0%CE. The GN solution enriched with LA was prepared by adding
94 different amount of LA pellet (1.0 and 2.0 mg/10 mL) into GN solution at 30 °C, and expressed as
95 GN/1.0%LA/0%CE and GN/2.0%LA/0%CE, respectively. The GN solution enriched with CE was
96 prepared by adding different amount (0.5 and 1.0 mg/mL) of CE into GN solution at 50 °C, and expressed
97 as GN/1.0%LA/0%CE and GN/2.0%LA/0%CE, respectively. The GN/LA/CE solution was prepared by
98 adding different amount of CE (0.5 and 1.0 mg/mL) into GN solution at 50 °C. Then, the different amount
99 of LA pellet (1.0 and 2.0 mg/ 10 mL) was added into the GN/CE solution when the solution was cooled
100 to 30 °C, and expressed as GN/1.0%LA/0.5%CE, GN/1.0%LA1.0/0%CE, GN/2.0%LA/0.5%CE and
101 GN/2.0%LA/1.0%CE, respectively. The film was prepared by casting the film-forming solution into a
102 petri dish and drying in an oven at 30 °C for 24 h.

103 2.5. Characterization of composite films

104 2.5.1 Antibacterial and antioxidant properties of the films

105 The inhibition of films against harmful microorganisms growth was determined according to the
106 previously reported method [22]. The *L. monocytogenes* was selected as the harmful microorganisms in
107 this work due to its importance in foodborne illness in China. The *L. monocytogenes* was cultured in
108 Tryptic Soy broth. The prepared *L. monocytogenes* cultures were packed in Erlenmeyer flask and
109 incubated at 37 °C for 24 h. Then, 100 mL of this *L. monocytogenes* culture ($\sim 10^8$ CFU/mL) was cultured
110 onto TSA plates at 37 °C for 24 h. The antibacterial property of the film was evaluated by determining
111 the diameter of the inhibition zone.

112 The DPPH scavenging ability of the film was measured to evaluate the antioxidant property in this work.
113 Briefly, 30 mg of the film sample was mixed with 5 mL of 0.1 mM DPPH solution (dissolved in ethanol)
114 and reacted for 45 min in the dark. The absorbance of the mixture was noted at 517 nm. DPPH scavenging
115 activity of the film was obtained based on Eq. (1):

$$116 \text{ DPPH scavenging activity (\%)} = [(A_0 - A_1)/A_0] \times 100 \quad (1)$$

117 Where A_1 and A_0 signify the absorbance of the DPPH solution mixed with or without film extract solution.

118 2.5.2. Mechanical and optical properties

119 The absorbance of the films was measured at 600 nm wavelengths using a spectrophotometer. The
120 opacity of film was evaluated based on the Eq. (2).

1 121 $\text{Opacity} = A_{600}/x$ (2)

2 122 Where A_{600} is the absorbance of the film at 600 nm, and x is film thickness.

3
4 123 The mechanical properties (Tensile strength (TS) and elongation percentage at break (EB)) of prepared

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6 124 films were determined according to the ASTM method (Standard D882-00, 2000) and calculated using

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8 125 Eq. (3) and (4):

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10 126 $TS = F/S$ (3)

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12 127 $EB(\%) = \Delta l/l_0$ (4)

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14 128 Where F is the maximum load; S is the initial cross-sectional area; Δl is film extension; l_0 is initial length.

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17 129 **2.5.3. Water vapor permeability (WVP)**

18 130 The film's WVP was measured referred to the procedure of Yang, Zhai, Zhang, Shi, Huang, Li, Zou,

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20 131 Gong, Holmes, Povey and Xiao [21]. Firstly, the fabricated film sample was selected to cover the

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22 132 centrifuge tube (25 mL) containing a certain amount of distilled water. After that, the centrifuge tube was

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24 133 placed in the dryer. The weight of the centrifuge tube was weighted every two hours. The following

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26 134 formula obtained the WVP:

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29 135 $WVP = (\Delta m \times \Delta P)/(A \times \Delta t \times x)$ (5)

30
31 136 Where Δm represents the weight change per unit time; Δp is the partial pressure difference between the

32
33 137 two sides of the film; A is the transfer area; Δt is per unit time; x is the thickness of film.

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35 138 **2.5.4. The fluorescence and visual appearance**

36 139 Films were stained with FDA (10 $\mu\text{g}/\text{mL}$) for 20 min at room temperature in the dark. The fluorescence

37
38 140 of the films at 480 nm excitation wavelength was noted with a confocal laser microscope (Leica TCS

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40 141 SP5). The visual appearance of the film was obtained with a scanner.

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43 142 **2.5.5. Morphological properties**

44 143 The surface of film morphology was characterized using scanning electron microscopy (JEOL, JSM-

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46 144 6360). All film samples were fixed on bronze stub using double-side adhesive and sputter-coated with

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48 145 gold in a vacuum evaporator.

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51 146 **2.6. Survivability of LA in the films**

52 147 The bioactive film incorporated with LA was stored at 25 ± 1 °C and 75 % RH for 24 d, and the

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54 148 survivability of the LA was determined every four days. In brief, the probiotic films were aseptically

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56 149 transferred into the sterile saline solution and agitated for 45 min. After that, the LA solution of an

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58 150 appropriate concentration was spread on an MRS medium. The counts of LA were measured using plate-

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151 count method. The relative survival rate of LA was obtained using Eq. (6) and (7):

152
$$\text{Survivability} = \log C_t - \log C_0 \quad (6)$$

153
$$\log C_t = \log C_0 - K \times t \quad (7)$$

154 Where C_0 is the number of initial viable *L. lactis* number, C_t is the number of viable *L. lactis* after a
155 specific time of storage (CFU/g), t is the storage time (d), k is the inactivation rate (log CFU/d).

156 **2.7. The procedure of printing patterns on the film**

157 The written patterns on the film was conducted by electrochemical writing according to our previous
158 method [20]. The GN hydrogels containing anthocyanin were selected in this work for electrochemical
159 writing. The anode and cathode of the electrochemical analyzer (CHI660E, CH Instruments Co.,
160 Shanghai, China) were connected with platinum (Pt) needle (0.2 mm) and Pt plate, respectively. The Pt
161 needle was contacted with the surface of the hydrogel that placed on the Pt plate and controlled by a
162 DOBOT M1 robotic arm (Yuejiang Technology Co., Ltd., Shenzhen, China) with a move precision of
163 0.1 μm .

164 **2.8. Biodegradable properties of films**

165 The biodegradability of films in soil was measured according to the method reported by Lu, Li, Ma, Li,
166 Jiang, Qin, Li, Li, Zhang and Wu [23]. Firstly, the square film samples (20 mm \times 20 mm) were prepared
167 and then placed in a natural environment in microorganisms-rich soil. The film figures were captured
168 every 3 days to observe the visually decrease in their area.

169 **2.9. Application for fresh-cut potatoes and apples**

170 Fresh apples (cultivar “Hongfushi”) and potatoes (cultivar “Huaen 1”) with good appearance quality
171 were obtained from local producers. The apples and potatoes were peeled and washed with distilled water.
172 After that, the apple were cut into 15 mm \times 30 mm pieces and the potatoes were cut into 20 mm \times 20
173 mm pieces. All the pieces were washed with distilled water and then air-dried for 1 h. Then pieces
174 samples were immersed in different film-forming solutions and air-dried again. The pieces only washed
175 with distilled water served as the control. Finally, all fresh-cut samples were placed in a polyethylene
176 tray and stored at 4 ± 1 $^{\circ}\text{C}$ and 75% RH for 6 days.

177 **2.10. Statistical analysis**

178 One-way analysis of variance (ANOVA) was used to assess the significantly difference between the
179 tested samples. All the presented results were the average of four measurements, and the mean values

180 marked with different letters indicate significant differences ($P < 0.05$).

181 **3. Results and discussions**

182 **3.1. SEM morphology and fluorescence figures of the film**

183 The SEM images of the surface of different films were presented in Fig. 1A. The surface of the pure GN
184 film was smooth and flat. For the film only containing CE, the surface of GN/0.5%CE film presented a
185 smooth and uniform morphology, indicating that CE has great compatibility with GN films. The
186 incorporation of 1.0% CE make the surface of GN film became uneven. Similar rough structure was also
187 observed in chitosan/jujube leaf extract [24] film and gelatin/corn starch/corn stigma extract film [15].
188 This was attributed to the fact that the increased CE concentration makes the insoluble particles not well
189 dispersed in the GN matrix [25]. Notably, the addition of LA obviously changed the morphology of the
190 surface of GN film, some LA cells were observed on the surface of the composite film. Li et al. [26] also
191 observed that the edible film surface became rough with the addition of probiotics. Similar result was
192 also observed in the GN-based film containing 2.0%LA. Notably, with the incorporation of CE, the
193 surface of the GN/LA film became rough and dense, and it became rougher with the increasing
194 concentration of CE. This result indicated the cross-linking between GN and LA was strengthened by the
195 addition of CE. However, the surface of GN/2.0%LA/1.0%CE film became very rough due to the large
196 accumulation of LA cells or excessive crosslinking between the different polymer chains. All GN/LA
197 films presented a rough surface and cross-section morphology. Fortunately, no crack or phase separation
198 was observed in all GN/LA/CE composite films. To check the presence and distribution of LA in the
199 film, the fluorescence of the films was also measured (Fig. 1B). No fluorescence was observed in the
200 figures of GN films without LA. The green fluorescence was evenly presented in GN/LA and GN/LA/CE
201 films, indicating the active and good distribution of LA in the matrixes. For the film containing the same
202 CE concentration, the number of green fluorescent dots in the 2.0% LA-loaded film was higher than that
203 of 1.0%LA-loaded film. Notably, the distribution of green fluorescent in the GN/1.0%LA/1.0%CE and
204 GN/2.0%LA/1.0%CE film became non-uniform, indicating the aggregation of the LA. This result was
205 inconsistent with the SEM result. The high concentrations of CE (1.0%) may cause the aggregation of
206 probiotics. This probiotic aggregation could be due to the increased roughness and unevenness of the
207 film making probiotic cells accumulate in the depressions on the film's surface. Singh et al. [27] also
208 found that *Lactobacillus rhamnosus* accumulate at the structural defects in cellulose-based films. These
209 results indicated that the GN-based probiotic film containing LA/CE was developed successfully in this

210 work. The incorporation of 1.0%CE/2.0%LA decreased the uniformity structure of the distribution of
211 probiotics in the film.

Fig. 1

212

213 3.2. The antibacterial and antioxidant properties

214 The antibacterial property of the films were presented in Fig. 2A. The pure GN has almost no inhibition
215 effect on *L. monocytogenes*. The maximum antibacterial properties (14.9 ± 0.6 mm) were obtained with
216 GN films containing 2.0%LA and 0.5%CE. For the film only incorporated with CE or LA, the
217 antibacterial activity of the composite film significantly increased with the increased LA or CE
218 concentration ($P < 0.05$). This result indicated that the incorporation of LA or CE possessed a positive
219 efficacy on the antibacterial property of the GN film. For the film both enriched with CE and LA, the
220 combination of 1.0%LA or 2.0%LA with 0.5%CE significantly enhanced the antibacterial property of
221 the film. Similar synergistic effect was also observed by Riešutė et al. [28], who pointed out that the
222 combination of plant extract and lactic acid bacteria could improve the antibacterial property of the
223 nutritional bar. The obviously increased antibacterial property in the CE/LA-loaded film was due to the
224 presence of LA in GN matrix may produce nisin and bacteriocin to break the cell structure of pathogens
225 [8]. In addition, the synergistic effect between the bioactive compounds in CE and LA could furtherly
226 strengthen the inhibition effect [29]. However, when the CE concentration was up to 1.0%, the
227 antibacterial property of the film containing CE and LA obviously decreased ($P < 0.05$). This may be
228 ascribed to that the excessive CE content changed the pH condition in the film so that inhibit the growth
229 of probiotics and reduce the production of antibacterial substances, resulting in a decreased antibacterial
230 property of the film [30]. Khalaphallah and Soliman [31] also reported that the positive effect of plant
231 extract on probiotic viability depended on the concentration of plant extract. This result indicated that
232 the incorporation of CE or LA favored the antibacterial property of the GN film, and the incorporation
233 of 2.0% LA and 0.5% CE obtain the best antibacterial property.

234 The antioxidant property of the films were presented in Fig. 2B. The antioxidant property of
235 GN/0%LA/0%CE film was $2.4 \pm 0.2\%$, and it increased to $10.6 \pm 0.7\%$ by the incorporation of 1.0%LA.
236 Notably, the incorporation of CE significantly affected the antioxidant property of the film ($P < 0.05$).
237 The satisfactory antioxidant property was obtained when the incorporation CE concentration was 1.0%.
238 This improved antioxidant property of the film could be due to the presence of flavonoids, Vitamin C,

1 239 and polyphenols in CE possessed excellent antioxidant capability [32]. It is interesting to observe that
2 240 the antioxidant property of the film containing CE and LA seems to be only sensitive to CE concentration.
3
4 241 For the composite film containing 0.5%CE or 1.0%CE, the addition of LA did not obviously affect the
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6 242 antioxidant property of the film ($P > 0.05$). Contrarily, Li et al. [26] pointed out the incorporated probiotic
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8 243 could improve the antioxidant activity of the film because some LAB strains could produce some
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10 244 exopolysaccharides (EPS). However, in this work, the antioxidant property of the LA-loaded film hardly
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12 245 altered. Settler-Ramírez et al. [9] also observed the antioxidant property of the PVOH film did not change
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14 246 with the addition of LA because *L. lactis* produces limited antioxidants in the film.
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18 247  Fig. 2
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20 248 3.3. Optical property and visual appearance

21 249 The optical property is an essential parameter for food packaging material. Hence, the opacity and visual
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23 250 appearance of the films were studied in this work. The opacity of the GN/0%LA/0%CE film was 0.85
24
25 251 A/mm (Fig. 2C), and it significantly increased with the increasing incorporation of CE or LA ($P < 0.05$).
26
27 252 The result indicated that the incorporation of CE or LA could reduce the film's transmittance. For each
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29 253 GN-based film containing 1.0%LA or 2.0%LA, the incorporation of 0.5%CE did not affect the opacity
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31 254 of the composite film. Our previous studies also observed that the addition of low concentration red
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33 255 radish anthocyanin did not affect the transmittance of the gelatin/gellan gum based film [33]. The highest
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35 256 opacity value was observed in the GN/2.0%LA/1.0%CE composite film (3.0 A/mm). This was due to the
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37 257 presence of a large amount of colored group of LA in the film could block the light so that decrease the
38
39 258 transmittance of the film.
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41
42 259 The visual appearance of GN/0%LA/0%CE film presented bright white color (Fig. 2D). The
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44 260 incorporation of CE makes the pure GN film become light pink, and the color of the film became
45
46 261 deepener and dark with the increasing CE concentration. The addition of LA increased the lightness of
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48 262 the GN film, and GN-based films only incorporated with LA became yellow. The color of probiotics film
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50 263 was mainly related to the density of probiotics [26]. The deepen color in GN/LA films was due to the
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52 264 presence of a higher LA concentration in the GN matrix block the passage of light. For each film, the
53
54 265 addition of CE making the color of the film became light pink, and the color of the film further became
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56 266 pink with the increasing CE concentration. Similar finding was also observed in starch/polyvinyl
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58 267 alcohol/roselle extract film [34]. The opacity and color depth of the film increased with the increasing
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1 268 CE or LA content. Even though the addition of 2.0% LA and 1.0%CE could enhance the biological
2 269 activity of the film (Fig. 2A and 2B), however, it presented a negative effect on the opacity of the film.

3 270 **3.4. TS and EB of the film**

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5
6 271 The mechanical property of the packaging material is a critical parameter to meet the external stress
7
8 272 during the processing and transport process. As can be despite in Table 1, the TS of the pure GN film
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10 273 was 17.92 ± 0.8 MPa, and it obviously decreased with the incorporating LA ($P < 0.05$). The lowest TS
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12 274 value (14.15 ± 1.2 MPa) was obtained with GN films containing 2.0% of LA. Similar result was also
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14 275 observed in the GN/LA/0.5%CE or GN/LA/1.0%CE films. Actually, the tensile strength of the composite
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16 276 films is related to the distribution and density of the incorporation polymer moleculars and their
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18 277 interactions [26]. The increased LA cells could aggregate in the matrix to decrease the compactness of
19
20 278 the film (Fig. 1). Similar decreased TS was also found in the cassava starch/carboxymethylcellulose film
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22 279 incorporated with 2% lactic acid bacteria [26]. The TS value of the film significantly strengthened with
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24 280 the addition of CE, and it furtherly enhanced with the increasing concentration of CE. The highest TS
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26 281 (32.52 ± 1.2 MPa) was obtained with the film containing 1.0% LA and 1.0% CE. This satisfactory TS of
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28 282 the composite film was in accord with the fact that the presence of phenolic compounds in CE enhanced
29
30 283 cross-linking and density in the polymer matrix (Fig. 1A) so that increase the TS of the film. The EB of
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32 284 the developed films was shown in Table 1. Generally, an enhanced TS results in a decrease in EB of the
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34 285 film. However, the EB of the composite film increased with the incorporation of 0.5%CE. This was
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36 286 because the addition of CE may form a layered structure so that increased the flexibility of the film. Zhai
37
38 287 et al. [33] also observed that the increasing anthocyanin concentration increases the TS and EB of the
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40 288 gelatin/gellan Gum film. For the pure GN film or GN film containing CE, the incorporation of 1.0%LA
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42 289 did not affect the EB of the films. Similar finding was also studied by Settier-Ramírez et al. [9]. However,
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44 290 the incorporation of 2.0% LA significantly decreased the EB of the film. The lowest EB ($9.15 \pm 0.7\%$)
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46 291 was observed in the GN/2.0%LA/0%CE film. This decreased EB of the composite film was due to that
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48 292 the agglomeration of a large number of LA negatively affected the uniformity of the film (Fig. 1C) so
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50 293 that decrease the EB [10]. The satisfactory EB value was obtained when the film containing high-level
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52 294 value of CE concentration (0.5%-1.0%) and the low-level value of LA (0%-1.0%). Several works also
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54 295 indicated that the addition of high anthocyanin extract could improve the mechanical property of the film
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56 296 [10, 35, 36]. This result revealed that the combination of CE (0.5~1.0%) and LA (0~1.0%) could enhance
57
58 297 the mechanical property of the GN film.
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298 3.5. Water barrier properties

299 The water barrier property of the film is essential for controlling the food quality during the storage and
300 transport process. Hence, the WVP of the films were checked in this work and the results were shown in
301 Table 1. For pure GN film, the incorporation of LA (1.0%~2.0%) or 0.5%CE did not significantly affect
302 the WVP of the GN film. The WVP of the pure GN film obviously decreased to $4.27 \pm 0.1 \text{ g}\cdot\text{mm}\cdot\text{m}^{-2}\cdot\text{Kpa}^{-1}\cdot\text{h}^{-1}$
303 when the concentration of incorporated CE was 1.0% ($P < 0.05$). This was because the
304 increased density of the GN/1.0%CE/0%LA film's structure (Fig. 1A) could effectively reduce the
305 passage of moisture. The WVP of the polymer film also related to the integrity of the matrix, hydrophilic,
306 and the interactions between each component [37]. For each film containing 2% LA, the WVP of the
307 film increased from 4.70 ± 0.1 to $4.81 \pm 0.2 \text{ g}\cdot\text{mm}\cdot\text{m}^{-2}\cdot\text{Kpa}^{-1}\cdot\text{h}^{-1}$ when the CE concentration increased
308 from 0.0% to 1.0%. The high concentration of LA cells and CE could aggregate in GN so that forming
309 rough and uneven structure (Fig. 1), resulting in an increased WVP of the film. Similar increased WVP
310 was also observed in the gelatin/gellan gum/radish extract composite film [33].

Table. 1

312 3.6. Survivability of *L. lactis* in the film

313 3.6.1. *L. lactis* survivability during the drying process

314 Fig. 3A presented the survivability of *L. lactis* cells in the different films during the drying process. The
315 viable cell numbers in all films decreased after drying at 30 °C for 24 h. This decreased viable cell
316 numbers is in accord with the fact that the increased osmotic pressures during the drying process broke
317 the cell structure of the microorganism [14]. Similar decreased viable probiotics numbers during the
318 drying process was also studied in starch/carboxymethylcellulose edible film [26]. Generally, the
319 suggested minimum doses in probiotic product was 10^6 CFU/g [38]. After the drying process, all
320 developed films exhibited satisfactory viable cell numbers ($> 6.0 \text{ log CFU/g}$). Regarding the
321 GN/1.0%LA-based composite film containing different CE concentrations, the CE concentration did not
322 affect the *L. lactis* viability in the GN film during the drying process. The similar finding was also studied
323 in the GN/2.0%LA-based composite films. The concentration of *L. lactis* also did not affect the
324 survivability of probiotics embedded in the film during the drying process, the viable cell numbers of all
325 developed film containing LA decreased by $\sim 1.0 \text{ Log CFU/g}$ after drying process. The probiotic
326 survivability in the film during the drying process is related to water evaporation rate, drying temperature,

1 327 moisture content, and film-forming matrix types [39]. In brief, the LA concentration or CE concentration
2 328 did not affect the probiotic survivability in the gellan gum film during the drying process in this work.

3 329 3.6.2. *L. lactis* survivability during the storage process

4 330 Probiotic viability during the storage process is an essential parameter for its health benefits and
5
6 331 biological activity [38]. The survivability of the *L. lactis* in all films exhibited a decreasing trend thought
7
8 332 the whole storage period (Fig. 3B). This reduction of viable cell numbers in all films was attributed to
9
10 333 the increased osmotic pressures and decreased nutrients during the storage process [40]. In the initial 4
11
12 334 d, no significant difference was observed in the survivability of *L. lactis* in all films. After 24 d of storage,
13
14 335 for each film containing the same concentration of LA, the addition of 0.5%CE significantly improved
15
16 336 the survivability of the probiotics. This improved survivability of *L. lactis* was due to the demonstrated
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18 337 prebiotic ability of CE could promote the growth of LAB [41]. Carine Raddatz et al. [29] also reported
19
20 338 that the presence of bioactive compounds, such as anthocyanin, phenolic, in the red onion residue extract
21
22 339 could stimulate the growth of probiotics. Notably, for each film containing the same concentration of LA,
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24 340 the *L. lactis* survivability in the film decreased by the incorporation of 1.0% CE. Zhu et al. [42] also
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26 341 pointed out that the promotion effect of the anthocyanin on the probiotic viability first increased and then
27
28 342 decreased with the increasing anthocyanin concentration. A high concentration of anthocyanin may
29
30 343 change the pH condition in the film so that inhibit the probiotic growth. In addition, the addition of 1.0%
31
32 344 CE causes the aggregation of probiotics in the film (Fig. 1B), which may intensify the space competition
33
34 345 of probiotics. Notably, the addition of a certain amount (1%~2.0%) of LA did not affect the *L. lactis*
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36 346 viability in the film. The inactivation rate (K) of *L. lactis* in all prepared films was also presented in Fig.
37
38 347 3C. The inactivation of all probiotics film was conformed to the first-order kinetic formula. As we
39
40 348 hypothesized, high concentrations of CE could reduce the viability of *L. lactis*. The GN/2.0%LA/1.0%CE
41
42 349 film obtained the highest inactivation rate (0.2057) while that in GN/1.0%LA/0.5%CE film was 0.1819.
43
44 350 This was because excessive concentration of quercetin in the CE may inhibit the growth of probiotics. C.
45
46 351 Iyer and K. Kailasapathy [43] also observed that the quercetin decreased the growth of *Lactobacillus*
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48 352 *acidophilus*. In addition, the content and types of the compounds in the plant extract also affect its
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50 353 prebiotic activity [44]. This result revealed that the *L. lactis* survivability in the GN film depends on the
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52 354 CE concentration, and the incorporation of CE (0.5%) could promote the survivability of *L. lactis* in the
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54 355 GN-based film.
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Fig. 3

1 356

2 357 **3.7. Electrochemical writing on composite film**

3 358 To provide essential manufacturing information to consumers, the colorful patterns were printed on the
4
5 359 anthocyanin-rich probiotic films (Fig. 4A). The images of the hourglass with two colors (pink and yellow)
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7 360 were printed successfully on the GN based probiotic films containing 0.5% CE or 1.0% CE. This
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9 361 development of multicolor patterns on the anthocyanin-rich probiotic film was based on structure
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11 362 changes of anthocyanin molecular under different pH conditions [20]. The principle of the fabrication of
12
13 363 the pink part of the hourglass pattern was due to that the structure of anhydrobase converted to flavylium
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15 364 ion under the low pH condition generated by positive electrolyte water reaction ($2\text{H}_2\text{O} - 4\text{e}^- = 4\text{H}^+ + \text{O}^2$)
16
17 365 (Fig. 4B). The principle of the fabrication of the yellow part of the hourglass pattern was due to that the
18
19 366 structure of anhydrobase converted to anhydrobase anion under the high pH condition generated by
20
21 367 negative electrolyte water reaction ($4\text{H}_2\text{O} + 4\text{e}^- = 4\text{OH}^- + 2\text{H}_2$). This structure change of anhydrobase
22
23 368 makes cranberry anthocyanin present different colors. Notably, the color of the hourglass patterns on the
24
25 369 composite film containing 1.0% CE is darker than that of the film containing 0.5% CE. This was because
26
27 370 a large number of anhydrobase anion or flavylium aggregates cause the deepening of the pattern color.
28
29 371 The addition of different concentration of LA did not affect the electrochemical writing performance of
30
31 372 the probiotic film. These colorful patterns could be used as a promising alternative to petroleum-based
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33 373 labels to provide some important product information to consumers, such as production date, shelf life.
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35 374 Furthermore, this patterned film could also broaden the application range of the probiotic film
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Fig. 4

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42 376 **3.8. Degradability of films in soil**

43 377 The photographs for checking the degradability of the composite film in soil was shown in Fig. 5. All
44
45 378 the GN-based film could be degraded in soil within 24 days. As a hydrophilic material, the pure GN film
46
47 379 was completely degraded within 15 days. This was because GN as a hydrophilic material could rapidly
48
49 380 absorb water from the environment, so that accelerate the growth of degradation-related microorganisms
50
51 381 [26]. The process of the film degradation in soil was slowed down with the incorporation of LA and CE,
52
53 382 and the GN/2.0%CE/1.0%LA film has the longest degradation time in soil. Similar increased degradation
54
55 383 time of the gelatin/corn stigma extract film was also studied by Boeira, Flores, Alves, Moura, Melo,
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57 384 Rolim, Nogueira-Librelo and Rosa [15]. This due to the presence of antibacterial and antioxidant
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1 385 agents in the film could inhibit the microbial activity in the soil. In addition, the density, water content
2 386 of the film, the microorganism types, and weather conditions also affected the biodegradability time of
3
4 387 the film [26]. The increased density structure by the addition of CE and LA, and the increased moisture
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6 388 content and loss of anthocyanin due to the rain erosion at 9th and 18th days may also affect the
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8 389 degradability of the GN-based composite film. This result indicated GN-based film presented the
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10 390 satisfactory biodegradability in the natural environment, and the incorporation of LA and CE may
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12 391 prolong the degradation time of the film in soil.
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16 392  Fig. 5

17 393 **3.9. Application of the film on fresh-cut potatoes and apples**

18 394 To evaluate the preservation effect of developed films on susceptible to bacterial infection and oxidized
19
20 395 food, the fresh-cut apple and potatoes was selected in this work. The appearance quality of the fresh-cut
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22 396 potatoes and apples were shown in Fig. 6A and 6B. At the beginning of storage, all fresh-cut fruits are
23
24 397 full and without damage, and the surface of all the fresh-cut apples and potatoes are bright color and
25
26 398 healthy. After storage for 6 d, the obviously surface browning and severe shrinkage due to water loss was
27
28 399 observed in the untreated fresh-cut potatoes and apples. The pure GN treatment presented a little effect
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30 400 on improving the quality of the fresh-cut samples. Notably, the visible browning degree in the GN/CE/LA
31
32 401 treated fresh-cut potatoes and apples was obviously lower over that of untreated samples, and the
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34 402 GN/2.0%LA/0.5%CE treated samples exhibited the optimal effect. This valid inhibition effect could be
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36 403 in accord with the fact that the presence of CE and LA in the GN/LA/CE films with excellent antioxidant
37
38 404 and antibacterial properties (Fig. 2A and 2B) could effectively inhibit the growth of pathogen and
39
40 405 scavenge free radical (Fig. 6C), so that decrease the browning degree and maintain good appearance
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42 406 quality of the fresh-cut apples or potatoes. In addition, the improved barrier properties also positively
43
44 407 control the water vapor exchange so that slow down the process of decay. This result proved that the GN-
45
46 408 based film incorporated with 0.5%CE and 2.0%LA expected to be an effective active packaging material
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48 409 for fresh-cut apples and potatoes.
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54 410  Fig. 6

55 411 **4. Conclusion**

56
57 412 A novel probiotic film with improved biological and probiotic viability based on GN, LA, and CE was
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59 413 fabricated in this work. The SEM and fluorescence image results revealed that the GN-based film
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1 414 containing LA/CE was prepared successfully. The incorporation of 1.0%CE and 2.0%LA positively
2 415 affect the antioxidant and antibacterial properties of the film while it had a negative effect on the film's
3
4 416 optical property and biodegradability. The addition of 0.5%CE significantly improved the LA
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6 417 survivability in the GN film during the storage period. Furthermore, the probiotic film with colorful
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8 418 patterns was successfully developed in this work, and the color depth of the patterns was sensitive to the
9
10 419 CE concentration. The best comprehensive properties were obtained with the film containing 2.0%LA
11
12 420 and 0.5%CE. The GN/2.0%LA/0.5%CE film also obtained the best preservation effect on fresh-cut
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14 421 apples and potatoes. Hence, GN/2.0%LA/0.5%CE probiotic film with green labels is expected to be a
15
16 422 novel active packaging material for food preservation.

18 423 **Acknowledgments**

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21 424 This study was supported by the National Natural Science Foundation of China (3210161415); Natural
22
23 425 Science Foundation of Jiangsu Province (BK202001030, BK20180865); Jiangsu Association for Science
24
25 426 and Technology youth Talent Promotion Project. Postgraduate Research & Practice Innovation Program
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27 427 of Jiangsu Province (KYCX21_3395);

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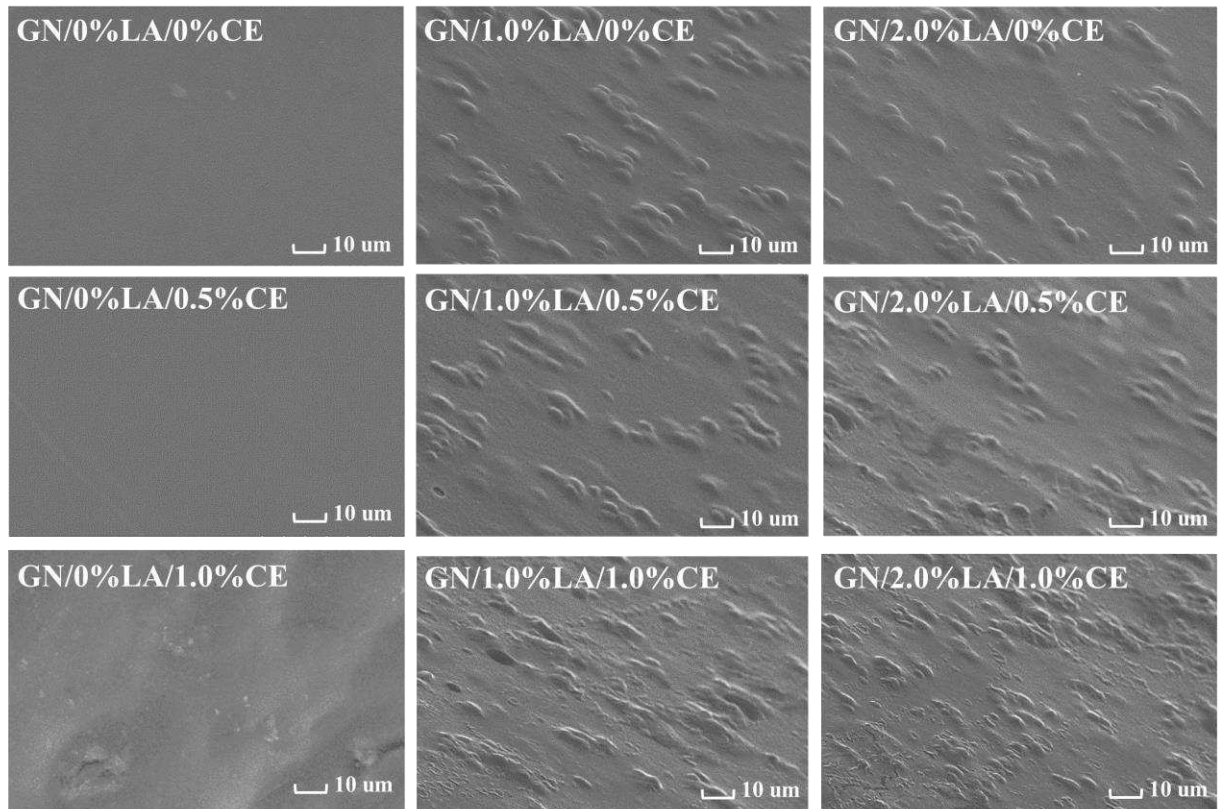
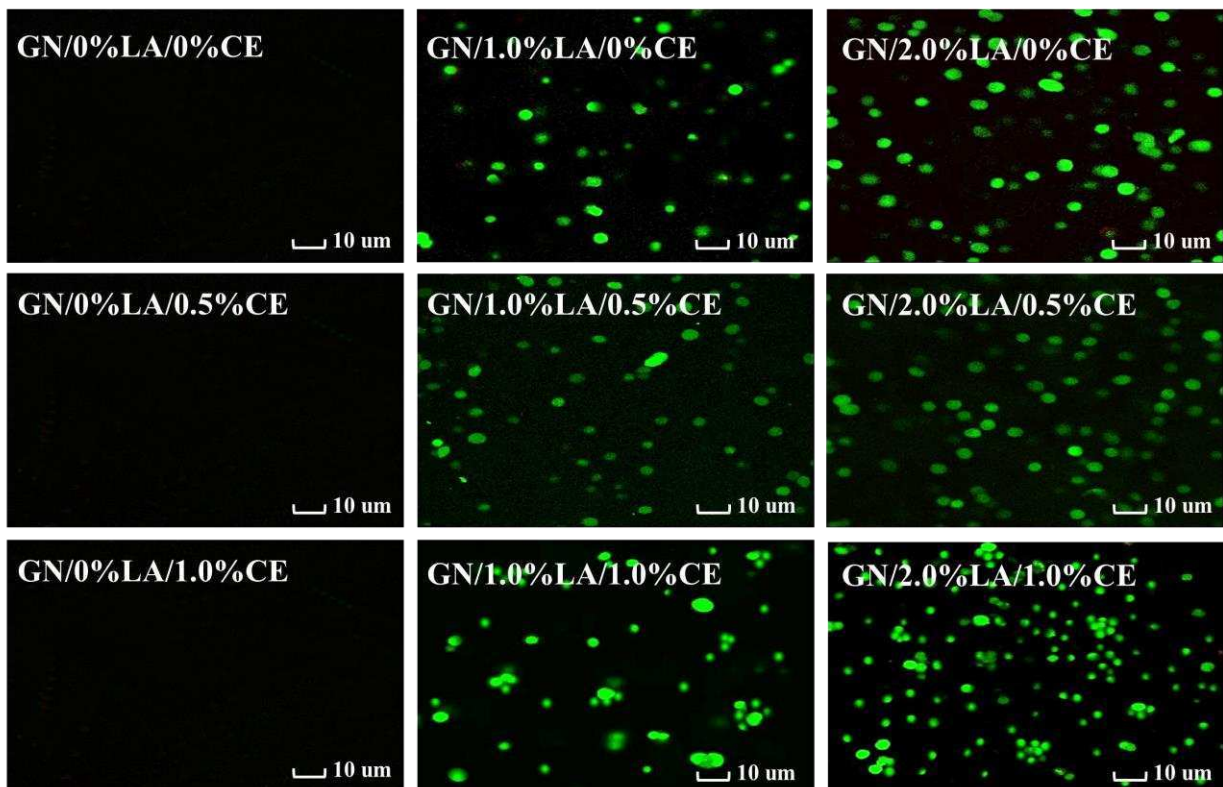
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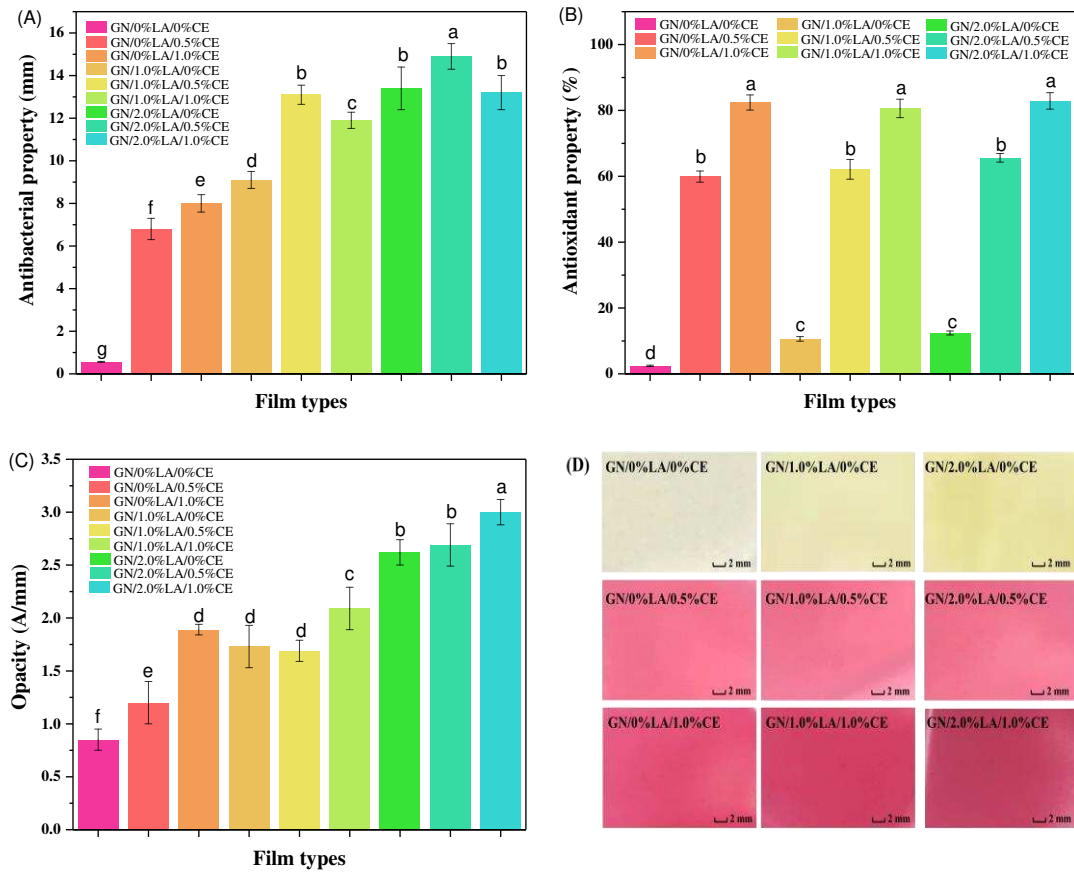
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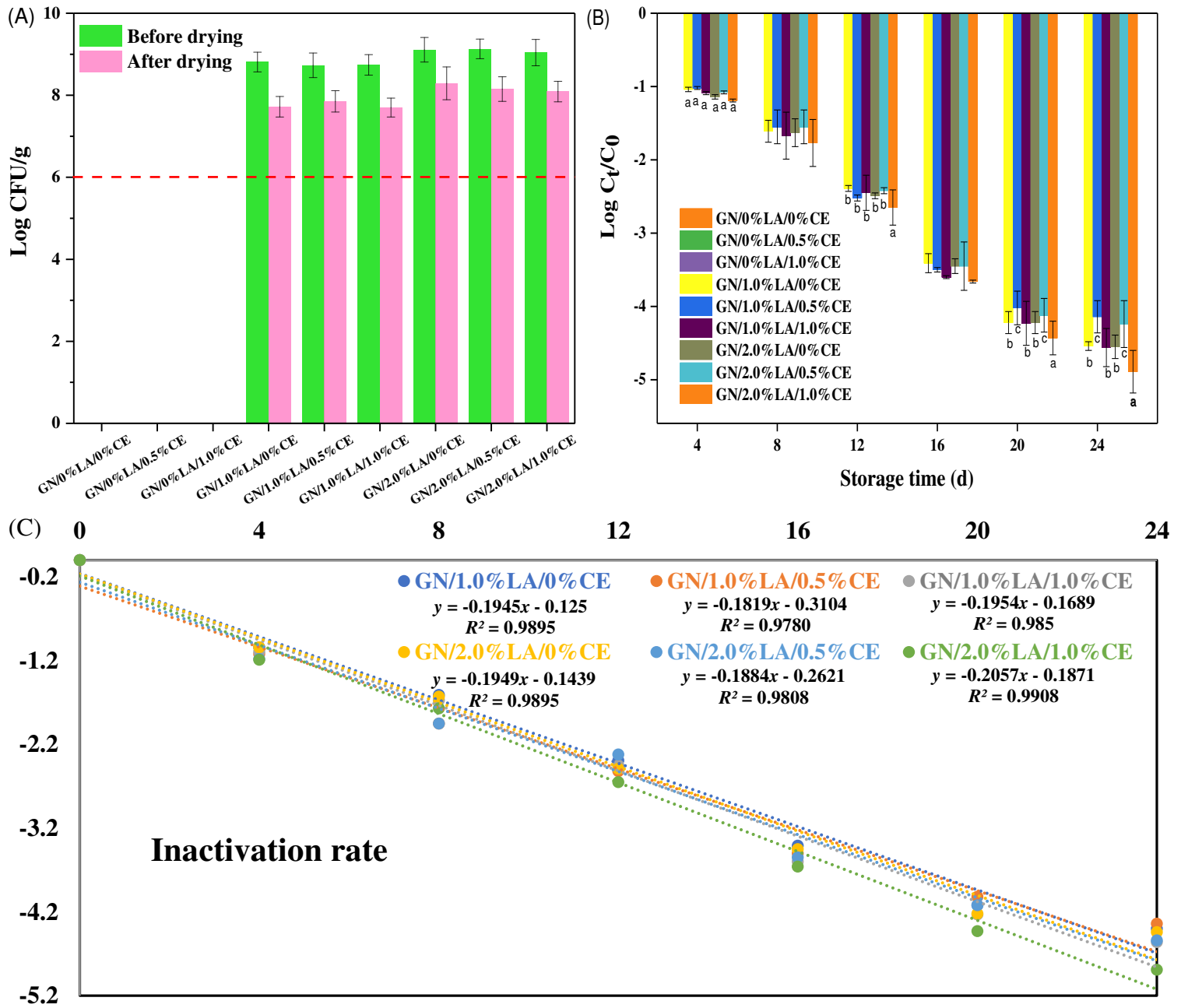
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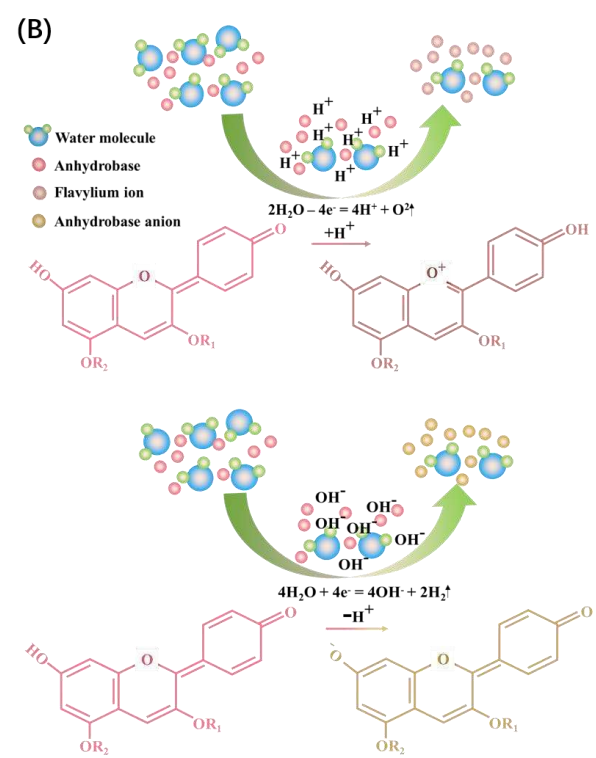
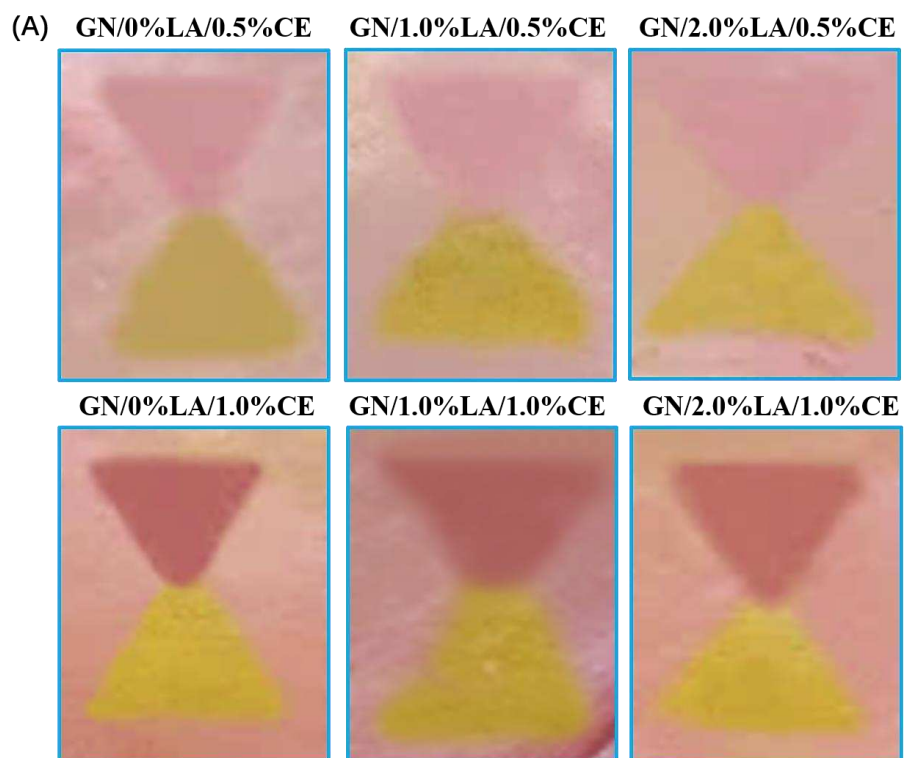
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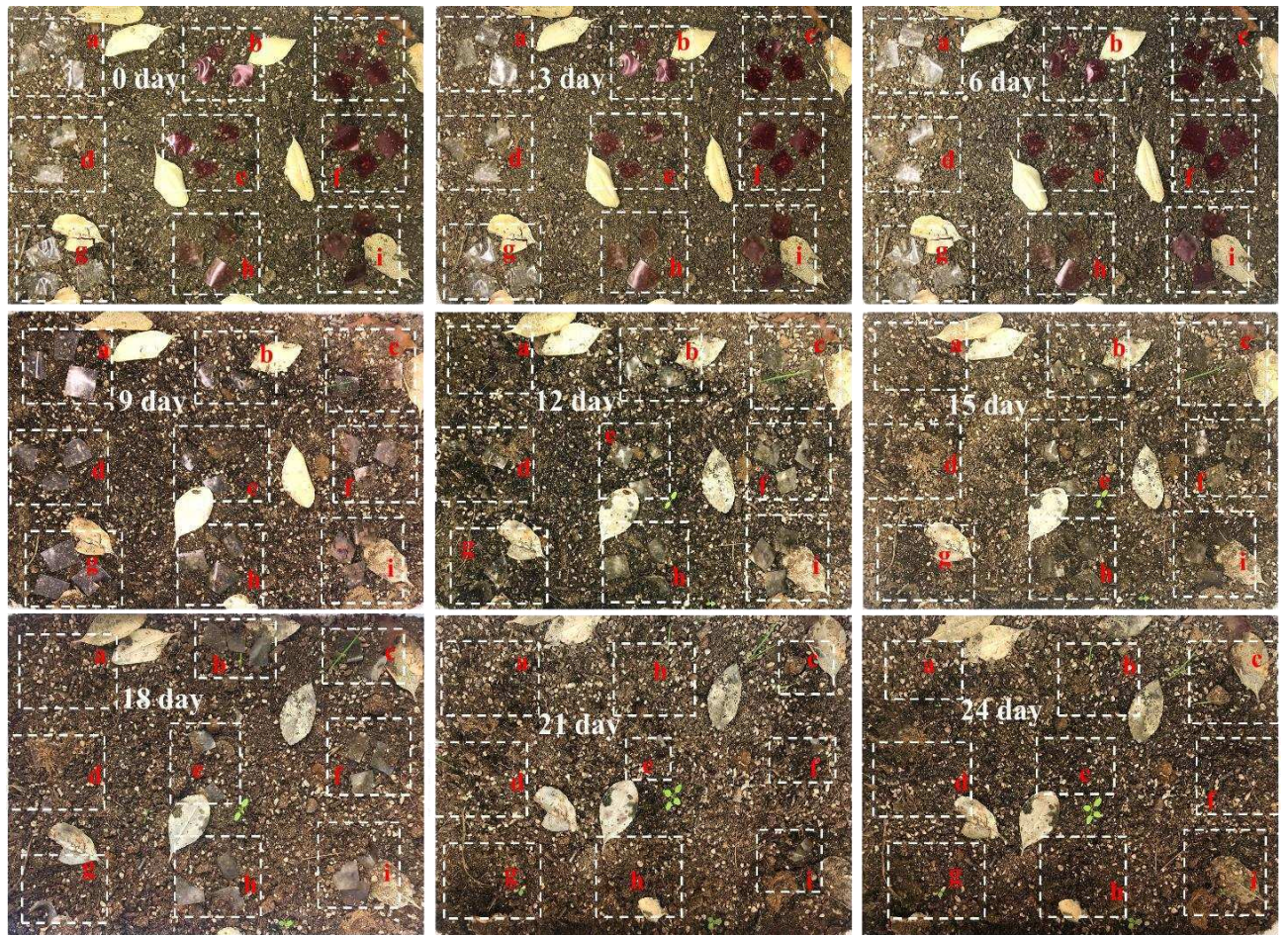
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(A)**(B)**









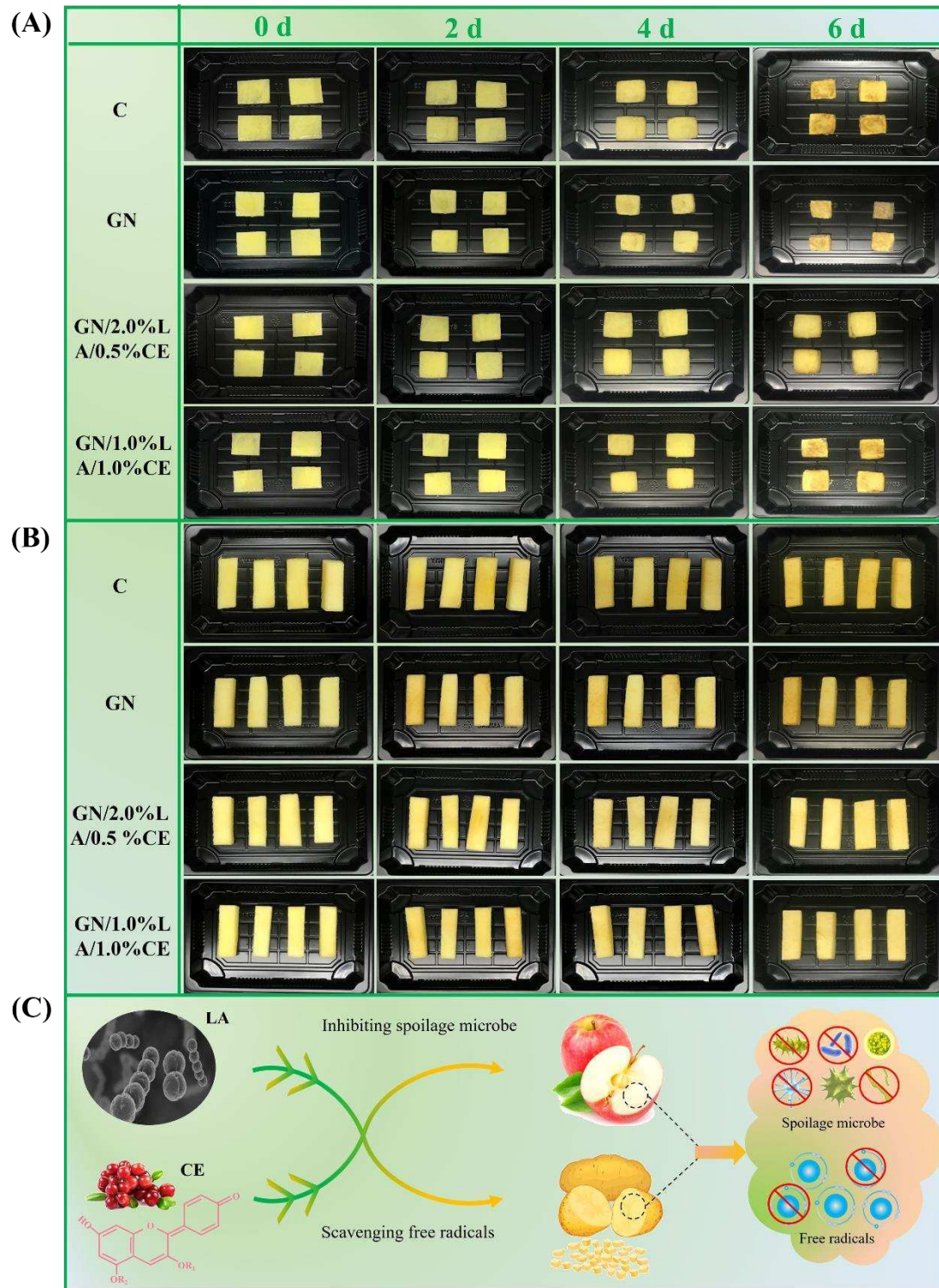


Fig. 1. The SEM morphology (A) and florescence images (B) of the different films.

Fig. 2. The antibacterial (A), antioxidant (B), optical (C) and appearance (D) properties of the different films.

Fig. 3. The survivability of *L. lactis* in the film during drying process (A) and storage process (B), and the first-order kinetic formula of inactivation (C).

Fig. 4. The colorful patterns (A) and electrochemical writing principle (B) on the different films

Fig. 5. The biodegradability of the films in soil in natural for 24 days. (a: GN/0%LA/0%CE films, b: GN/0%LA/0.5%CE films, c: GN/0%LA/1.0%CE films, d: GN/1.0%LA/0%CE films, e: GN/1.0%LA/0.5%CE films, f: GN/1.0%LA/1.0%CE films, g: GN/2.0%LA/0%CE films, h: GN/2.0%LA/0.5%CE films, i: GN/2.0%LA/1.0%CE films)

Fig. 6. The preservation effect of different films on fresh-cut potatoes (A) and apples (B) during the storage period.

Table 1. The tensile strength (TS) and elongation percentage at break (EB) and water vapor permeability (WVP) of the films.

Film samples	TS	EB	WVP
	(MPa)	(%)	(g·mm·m ⁻² ·Kpa ⁻¹ ·h ⁻¹)
GN/0%LA/0%CE	17.92 ± 0.8 ^e	12.32 ± 1.3 ^c	4.71 ± 0.1 ^a
GN/0%LA/0.5%CE	22.62 ± 1.2 ^d	19.0 ± 0.8 ^a	4.66 ± 0.3 ^a
GN/0%LA/1.0%CE	31.70 ± 1.4 ^a	19.23 ± 1.0 ^a	4.27 ± 0.1 ^b
GN/1.0%LA/0%CE	17.12 ± 2.0 ^e	12.47 ± 1.0 ^c	4.63 ± 0.3 ^a
GN/1.0%LA/0.5%CE	22.60 ± 1.3 ^d	18.50 ± 0.6 ^a	4.57 ± 0.1 ^a
GN/1.0%LA/1.0%CE	32.52 ± 1.2 ^a	17.9 ± 2.1 ^a	4.30 ± 0.2 ^b
GN/2.0%LA/0%CE	14.15 ± 1.2 ^f	9.15 ± 0.7 ^d	4.70 ± 0.1 ^a
GN/2.0%LA/0.5%CE	25.50 ± 0.8 ^c	15.44 ± 1.2 ^b	4.65 ± 0.2 ^a
GN/2.0%LA/1.0%CE	28.17 ± 1.7 ^b	15.10 ± 1.0 ^b	4.81 ± 0.2 ^a

Values are expressed as mean ± standard deviation (n = 4). Different small letters indicate significant differences.

Credit author statement

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Conflict of Interest

Xiaobo Zou declares that he has no conflict of interest. Zhikun Yang declares that he has no conflict of interest. Jiyong Shi declares that he has no conflict of interest. Xiaodong Zhai declares that he has no conflict of interest. Xiaowei Huang declares that she has no conflict of interest. Zihua Li declares that he has no conflict of interest. Tao Wang declares that he has no conflict of interest. Li Chuang declares that she has no conflict of interest. Yunyun Gong declares that she has no conflict of interest. Melvin Holmes declares that he has no conflict of interest. Megan Povey declares that she has no conflict of interest.