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2

3 **Novel method for improving the water dispersibility and flowability of fine green**
4 **tea powder using a fluidized bed granulator**

5

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22 **ABSTRACT**

23 To develop new green tea beverages and dietary supplements in powder form, the surface of green
24 tea powder (GTP) was modified by spraying a hydrophilic polymer Soluplus[®] (Sol) solution using a
25 top-spray fluidized bed granulator. This enhanced the water dispersibility and powder flowability of
26 the GTP. Spraying with levels of 0.5%–9% Sol polymers improved the flow properties but
27 significant agglomeration occurred in products coated with 3%–9% Sol, but not in those coated with
28 0.5% and 1% Sol. This suggested that low levels of coating solution inhibited agglomeration. The
29 highest level of water dispersibility and improved flowability with no agglomeration were exhibited
30 by GTP with 1% Sol coating. Overall, surface modification with hydrophilic polymer can improve
31 the flow properties, dispersibility and other properties of GTP. This method might also be useful for
32 developing other novel food ingredient powders.

33

34 *Keywords:* Green tea powder; Dispersibility; Flowability; Surface modification; Fluidized bed
35 coating.

36

37 *Abbreviations:* GTP, green tea powder; Sol, Soluplus; d_{50} , the volume-based mean particle size; R_w ,
38 relative particle size distributions width.

39

40 **1. Introduction**

41 Green tea is one of the most widely consumed beverages in the world (Sinija et al., 2007),
42 especially in Asian countries such as China and Japan. Green tea leaves contain several hydrophilic
43 active ingredients: polyphenolic compounds such as catechins and amino acids such as theanine,
44 which exhibit anti-oxidative, anti-inflammatory, and anti-carcinogenic properties (Moore et al., 2009;
45 Ojo et al., 2007; Sadzuka et al., 1996, 2000). These hydrophilic active ingredients can be easily
46 extracted and diffused in water from the surface of the tea leaves, providing beverages with certain
47 health benefits for consumers (Koo and Cho, 2004). However, other hydrophobic active ingredients,
48 located inside the tea leaves, are less easy to extract and can easily be precipitated in water because
49 of their low water solubility. This means that, at present, green tea beverages cannot provide the full
50 advantage of these active ingredients. Therefore, if beverages can be developed where the green tea
51 powder (GTP) is dispersed well, they would provide several types of active compounds
52 simultaneously compared with those containing only extracts from the tea leaves.

53 Particle size is a key parameter influencing the efficacy of extracting and diffusing active
54 ingredients from GTP and the dispersibility of GTP in water. The appropriate grinding method can
55 reduce the particle size of food ingredients to between 1 and 100 μm . These superfine particles of tea
56 leaf have been reported to increase the extraction efficacy and dispersibility in water because of the
57 increase in particle surface area and the breakdown of the cell walls of the leaf (Hu et al., 2012; Park
58 et al., 2001). When GTP with a smaller particle size is dispersed in water, oral discomfort is also

59 reduced and the rate of sedimentation inhibited. However, for smaller particles, the adhesive forces
60 such as electrostatic and van der Waal's forces increase, while the gravitational or inertial forces
61 decrease, leading to undesirable changes in dispersibility in water and flow properties (Ehlers et al.,
62 2008; Heim et al., 1999). Therefore, during the manufacturing process, it is essential to keep the
63 particle sizes of GTP small without reducing their dispersibility in water and flow properties.

64 Some methods for producing tea powder, such as freeze-drying tea leaves after wet grinding
65 (Saito and Shobu, 1970), or milling tea leaves with saccharides (Park et al., 2001), have been
66 developed to solve these problems. However, freeze-drying improved dispersibility but not the flow
67 properties because of the non-spherical and porous forms of tea powder produced. Direct milling
68 with saccharides improved dispersibility, but was not suitable for practical use because of the high
69 quantity of additives needed. Recently, Xiao et al. (2017) have reported the successful development
70 of superfine black tea powders with high levels of infusion and dispersibility through using a
71 water-dispersible stabilizer, but the procedure required a large quantity of additives.

72 Generally, the surface conditions of powders greatly affect the physical properties of
73 powders such as flowability and dispersibility, which can determine their storage and handling
74 properties as well as their subsequent processing or application (Gaiani et al., 2006, 2011; Kim et al.,
75 2002; Murrieta-Pazos et al., 2012; Nijdam and Langrish, 2006; Shrestha et al., 2007; Szulc and
76 Lenart, 2013). Surface modification has been widely used in the pharmaceutical field to improve the
77 flow properties and solubility of drugs (Ehlers et al., 2008, Miyadai et al., 2010). However, studies

78 on modifying the surface of food ingredient powders such as GTP have not yet been reported.

79 In the present study, we aim to modify the surface properties of GTP by spraying a dilute
80 solution of Soluplus[®] (5%, w/w, BASF Japan Co. Ltd., Tokyo, Japan) using a top-spray fluidized bed
81 granulator to enhance its dispersibility, flowability, and other properties. Fluidized bed coating is
82 selected because this method is widely used in the pharmaceutical and food industries to modify
83 powders to enhance their processability, to mask any unpleasant taste or appearance and to enhance
84 or create functional features such as delayed release or increased stability of the active ingredients
85 (Ehlers et al., 2008; Tenou and Poncelet, 2003; Turton, 2008; Werner et al., 2007). As preliminary
86 experiments, only just GTP did not flow in and adhered to the wall of fluidized bed chamber because
87 of strong electrostatic and van der Waal's forces of GTP, 1% (w/w) fumed silica was added to GTP as
88 a flowing and glidant agent. In addition, surfactants have been reported to decrease the interfacial
89 tension between particles and the dispersion media, leading to enhanced levels of dispersibility
90 (Martin et al., 1993). Polyvinyl caprolactam-polyvinyl acetate-polyethylene glycol graft copolymer
91 Soluplus[®] (Sol), a synthesized amphiphilic polymer with a surfactant action, has recently been
92 developed as a safe polymer for use in the hot-melt extrusion technique for pharmaceutical sciences
93 (Ashour et al., 2016; Thiry et al., 2016). We will use a dilute Sol solution to spray onto GTP at levels
94 up to 9% by weight of the coating and thereby modify its surface characteristics. We will then
95 investigate the effect of Sol on the physical properties, particle size distribution, flow properties,
96 wettability, dispersibility, and dispersion stability of GTP, and its application in developing beverages

97 containing dispersed fine GTP.

98

99

100 **2. Materials and methods**

101 *2.1. Materials*

102 Green tea leaves were provided by Arahataen Co., Ltd. (Shizuoka, Japan). Fumed silica
103 (Aerosil[®] 200, Nippon Aerosil Co. Ltd., Tokyo, Japan) was used as a flowing and glidant agent.
104 Soluplus[®] (Sol) was provided by BASF Japan Co. Ltd. as a coating material.

105

106 *2.2. Preparation of finely green tea powders modified by coating materials*

107 To produce GTP, green tea leaves were milled using a Bisaiki[®] powder mill (Terada
108 Seisakusho Ltd., Shizuoka, Japan) with running water to prevent denaturation and the loss of tea
109 flavor. The GTP was dried for 24 h at 50 °C in an oven. Then 1.5 g fumed silica was added to 150 g
110 of the bulk powder and resultant powders were manually pre-mixed by shaking in a polyethylene bag
111 for 5 min. In the present study, the coating experiments were performed using a top-spray fluidized
112 bed coater (Flow Coater[®] FL-Mini, Freund Corp., Tokyo, Japan) (Fig. 1A). The chamber of the
113 coater was preheated to 35 °C then 150 g of the GTP/fumed silica mixture was put into the chamber
114 and mixed at 30 °C. The coating solution was then sprayed into the chamber, and the flow rate and
115 spray air pressure were adjusted to 1.0 g min⁻¹ and 0.15 MPa, respectively. The composition of the
116 solutions is shown in Table 1. After all the coating solution was added to the GTP, a stream of air at
117 35 °C was passed over the particles to dry them completely.

118

119 *2.3. Characterization of final product properties*

120 *2.3.1. Particle size distribution*

121 The particle size distributions of the uncoated intact GTP and the coated final products were
122 determined through laser diffraction measurement. The Mastersizer 2000 device was used in
123 combination with the Hydro 2000 S dispenser (both Malvern Instruments Ltd, Malvern, UK) and
124 deionized water was used as the dispersion medium in the measuring cell. The relative width of
125 particle size (R_w) was calculated using the following formula.

126
$$R_w = (d_{90} - d_{10}) / d_{50}$$

127 where d_{10} , d_{50} , and d_{90} are 10%, 50%, and 90% of typical particle size distribution,
128 respectively.

129

130 *2.3.2. Carr's fluidity index*

131 The flow properties of the products were determined using Carr's method (Carr, 1965). Four
132 tests measured compressibility, angle of repose, angle of spatula and uniformity coefficient. The
133 uniformity coefficient was obtained by particle size analysis and the other indices using a powder
134 characteristics tester (Powder Tester[®], Hosokawa Micron Co., Ltd., Osaka, Japan).

135 To determine the compressibility of the products, the bulk density was first measured using
136 a 100-mL cylinder filled with an accurately weighed sample of the product with the top leveled off.
137 The bulk density was calculated as the ratio of the mass to the volume of the sample. The tapped

138 density was determined using the same method and mass of powder, but the volume was measured
139 after 200 taps. The compressibility was calculated by using the bulk and tapped density values in the
140 following equation:

$$141 \text{ Compressibility (\%)} = [(\text{tapped density} - \text{bulk density}) / \text{tapped density}] \times 100.$$

142 The angle of repose was measured using a protractor from a heap of the powder formed by
143 dropping the GTP product through a funnel. The angle of spatula was measured using a protractor
144 and a steel spatula with a blade (22 mm × 120 mm). The spatula was inserted into the bottom of a
145 carefully built heap of powder. The spatula was then withdrawn vertically, and the angle of the heap
146 formed by the spatula was measured as the angle of spatula.

147 The uniformity coefficient was calculated using the following equation:

$$148 \text{ Uniformity coefficient} = d_{60} / d_{10}$$

149 where d_{10} and d_{60} are 10% and 60% of the accumulated particle size, respectively.

150 Twenty five scores were assigned for each examination, and the flowability index was then
151 calculated using the point scores out of 100 as described previously (Carr, 1965), where scores from
152 90 to 100 represent “excellent” flowability, 80 to 89, “good”; 70 to 79, “fair”; 60 to 69, “passable”;
153 40 to 59, “poor”; 20 to 39, “very poor”; and 0 to 19, “extremely poor”.

154

155 2.3.3. *Wettability*

156 The wettability of the uncoated intact GTP and the coated products was determined by

157 measuring the contact angle of a water droplet and its penetration time. A 500-mg GTP sample was
158 compressed using a hydraulic single-punch tableting machine set at 100 MPa. A 10- μ L water droplet
159 was then placed on the tablet of GTP and the contact angle measured using the $\theta/2$ method (Yang and
160 Lin, 2003). The penetration time was the time taken for the droplet to soak completely into the GTP
161 tablet.

162

163 *2.3.4. Dispersibility and Dispersion stability*

164 For dispersibility, 100 ml distilled water were poured into a measuring cylinder then a
165 200-mg GTP sample was put on the water surface. The degree of turbidity of the water 6.5 cm below
166 the surface was measured as an index of dispersibility using a UV-Vis spectrophotometer
167 (UVmini-1240[®], Shimadzu Corp., Kyoto, Japan) at an absorbance wavelength of 650 nm, 3, 5 and
168 10 min after putting the GTP on the water surface (Fig. 1B).

169 For the dispersion stability, 100 mL of water dispersions containing 200 mg of intact GTP
170 and coated GTP products were stirred for 1 min and then transferred to an empty measuring cylinder.
171 The degree of turbidity was then determined as described above. The relative dispersion stability was
172 calculated using the following equation:

$$173 \quad \text{Relative dispersion stability (\%)} = [\text{Turbidity}_{10\text{min}} / \text{Turbidity}_{3\text{min}}] \times 100$$

174 where $\text{Turbidity}_{3\text{min}}$ and $\text{Turbidity}_{10\text{min}}$ are the turbidities of the dispersion measured 3 and
175 10 min after being transferred to the empty measuring cylinder (Fig. 1B).

176

177 *2.4. Statistical analysis*

178 Statistical analysis was carried out using the Student's *t*-test and ANOVA. A probability

179 value of $p < 0.05$ was considered to indicate a significant difference between mean values.

180 3. Results and discussion

181 3.1. Physicochemical characteristics of GTPs coated with Sol

182 The particle size distributions of GTP and GTP sprayed with Sol are shown in Fig. 2. The
183 volume-based mean particle size (d_{50}) and width of the relative particle size distributions (R_w) of
184 these GTPs are shown in Table 2. These results indicated that the agglomeration of GTP increased as
185 the level of Sol coating increased. In particular, the 1%, 3%, 6%, and 9% Sol coatings caused
186 significant agglomeration. In contrast, there were no significant differences in d_{50} and R_w values
187 between the intact GTP and the GTP with a Sol 0.5% coating, suggesting that using a small amount
188 of the coating solution did not accelerate the agglomeration progress.

189 Table 3 shows the mean values of flowability of GTP and GTP coated with Sol. In general,
190 smaller values of compressibility, angle of repose, angle of spatula and uniformity coefficient led to
191 larger values of Carr's fluidity index, indicating better flowability. Table 3 shows that Carr's fluidity
192 index for GTP was 51.5 (Poor). This value indicates that the intact GTP was difficult to handle
193 because of its high cohesive properties. Carr's fluidity index for all the products increased as the
194 level of Sol coating increased, ranging from 59.0 to 74.0.

195 The particle size of powders strongly influences their flowability (Jinapong et al., 2007). In
196 other words, the improvement in flowability as the level of Sol coating increased may be influenced
197 by particle agglomeration. However, even though the particle size of the products coated with 0.5%
198 Sol did not change, the Carr's fluidity index increased from 51.5 to 59.0 with the flow properties

199 changing from “Poor” to almost “Passable” (60 to 69). Based on these results, the flowability
200 improved without any significant increase in the mean particle size, and thus it might be possible to
201 modify the surface of intact GTP just by spraying.

202

203 *3.2. Wettability, Dispersibility and Dispersion stability of GTP*

204 The penetration time and contact angle decreased as the level of Sol coating increased (Fig.
205 3), meaning that the wettability increased. In particular, the wettability of the products coated with
206 3%, 6%, and 9% Sol was significantly lower than that of the intact GTP because the surface area
207 covered by hydrophilic polymers increased.

208 To illustrate the dispersibility characteristics of GTP, Fig. 4A shows representative photos,
209 30 s after putting intact GTP and GTP coated with 1% Sol on the water surface. The intact GTP was
210 poorly dispersed in water because of its cohesive properties, thus tending to form coarse
211 agglomerations, which readily settled as a sediment. In contrast, the GTP coated with 1% Sol
212 exhibited a remarkably high water dispersibility, indicating a decreasing adhesive force between the
213 particles. Fig. 4B shows that the water dispersibility of GTP coated with 1% Sol increased
214 significantly but decreased in products coated at Sol levels of 9%. Overall, the flowability and
215 wettability of GTP products coated with 0.5% and 1% Sol tended to improve through surface
216 modification because more particles could make contact with the water and be dispersed without
217 agglomerating. However, in products coated with high levels of Sol, the increase in the mean particle

218 size increased the sedimentation rate. The sedimentation rate in water increases in proportion to the
219 square of the mean particle size according to Stokes' Law for the same density and viscosity of the
220 medium and the same particle density. Therefore, this could explain the reason for the poor
221 dispersibility of products coated with high levels of Sol.

222 The dispersion stability of intact GTP and the other coated GTP products was also evaluated
223 over time. Fig. 5A shows the high dispersion stability of intact GTP and GTP products coated with
224 0.5% Sol. Since there was a possibility that differences in the numbers of particles associated with
225 both d_{50} and R_w affected these results, the relative dispersion stabilities shown in Fig. 5B were
226 calculated using the equation described in section 2.3.4. The intact GTP and GTP products with 0.5%,
227 1% and 6% Sol coatings exhibited high values of relative dispersion stability because the mean
228 particle size of products with 0.5% and 1% Sol coatings was small. However, GTP coated with 6%
229 Sol would have been predicted to exhibit a low value of relative dispersion stability, but its value was
230 high because it contained a large proportion of fine particles as shown in Fig.2. As expected, those
231 powders with a large mean particle size (coated with 3% and 9% Sol) exhibited a low relative
232 dispersion stability.

233

234 **4. Conclusions**

235 GTP produced by grinding green tea leaves to a particle size of less than 100 μm could be a
236 useful material for developing new beverages and dietary supplements. When the size of the primary

237 particles is small, the adhesive forces between them become larger, resulting in poorer flow
238 properties and lower dispersibility in water. The surface properties of GTP were therefore modified
239 by spraying with a hydrophilic polymer (Sol solution) using a top-spray fluidized bed granulator at
240 coating levels between 0.5% and 9%. The particle size distribution, flow properties, wettability,
241 dispersibility and dispersion stability were all evaluated, which showed that the flow properties of all
242 the coated products had improved. The GTP product coated with 1% Sol exhibited the highest level
243 of dispersibility. The present study has shown that coating GTP particles with a small amount of a
244 hydrophilic polymer improved physical properties, such as flowability and dispersibility, which
245 depend on the surface properties of the particles. Although further study using other polymers would
246 be need, GTP modified in this way may be useful for developing new green tea beverages and other
247 processed products including dietary supplements in the form of food powders.

248

249 **Acknowledgements**

250 We are extremely grateful to the BASF Japan Co. Ltd. for providing the Soluplus[®] for this
251 study. We would also like to thank the Arahataen Co. Ltd. for providing the green tea leaves.

252

253 **Conflict of Interest**

254 The authors declare no conflict of interest.

255

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327

328 **Figure captions**

329 **Fig. 1.** The top-spray fluidized bed coater (A) and the procedure for evaluating the dispersibility and
330 dispersion stability of GTP products.

331

332 **Fig. 2.** Particle size distribution of intact GTP and Sol-coated GTP products.

333

334 **Fig. 3.** Contact angle and penetration time of intact GTP and GTP products coated with Sol at levels
335 from 0.5% to 9%. Data are means \pm SD (bars), $n = 3$, significance of difference with GTP: *, $p <$
336 0.05 and **, $p < 0.01$.

337

338 **Fig. 4.** (A) Representative photos showing dispersibility 30 s after placing powder on the water
339 surface for (a) Intact GTP and (b) GTP with 1% Sol coating. (B) Changes in the dispersibility of
340 intact GTP and GTP products coated with Sol at levels from 0.5% to 9% after 3, 5 and 10 min. Data
341 are means \pm SD (bars), $n = 3$, significance of difference with GTP: *, $p < 0.05$.

342

343 **Fig. 5.** Time-dependent changes in (A) dispersion stability, and (B) relative dispersion stability, of
344 intact GTP and GTP products coated with Sol at levels from 0.5% to 9%. Data are means \pm SD (bars),
345 $n = 3$, significance of difference with GTP: *, $p < 0.05$ and **, $p < 0.01$.

346

Figure 1

A)



B)

Dispersibility Dispersion stability

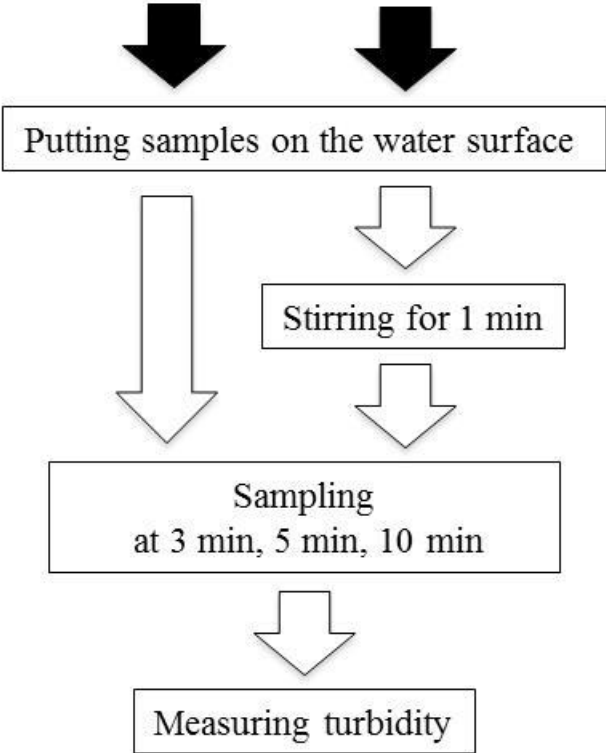
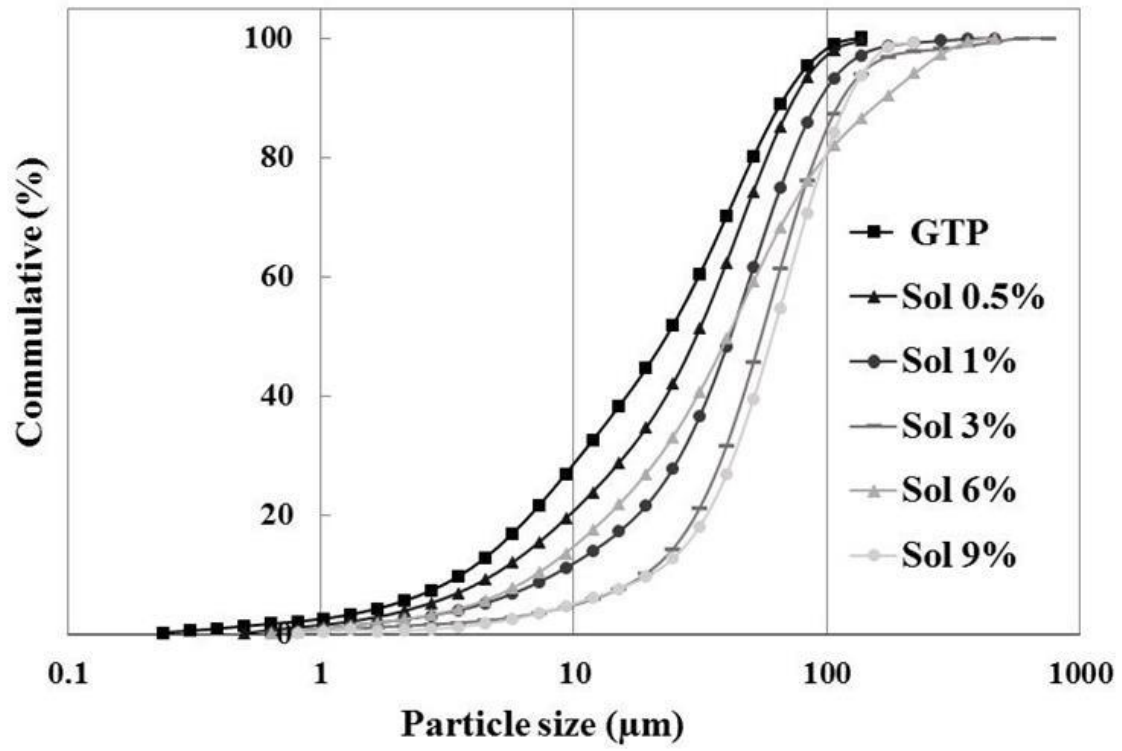
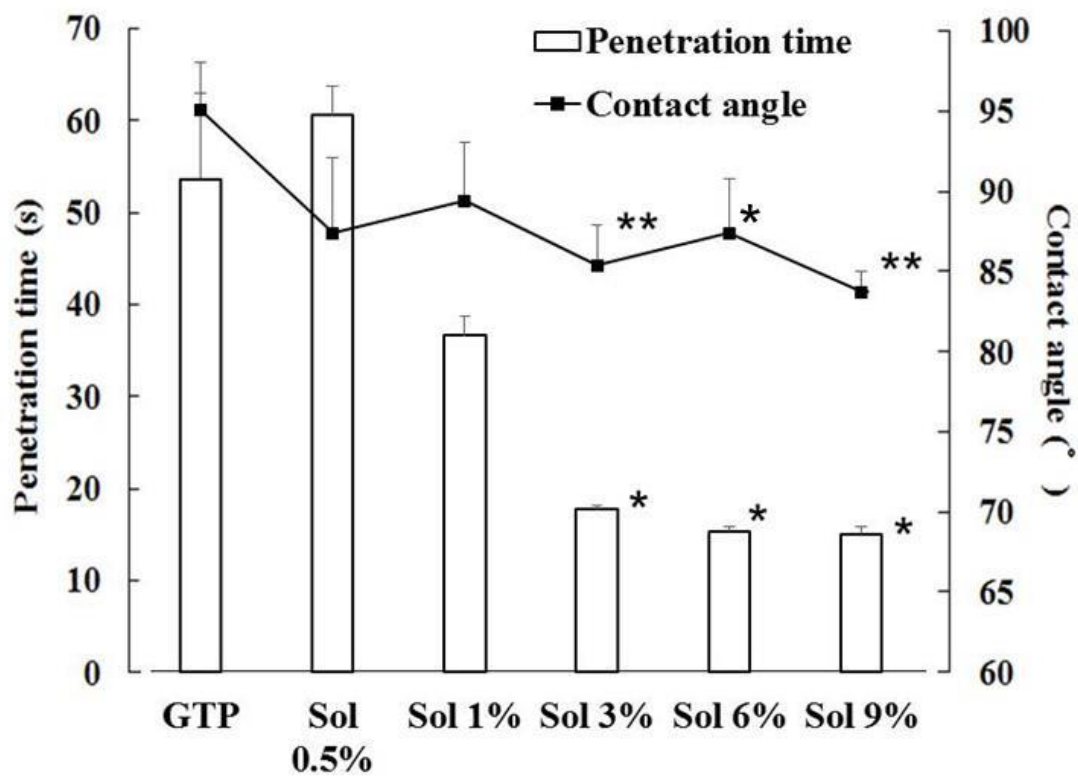


Figure 2



348

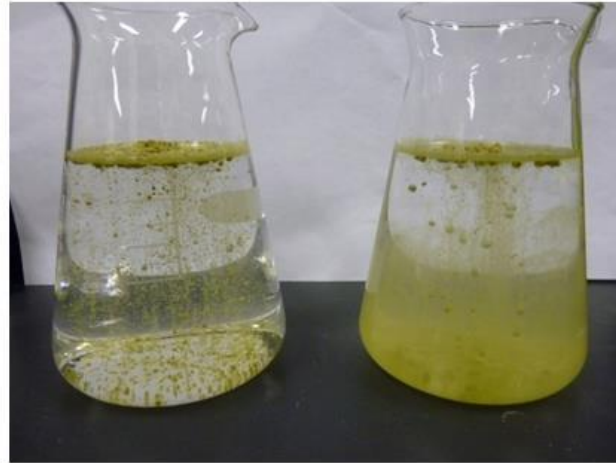
Figure 3



349

Figure 4

A)



(a) Intact GTP

(b) Sol 1% coating

B)

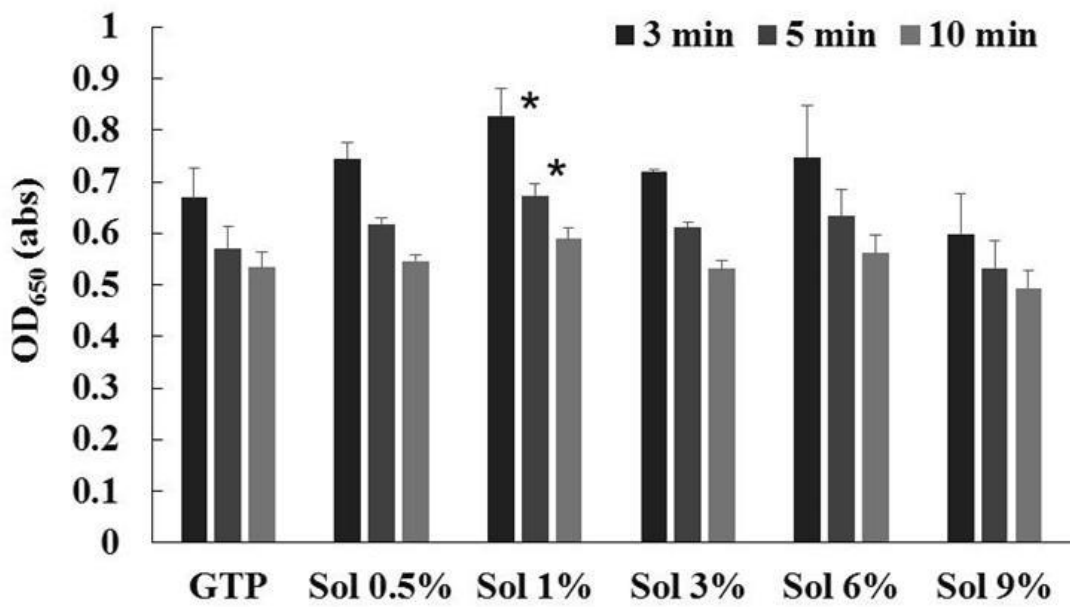
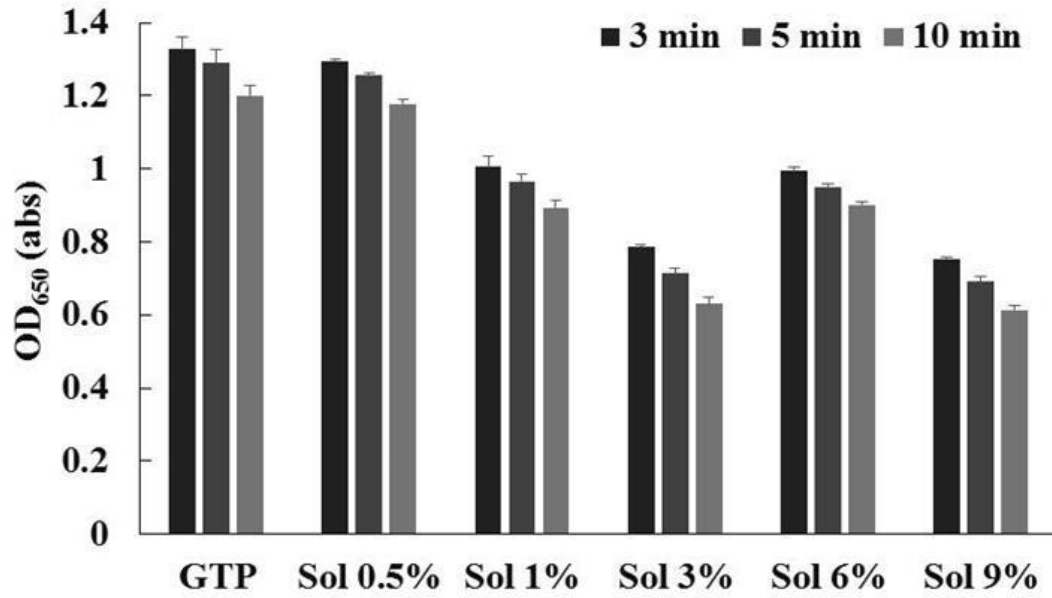
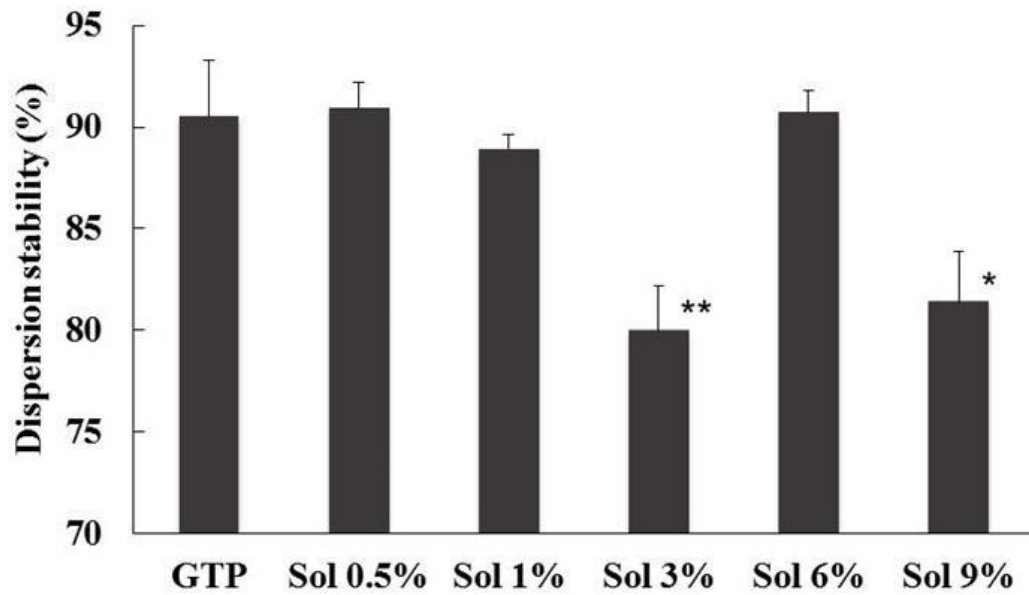


Figure 5

A)



B)



351

352

353 **Table 1.**

354 The composition of GTP coated with Sol at levels from 0.5% to 9%.

355

		Sol				
		0.5%	1%	3%	6%	9%
Physical mixture						
GTP	(g)	150	150	150	150	150
Fumed silica	(g)	1.50	1.50	1.50	1.50	1.50
Coating solution						
Sol	(g)	0.75	1.50	4.50	9.00	13.5

356

357 **Table 2.**358 The mean particle size (d_{50}) and relative particle size distributions width (R_w) of intact GTP and GTP
359 products coated with Sol at levels from 0.5% to 9%.

360

GTP		Sol				
		0.5%	1%	3%	6%	9%
d_{50} (μm)	28.2 ± 4.2	30.9 ± 0.8	$39.9 \pm 1.1^{**}$	$51.2 \pm 3.6^{**}$	$41.3 \pm 5.5^{**}$	$62.7 \pm 2.2^{**}$
R_w	2.41 ± 0.31	2.33 ± 0.09	1.93 ± 0.01	1.76 ± 0.01	3.06 ± 0.81	1.72 ± 0.03

361 $n = 3$, mean \pm SD. Significance of difference with GTP: **, $p < 0.01$.

362

363 **Table 3.**

364 Flowability of intact GTP and GTP products coated with Sol at levels from 0.5% to 9%.

365

GTP		Sol				
		0.5%	1%	3%	6%	9%
Uniformity	7.6 ± 0.8	8.1 ± 0.1	6.1 ± 0.7	4.2 ± 0.7	7.6 ± 0.2	3.5 ± 0.3
Compressibility (%)	36.5 ± 0.6	30.0 ± 1.2	30.2 ± 1.0	29.0 ± 1.0	27.5 ± 0.9	23.9 ± 0.9
Angle of repose ($^\circ$)	45.6 ± 1.9	45.4 ± 0.4	43.8 ± 0.7	47.4 ± 1.6	39.3 ± 1.9	38.4 ± 1.7
Spatula angle ($^\circ$)	68.0 ± 0.4	63.9 ± 0.2	60.4 ± 2.9	54.8 ± 1.7	53.9 ± 4.3	53.5 ± 1.3
Carr's fluidity index	51.5	59.0	65.0	64.0	66.0	74.0
Flow property	Poor	Poor	Passable	Passable	Passable	Fair

366 $n = 3$, mean \pm SD.