Supplementary information

Development of Strong and High-Barrier Food Packaging Films from

Cyclic-Anhydride Modified Bacterial Cellulose

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| Properties | BC | DSA/ | DMSO (| (w/w) | | | OSA/ | DMSO (v | v/w) | ODSA/DMSO (w/w) | | | | MA/DMSO (w/w) | | |
|------------|------|------|--------|-------|------|------|-----------|-----------|------|-----------------|------|------|--|---------------|-------|------|
| 1 | | 0.03 | 0.05 | 0.1 | 0.15 | 0.2 | 0.03 | 0.05 | 0.1 | 0.1 | 0.15 | 0.2 | | 0.01 | 0.03 | 0.05 |
| TS | 28.2 | 49.5 | 66.3 | 83.5 | 83.3 | 35.1 | 3.1 | 110.7 | 88.6 | 45.0 | 53.6 | 22.9 | | 43.5 | 133.4 | 36.9 |
| (MPa, dry) | ±1.5 | ±3.3 | ±5.1 | ±4.5 | ±3.5 | ±2.6 | ± 0.4 | ±3.5 | ±3.5 | ±2.3 | ±3.2 | ±3.4 | | ±1.1 | ±3.3 | ±3.5 |
| TS | | 0.5 | 3.2 | 15.8 | 11.5 | 30.1 | 0.6 | 0.5 | 0.7 | 5.2 | 5.6 | 0.7 | | 13.7 | 22.5 | 25.4 |
| (MPa, wet) | - | ±0.2 | ±0.6 | ±1.2 | ±2.1 | ±3.3 | ± 0.1 | ± 0.1 | ±0.1 | ±0.3 | ±0.2 | ±0.2 | | ±1.2 | ±1.9 | ±2.1 |

Table S1Dry and wet TS of the pristine BC film and anhydride modified films at 70 °C for 5 min

Table S2

Chemical compositions of the pristine BC and anhydride modified films at 10 min-3 h

| Atom (wt %) | | 3%С0- | BC | | | 5%C8- | BC | | | 10%C1 | 2-BC | | | 15%C18-BC | | | |
|----------------|-------|------------|------------|------------|------------|-----------|-----------|------------|-------|------------|-----------|-------|-------|-----------|-----------|------------|------------|
| | BC | 10 min | 30 min | 1 h | 3 h | 10 min | 30 min | 1 h | 3 h | 10 min | 30 min | 1 h | 3 h | 10 min | 30 min | 1 h | 3 h |
| C | 46.39 | 49.97 | 49.68 | 49.79 | 50.19 | 51.07 | 52.16 | 52.65 | 52.75 | 50.23 | 50.68 | 50.81 | 51.73 | 49.70 | 51.88 | 53.36 | 54.56 |
| C | ±0.29 | ±0.16 | ± 0.18 | ± 0.22 | ±0.24 | ±0.20 | ±0.23 | ± 0.25 | ±0.23 | ±0.17 | ±0.27 | ±0.19 | ±0.26 | ±0.16 | ±0.23 | ± 0.24 | ± 0.28 |
| 0 | 53.49 | 48.93 | 49.43 | 49.13 | 49.18 | 47.60 | 45.91 | 45.85 | 46.00 | 48.94 | 48.30 | 48.02 | 46.90 | 50.12 | 46.14 | 45.34 | 43.69 |
| 0 | ±0.33 | ±0.21 | ±0.12 | ±0.15 | ±0.19 | ±0.16 | ±0.15 | ± 0.18 | ±0.16 | ± 0.11 | ±0.13 | ±0.12 | ±0.15 | ±0.20 | ±0.17 | ±0.13 | ±0.12 |
| c | 0.12 | 1.09 | 0.88 | 1.08 | 0.63 | 1.33 | 1.93 | 1.47 | 1.26 | 0.83 | 1.03 | 1.17 | 1.37 | 0.19 | 1.98 | 1.30 | 1.75 |
| 5 | ±0.02 | ± 0.05 | ±0.10 | ± 0.04 | ± 0.07 | ±0.09 | ±0.10 | ± 0.05 | ±0.10 | ±0.07 | ±0.06 | ±0.03 | ±0.07 | ±0.03 | ±0.06 | ± 0.07 | ± 0.08 |

Table S3Mechanical strength of the pristine BC film and anhydride modified films at 10 min–3 h

| | | 3%C0 |)-BC | | | 5%C8 | -BC | | | 10%C | 12-BC | | | 15%0 | C18-BC | | |
|------------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|------|-----------|-----------|-----------|-------|-----------|
| Properties | BC | 10 min | 30 min | 1 h | 3 h | 10 min | 30 min | 1 h | 3 h | 10 min | 30 min | 1 h | 3 h | 10 min | 30 min | 1 h | 3 h |
| TS | 28.2 | 87.8 | 89.5 | 49.9 | 58.9 | 88.8 | 116.6 | 66.1 | 66.4 | 84.6 | 124.3 | 93.3 | 48.1 | 49.7 | 110.4 | 116.3 | 91.2 |
| (MPa, dry) | ±1.5 | ±2.6 | ±2.6 | ± 3.8 | ±3.5 | ±3.7 | ±2.3 | ±9.9 | ±3.1 | ± 2.8 | ±2.1 | ±2.5 | ±3.1 | ± 3.8 | ± 2.9 | ±3.5 | ± 4.0 |
| EAB | 13.6 | 7.7 | 7.3 | 3.8 | 5.3 | 10.0 | 13.7 | 8.6 | 10.4 | 12.9 | 20.1 | 10.3 | 5.1 | 4.8 | 11.4 | 12.2 | 10.1 |
| (%, dry) | ± 0.1 | ±0.6 | ±0.4 | ±0.3 | ± 0.4 | ± 1.1 | ±2.0 | ±1.5 | ± 1.0 | ±0.6 | ± 0.5 | ±0.9 | ± 1.0 | ± 0.5 | ±1.2 | ±0.3 | ±0.6 |
| TS | | 16.5 | 23.0 | 75.4 | 48.1 | 7.4 | 58.8 | 53.9 | 30.9 | 37.5 | 81.0 | 75.7 | 22.7 | 15.1 | 25.3 | 71.4 | 73.6 |
| (MPa, wet) | - | ± 1.8 | ±1.5 | ±3.3 | ±2.2 | ±1.4 | ±3.7 | ± 4.0 | ±3.1 | ±1.1 | ±1.7 | ±3.1 | ±3.1 | ±1.2 | ±1.9 | ±4.1 | ±3.3 |
| EAB | | 2.6 | 4.6 | 13.5 | 8.9 | 2.8 | 26.4 | 16.6 | 9.6 | 15.4 | 15.0 | 17.5 | 5.8 | 7.0 | 7.9 | 17.8 | 13.9 |
| (%, wet) | - | ±0.2 | ±0.3 | ±0.5 | ±0.5 | ±0.3 | ±2.2 | ±1.5 | ±0.5 | ±1.7 | ± 0.8 | ±0.9 | ±0.5 | ±0.4 | ±0.4 | ±0.6 | ±1.6 |



Figure S1. XRD of the pristine BC and 3%C0, 5%C8, 10%C12, 15%C18-30 min, 1 h films.



Figure S2. SEM images (cross-section) of **(a)** 3%C0-BC-1 h, **(b)** 5%C8-BC-1 h, **(c)** 10%C12-BC-1 h, **(d)** C15-BC-1 h at 20,000×, and **(e)** the pristine BC, **(f)** 5%C8-BC-1 h at 5,000×.

| Table S4 |
|---|
| Water resistance of the pristine BC and anhydride modified BC films at 10 min-3 h |

| Watan | | 3%C0- | BC | | | 5%C8- | BC | | | 10%C | l2-BC | | | 15%C18-BC | | | |
|--|---------------|---------------|---------------|--------------------|---------------|---------------|---------------|---------------|---------------|---------------|---------------|---------------|---------------|---------------|---------------|---------------|---------------|
| resistance | BC | 10 min | 30 min | 1 h | 3 h | 10 min | 30 min | 1 h | 3 h | 10 min | 30 min | 1 h | 3 h | 10 min | 30 min | 1 h | 3 h |
| WCA, | 64 | 73 | 78 | 76 | 81 | 93 | 96 | 88 | 92 | 82 | 96 | 97 | 101 | 93 | 96 | 103 | 104 |
| 1 s (°) | ±2 | ±3 | ±3 | ±1 | ±2 | ±3 | ±2 | ±1 | ±2 | ±3 | ±2 | ±3 | ±2 | ±3 | ±2 | ±2 | <u>+2</u> |
| WCA, | 18 | 47 | 52 | 54 | 54 | 78 | 83 | 71 | 75 | 52 | 70 | 76 | 78 | 77 | 83 | 90 | 89 |
| 5 min (°) | ±2 | ± 3 | ±2 | ±1 | ±2 | ±3 | ±1 | ±3 | ±3 | ±3 | ±2 | ±3 | ±2 | ±2 | ± 3 | ±2 | ±1 |
| WS (%) | - | 18.2 ±1.3 | 9.5 ±0.7 | $^{8.3}_{\pm 0.5}$ | 4.4 ±0.5 | 45.8 ±2.2 | 25.7 ±1.9 | 21.2 ±0.7 | 5.7 ±0.3 | 56.5 ±2.3 | 40.0 ±2.1 | 39.1 ±1.2 | 26.7 ±1.3 | 27.8 ±1.2 | 19.2 ±0.4 | 16.7 ±0.8 | 15.8 ±1.3 |
| WVP (×10 ⁻¹⁰ g· $m^{-1} \cdot s^{-1} \cdot pa^{-1}$) | 3.11 ±0.08 | 2.43 ±0.07 | 2.48 ±0.06 | 2.84 ±0.07 | 2.21 ±0.06 | 2.93 ±0.11 | 1.92 ±0.06 | 1.46 ±0.05 | 1.26 ±0.09 | 2.82 ±0.07 | 2.35 ±0.11 | 2.47 ±0.16 | 2.10 ±0.09 | 2.53 ±0.10 | 1.89 ±0.17 | 1.03 ±0.10 | 1.57 ±0.15 |

| Property | 10%C8- | BC | | | 15%C8-BC | | | | | | |
|---|---------------|---------------|---------------|---------------|---------------|---------------|---------------|---------------|--|--|--|
| | 10 min | 30 min | 1 h | 3 h | 10 min | 30 min 1 h | | 3 h | | | |
| WVP (×10 ⁻¹⁰ g·m ⁻¹ ·s ⁻¹ ·pa ⁻¹) | 1.16 ±0.03 | 0.87 ±0.02 | 0.51 ±0.03 | 0.92 ±0.01 | 2.48 ±0.02 | 2.39 ±0.04 | 1.77 ±0.06 | 1.06 ±0.04 | | | |

Table S5 WVP of 10%C8-BC-10 min–3 h and 15%C8-BC-10 min–3 h at 30 °C, 75%

Table S6

Mechanical strength of 10%C8-BC-10 min-3 h

| Properties | 10%C8- | 10%C8-BC | | | | | | | | | | |
|-------------------------|-----------|-----------|-----------|-----------|--|--|--|--|--|--|--|--|
| | 10 min | 30 min | 1 h | 3 h | | | | | | | | |
| TS (MDa dmy) | 54.6 | 95.1 | 71.4 | 62.6 | | | | | | | | |
| 15 (IVIFa, uly) | ±2.5 | ±1.7 | ±3.3 | ±3.3 | | | | | | | | |
| EAR(0/a) | 7.8 | 15.4 | 11.3 | 10.7 | | | | | | | | |
| EAD $(70, \text{ ury})$ | ± 0.5 | ± 0.5 | ± 0.5 | ± 0.8 | | | | | | | | |
| TS (MDa wat) | 25.2 | 52.8 | 37.8 | 59.8 | | | | | | | | |
| 15 (MFa, wet) | ±2.1 | ±3.9 | ±3.0 | ±3.6 | | | | | | | | |
| EAP(0/2) wet | 9.3 | 15.6 | 12.4 | 10.5 | | | | | | | | |
| LAD (70, wet) | ±0.4 | ±0.5 | ±0.9 | ± 0.8 | | | | | | | | |

Table S7

Mechanical strength of 15%C8-BC-10 min-3 h

| Properties | 10%C8- | BC | | |
|-------------------------|-----------|-----------|-----------|-----------|
| Toperties | 10 min | 30 min | 1 h | 3 h |
| TS (MDa dury) | 46.2 | 59.0 | 35.9 | 48.4 |
| 15 (MPa, dry) | ±2.8 | ± 2.8 | ±4.7 | ±3.0 |
| EAP(0/am) | 4.7 | 8.7 | 3.4 | 5.6 |
| EAD $(70, \text{ ury})$ | ±0.9 | ± 0.5 | ± 0.4 | ±0.6 |
| TS (MDa wet) | 26.4 | 35.5 | 37.6 | 41.8 |
| 15 (MFa, wet) | ± 4.0 | ±3.4 | ±4.9 | ± 3.8 |
| EAP(0/a) wet | 6.8 | 10.1 | 9.6 | 10.3 |
| LAD (70, wet) | ±0.5 | ±1.2 | ± 0.5 | ±1.1 |



Figure S3. The photographs of the strawberries covered by the (a) pristine BC, (b) 3%C0-BC-1 h, (c) 10%C8-BC-1 h, and (d) LDPE films after storing at 4 °C for 7 d.

Table S8

| Weight loss (%) of the uncovered, and pristine BC/3%C0-BC-1 h/10%C8-BC-1 h/LDPE covered |
|---|
| strawberries during storage in refrigerator for 1–7 d |

| Samples | Weight l | oss (%) | | |
|-------------------------|------------|------------|------------|------------|
| | 1 d | 3 d | 5 d | 7 d |
| | 4.73 | 12.91 | 22.05 | 36.44 |
| uncovered | ±0.16 | ±0.15 | ±0.21 | ±1.09 |
| DC film asymptot | 2.10 | 6.65 | 11.13 | 20.07 |
| BC IIIII covered | ± 0.09 | ± 0.10 | ± 0.11 | ± 0.40 |
| 20/C0 DC 1 h asymptotic | 2.03 | 6.33 | 16.22 | 16.09 |
| 3%CO-BC-1 Il covered | ± 0.07 | ± 0.18 | ±0.26 | ±0.10 |
| 10%C8-BC-1 h | 1.14 | 3.59 | 5.77 | 9.04 |
| covered | ±0.12 | ± 0.18 | ±0.13 | ±0.19 |
| I DDE film | 1.01 | 3.13 | 4.85 | 7.88 |
| | ± 0.04 | ± 0.07 | ± 0.07 | ±0.26 |

Table S9

| | | 3%C0-BC | | | 5%C | 5%C8-BC | | | | С12-В | С | | 15% | 15%C18-BC | | | |
|-----------------------|-----|-----------|-----------|-----|-----|-----------|-----------|-----|-----|-----------|-----------|-----|-----|-----------|-----------|-----|-----|
| Properties | BC | 10 min | 30 min | 1 h | 3 h | 10 min | 30 min | 1 h | 3 h | 10 min | 30 min | 1 h | 3 h | 10 min | 30 min | 1 h | 3 h |
| T _{max} (°C) | 338 | 370 | 361 | 364 | 357 | 349 | 371 | 369 | 364 | 360 | 371 | 370 | 362 | 364 | 367 | 369 | 363 |

Thermal stability of the pristine BC and 3%C0, 5%C8, 10%C12, 15%C18-BC-10 min-3 h