

Supplementary Information

Self-healable and biodegradable konjac-glucomannan-blended sodium alginate films for food packaging

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Supplementary Notes

Supplementary Note 1	S-2
Supplementary Note 2	S-3
Supplementary Note 3	S-4
Supplementary Note 4	S-5
Supplementary Note 5	S-6
Supplementary Note 6	S-7
Supplementary Note 7	S-8

Supplementary Notes

Supplementary Note 1

Surface morphologies of self-healable and biodegradable films

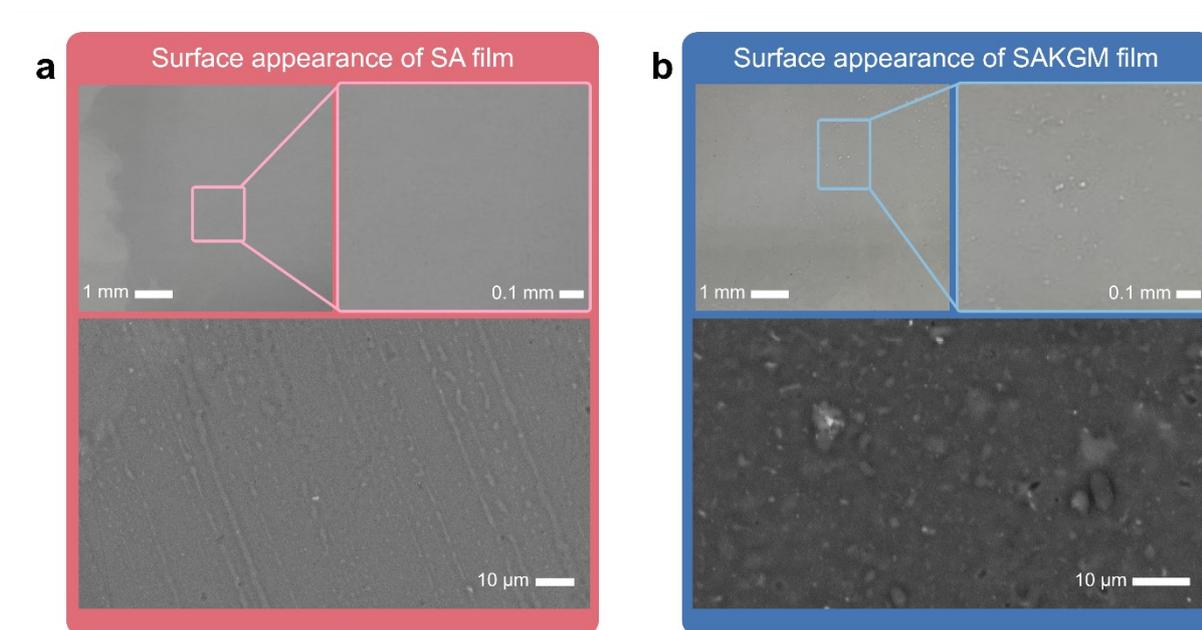


Figure S1. Surface morphologies of self-healable and biodegradable films. **a** Pure sodium alginate (SA) film. **b** Konjac-glucomannan-blended sodium alginate (SAKGM) film. Both SA and SAKGM films show crack-free and pore-free surfaces indicating the successful film formation. SA film exhibits continuous and smooth surface, while SAKGM film shows rougher surface with numerous ungelatinized polysaccharide granules.

Supplementary Note 2

Self-healing performances of biodegradable films

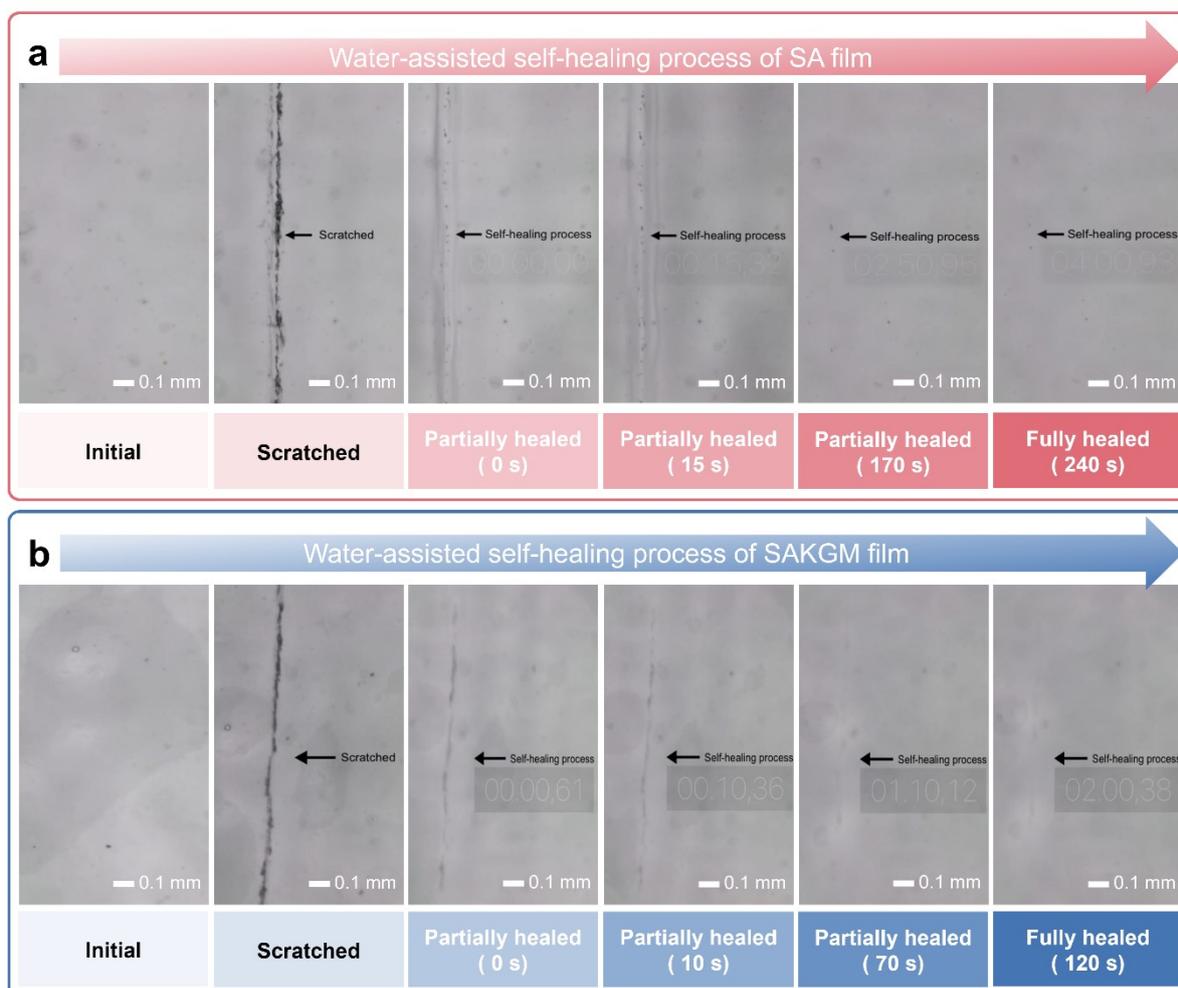


Figure S2. Water-assisted self-healing processes of biodegradable films observed under an optical microscope. **a** Pure sodium alginate (SA) film. **b** Konjac-glucomannan-blended sodium alginate (SAKGM) film. The water droplet from a syringe initiates intrinsic mechanism of self-healing in the films. The SA and SAKGM films can be completely healed after 240 and 120 s, indicating that konjac-glucomannan (KGM) can enhance the chain mobility, water absorption, and hydrogen bond [1].

Supplementary Movie 1 and **Supplementary Movie 2** have been uploaded to explain more clearly the self-healing process.

Supplementary Note 3

Performance comparison of different self-healable and biodegradable films

Table S1. Self-healing performances of different biodegradable films. Comparison is made between SAKGM film developed in this work and other biodegradable films reported in other previous studies. Several key parameters of films are compared to evaluate their performances, which include material, fabrication method, healing mechanism, healing trigger, and recovery time.

Material	Fabrication method	Healing mechanism	Healing trigger	Recovery time	Ref.
^a CS–CA–C C	Solution casting from aqueous ^e NADES	Hydrogen-bond	Slight pressure	60 s	[2]
^b SA–CS	^f LbL dipping deposition	Electrostatic interaction	Water droplet	80 s	[3]
^c CS–CMC	^f LbL electrostatic deposition	Intrinsic electrostatic	Mist (high RH)	600 s	[4]
^d SAKGM	Single-step casting method	Intrinsic mechanism by hydrogen-bond	Water droplet	120 s	This work

^aCS–CA–CC: chitosan – citric acid – choline-chloride; ^bSA–CS: sodium alginate – chitosan; ^cCS – CMC: chitosan – carboxymethyl-cellulose; ^dSAKGM: sodium alginate – konjac-glucomannan; ^eNADES: natural deep eutectic solvent; ^fLbL: Layer-by-layer.

Table S1 shows that our konjac-glucomannan-blended sodium alginate (SAKGM) film fabricated by a single-step casting method is promising because they have higher performance than other state-of-the-art biodegradable films. The SAKGM film can recover its damage area by its own only within 120 s using water-based trigger. This duration is shorter than recovery times needed by sodium alginate – chitosan (SA-SC) and chitosan – carboxymethyl-cellulose (CS-CMS) films [2], [3], [4]. The chitosan – citric acid – choline-chloride (CS-CA-CC) film demonstrates the shortest recovery time of 60 s because its healing process is triggered or activated by an external pressure (compressive stress) rather than by a passive water stimulus, introducing an additional mechanical aid from the other system. Compared to layer-by-layer (LbL) methods that require several steps for fabrication, the single-step casting method can be an alternative for scale-up in mass production because of its low cost and simplicity.

Supplementary Note 4

Physical and mechanical properties of biodegradable films

Table S2. Physical and mechanical properties of biodegradable films. The fabricated films possess similar properties before (SA and SAKGM) and after scratching and water-assisted healing processes (SA-SH and SAKGM-SH).

Polymer	Thickness (mm)	Tensile strength (MPa)	Elongation at break (%)	Young's modulus (MPa)	WVTR ($\text{g m}^{-2} \text{h}^{-1}$)
SA	(0.15 ± 0.01) ^a	(0.23 ± 0.04) ^a	(78.55 ± 13.79) ^a	(0.30 ± 0.06) ^a	(7.81 ± 0.83) ^a
SA-SH	(0.15 ± 0.01) ^a	(0.22 ± 0.08) ^a	(73.69 ± 15.86) ^a	(0.31 ± 0.13) ^a	(6.56 ± 1.36) ^a
SAKGM	(0.19 ± 0.01) ^a	(0.27 ± 0.08) ^a	(43.53 ± 10.71) ^a	(0.62 ± 0.12) ^a	(6.18 ± 1.25) ^a
SAKGM-SH	(0.20 ± 0.01) ^a	(0.20 ± 0.10) ^a	(34.65 ± 6.28) ^b	(0.60 ± 0.39) ^a	(7.09 ± 0.49) ^a

*Different superscripts (^a, ^b) within the same column indicate significant differences among samples ($p < 0.05$).

Table S2 demonstrates that the intrinsic self-healing mechanism restores both mechanical properties and barrier performance. A reduction in elongation at break (EAB) was observed only for SAKGM-SH film, likely because the addition of KGM reinforces the film matrix and consequently diminishes its extensibility.

Supplementary Note 5

Mechanical characterizations of biodegradable films

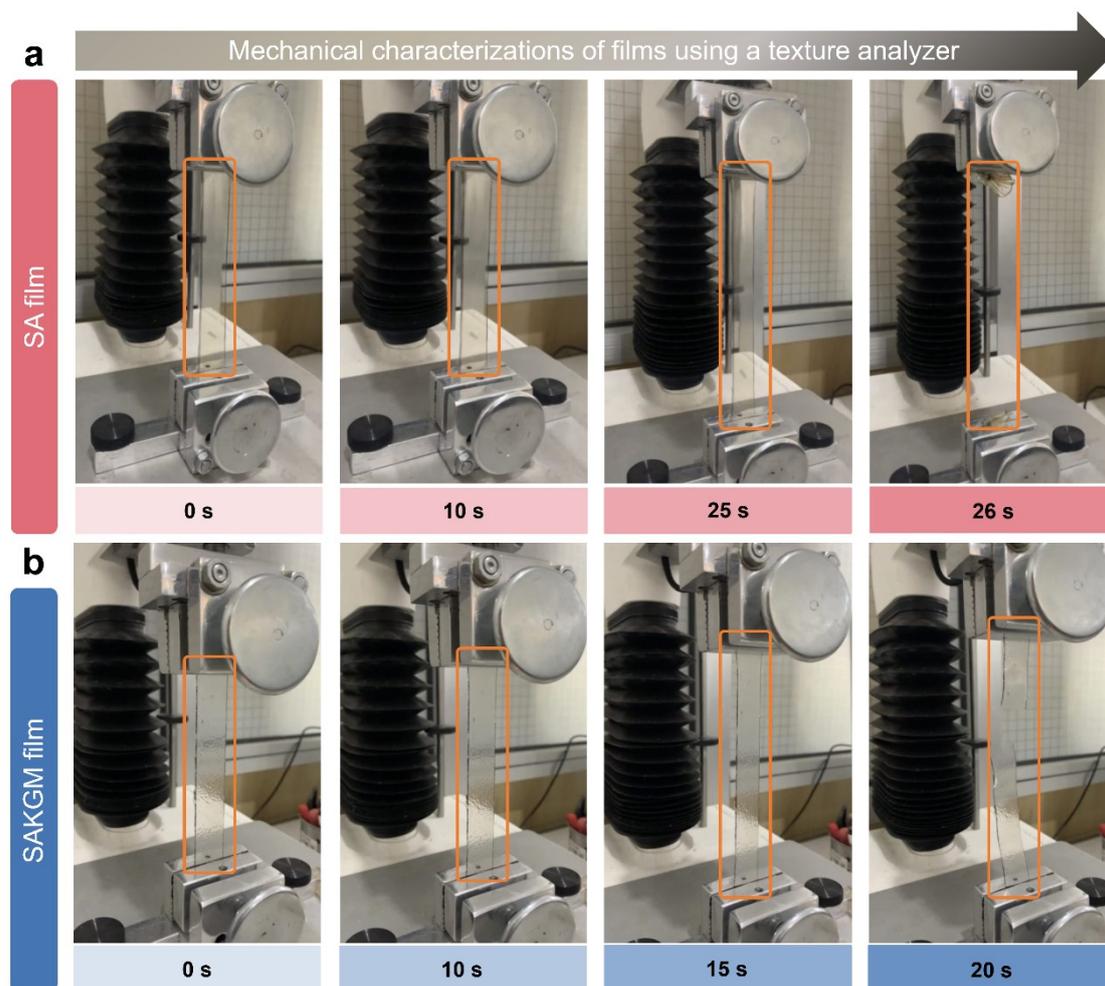


Figure S3. Mechanical characterizations of biodegradable films. **a** Pure sodium alginate (SA) film. **b** Konjac-glucomannan-blended sodium alginate (SAKGM) film. The mechanical measurements of film properties were recorded using a texture analyzer (TA-Xt, Stable Microsystems, UK). The orange rectangle marks the initial gauge section. The test comprises three different stages (i.e., progressive stretching, necking, and final rupture). The SA and SAKGM films were broken after the tests were conducted for ~26 s and ~20 s, respectively. The force displacement data from these sequences were used to calculate tensile strength, elongation at break (EAB), and Young's modulus for each film.

Supplementary Note 6

Biodegradability test of biodegradable films

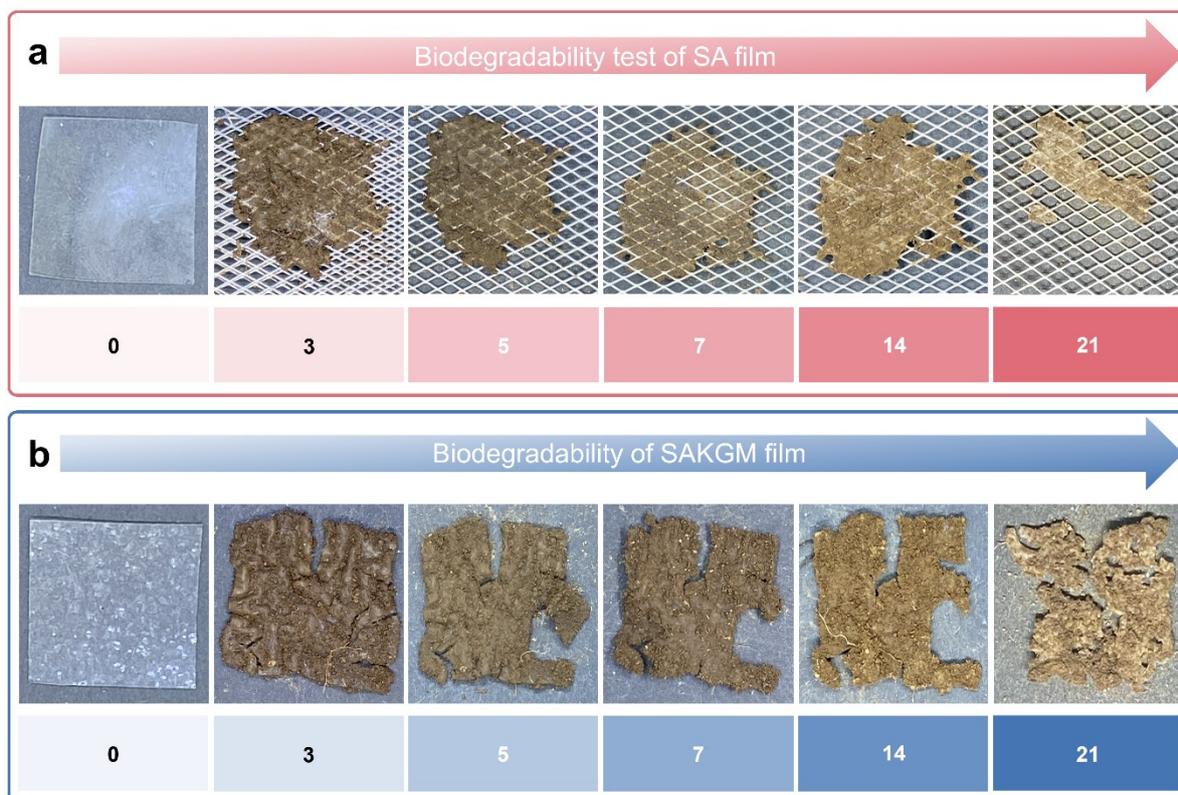


Figure S4. Biodegradability test of self-healable biodegradable film. a Pure sodium alginate (SA) film. **b** Konjac-glucomannan-blended sodium alginate (SAKGM) film. The biodegradability test was conducted over 21-day soil burial test to evaluate the degradation behavior of the films. Visual observation shows progressive structural breakdown and surface deterioration of both SA and SAKGM films over the time, indicating their biodegradability under soil conditions.

Supplementary Note 7

Physicochemical characteristics of grapes after 20 days of storage

Table S3. Physicochemical characteristics of grapes after 20 days of storage. The applications of konjac-glucomannan-blended sodium alginate (SAKGM) films on grapes were conducted using brushing and wrapping methods. Changes in weight loss, firmness, titratable acidity, vitamin C and color difference of samples after 20 days of storage were compared and analyzed.

Treatment	Weight loss (%)		Firmness (N)		Titratable acidity (mg/g)		Vitamin C (mg/g)		Color difference
	5	20	0	20	0	20	0	20	
Uncoated	(11.38 ± 0.53)	*na	(7.00 ± 1.84)	*na	(0.38 ± 0.02)	*na	(2.02 ± 0.44)	*na	(3.73 ± 0.60) ^a
Brushing	(7.31 ± 1.67)	(22.93 ± 1.67)	(6.90 ± 1.25)	(5.51 ± 1.25)	(0.37 ± 0.11)	(0.44 ± 0.11)	(2.27 ± 0.31)	(2.40 ± 0.31)	(2.38 ± 0.60) ^b
Wrapping	(6.53 ± 1.67)	(21.82 ± 1.00)	(8.00 ± 1.25)	(6.12 ± 1.48)	(0.42 ± 0.08)	(0.44 ± 0.08)	(2.24 ± 0.33)	(2.49 ± 0.33)	(2.27 ± 0.60) ^b

*Different superscripts (^a, ^b) within the same column indicate significant differences among samples ($p < 0.05$), *na: the data are not available because the grapes had spoiled after 8 days of storage.

Table S3 demonstrates that physicochemical and nutritional components of grapes after 20 days of storage. There was no significant difference for the samples that were treated using brushing and wrapping methods. This phenomenon indicates that the application of SAKGM treatment can retain the physicochemical and nutritional value of grapes.

References

- [1] L. Guo, W. Yokoyama, L. Chen, F. Liu, M. Chen, and F. Zhong, 'Characterization and physicochemical properties analysis of konjac glucomannan: Implications for structure-properties relationships', *Food Hydrocoll*, vol. 120, Nov. 2021, doi: 10.1016/j.foodhyd.2021.106818.
- [2] M. A. Smirnov *et al.*, 'Self-healing films based on chitosan containing citric acid/choline chloride deep eutectic solvent', *Polym Test*, vol. 97, May 2021, doi: 10.1016/j.polymertesting.2021.107156.
- [3] Y. Du *et al.*, 'Fabrication of novel self-healing edible coating for fruits preservation and its performance maintenance mechanism', *Food Chem*, vol. 351, p. 129284, Jul. 2021, doi: 10.1016/j.foodchem.2021.129284.
- [4] M. Sultan, O. M. Hafez, and M. A. Saleh, 'Quality assessment of lemon (*Citrus aurantifolia*, swingle) coated with self-healed multilayer films based on chitosan/carboxymethyl cellulose under cold storage conditions', *Int J Biol Macromol*, vol. 200, pp. 12–24, Mar. 2022, doi: 10.1016/j.ijbiomac.2021.12.118.

