Supplementary Information

Double Noncovalent Network Chitosan/Hyperbranched Polyethyleneimine/Fe^{3+} Films with High Toughness and Well Antibacterial Activity

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Preparation of CF-1 Films

1 g chitosan powder was dissolved in a 1% acetic acid solution and magnetically stirred for 12 h to make a uniformly solution (100 ml), the prepared Fe^{3+} solution (5 mg/ml) was added to CHP-4 solution. The samples with Fe^{3+} content (1 mg) were named CF-1. Then the matrix was heated and magnetically stirred in a water bath at 60 °C for 1 h and magnetically stirred at room temperature for 12 h. Finally, they were transferred into Petri dishes (Φ = 12 cm) and put in a constant temperature and humidity box at 45 °C and 50% RH until the films were dried.

Table S1. The detailed composition of the chitosan-based films

Sample	Chitosan (g)	HPEI (g)	$FeCl_3$ (mg)
CS	1.0		
CHP-1	1.0	0.05	
CHP-2	1.0	0.10	
CHP-3	1.0	0.15	
CHP-4	1.0	0.20	
CHPF-1	1.0	0.20	0.5
CHPF-2	1.0	0.20	1.0
CHPF-3	1.0	0.20	2.0
CHPF-4	1.0	0.20	3.0
CHPF-5	1.0	0.20	6.0
CF-1	1.0		1.0

Characterization

Thickness

The thickness (mm) of films was measured, using a digital vernier caliper (Deli, China), at three different positions in each sample to the nearest 0.001mm.

Water vapor transmission (WVT) and water vapor permeability (WVP)

The water vapor transmission (WVT) and water vapor permeability (WVP) of films was determined by procedure for water method, using a modified ASTM E96/E96M-16-Standard Test Methods for Water Vapor Transmission of Materials[1, 2]. The obtained dry chitosan-based films were covered on the mouth of the sample bottle and sealed by waterproof tape. The bottle was filled with more than 80% ultrapure water to produce 100% RH. Finally, the sample vial was placed in a desiccator filled with silica gel desiccant in order to provide 0% RH. The weight change of the sample vial was recorded every 6 hours, at room temperature (25 °C). Measure at least three times each time and take the average value to reduce errors. The water vapor permeability was calculated as follows:

$$WVT(gm^{-2}h^{-1}mm) = \frac{\Delta M \times d}{A \times T} \quad (1)$$

$$WVP(gm^{-2}h^{-1}Pa^{-1}mm) = \frac{WVT}{\Delta P} = \frac{\Delta M \times d}{A \times T \times \mathrm{P}(\mathrm{RH}_1\text{-}\mathrm{RH}_2)} \quad (2)$$

Where: ΔM = the weight loss over time (g), d = the thickness of sample (mm),

A = the test area or cup mouth area (m^2), T = the time (h),

ΔP = the partial vapor pressure difference of the atmosphere with silica gel and pure water (Pa),

P = the saturation vapor pressure at test temperature (3.169×10^3 Pa, 25 °C),

RH_1 = the relative humidity at test cup (100% RH), RH_2 = the relative humidity at desiccator (0% RH).

From the results of Table S2, the WVT and WVP of CS film were better than CHP-4 film, it can be illustrated by the addition of HPEI. Large number of amino groups and hydroxyl groups exist in the CHP-4 film, and water vapor can pass through the film more easily. After adding 1 mg of iron ions, the addition of iron ions combines with the amino groups and hydroxyl groups in the internal structure to form metal coordination bonds, resulting in a decrease in the amount of water vapor passing through the film.

Table S2. Thickness, water vapor transmission (WVT) and water vapor permeability (WVP) of chitosan-based films.

Sample code	Thickness (mm)	WVT ($gm^{-2}h^{-1}mm$)	WVP ($gm^{-2}h^{-1} Pa^{-1}mm\times10^{-3}$)
CS	0.153 ± 0.003	6.3666 ± 0.11	2.0090 ± 0.05
CHP-4	0.298 ± 0.005	9.6227 ± 0.32	3.0365 ± 0.13
CHPF-2	0.253 ± 0.006	6.1487 ± 0.10	1.9403 ± 0.04

The results of WVT and WVP are calculated based on the 24-hour weight loss.

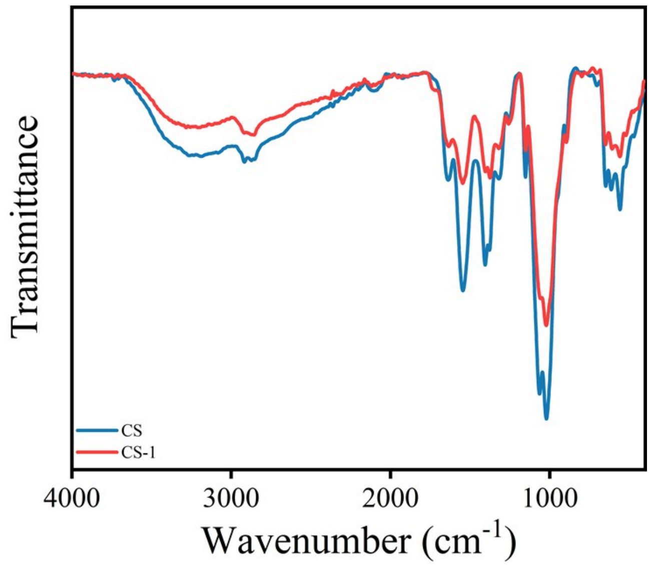

Figure S1. FT-IR spectra of CS and CF-1 films.

1. S. S. Narasagoudr, V. G. Hegde, R. B. Chougale, S. P. Masti, S. Vootla and R. B. Malabadi, *Food Hydrocoll.*, 2020, **109**, 106096.
2. R. Priyadarshi, Sauraj, B. Kumar and Y. S. Negi, *Carbohydr. Polym.*, 2018, **195**, 329-338.