

## APPENDIX A: Preparation, film-forming property and antifungal activity of 28.6% chitosan composite film (CCF)

The CCF was prepared by the following steps.

(1) The film-forming effects of different concentrations of chitosan and dextrin were shown in Table S1. The better film-forming concentrations of chitosan were 6.00, 8.00, 10.00% and dextrin were 10.00, 20.00 and 30.00%.

Table S1. The film-forming effects of different concentrations of chitosan and dextrin

Film materials	Concentration (%)	Ductility	Degree of wire drawing	Extended area (cm <sup>2</sup> )	Dilution property
Water	0.00	0	0	1.32	--
	2.00	+	+	1.04	Easy
	4.00	++	+	1.19	Easy
	6.00	+++	++	1.36	Easy
	8.00	++++	+++	1.45	Easy
	10.00	++++	+++	1.40	Easy
	12.00	++++	+++	1.20	Difficult
Chitosan	10.00	+	+	1.18	Easy
	20.00	++	+	1.27	Easy
	30.00	++	++	1.30	Easy
	40.00	++	++	1.52	Difficult
	50.00	+++	+++	1.90	Difficult
	60.00	+++	+++	1.70	More difficult
Dextrin					

(2) The film-forming effects of different mixture ratios of chitosan-dextrin were shown in Table S2. The 8:10 and 6:20 of chitosan: dextrin had very good film-forming effects.

Table S2. The film-forming effects of different mixture ratios of chitosan-dextrin

Chitosan : Dextrin (%)	Ductility	Degree of wire drawing	Extended area (cm <sup>2</sup> )	Dilution property
0.00	0	0	1.13	--
4:10	++	++	1.26	Easy
6:10	++++	+++	1.33	Easy
8:10	++++	+++	1.47	Easy

4:20	++++	+++	1.29	Easy
6:20	++++	+++	1.49	Easy
8:20	++++	+++	1.58	Difficult
4:30	++++	+++	1.20	Easy
6:30	++++	+++	1.53	Difficult
8:30	++++	+++	1.43	Difficult

(3) The inhibition of chitosan and dextrin to *B. dothidea* and *Phomopsis* sp. were shown in Table S3, two materials had certain antibacterial properties, and chitosan was better than dextrin. The inhibition of different mixture ratios of chitosan-dextrin to *B. dothidea* and *Phomopsis* sp. were shown in Table S4 and Table S5, respectively. Among all the tested mixture ratios, the 8:10 of chitosan: dextrin caused the inhibition of mycelia growth in both *B. dothidea* and *Phomopsis* sp. with mycelial EC<sub>50</sub> values of 430.00 mg L<sup>-1</sup> and 252.32 mg L<sup>-1</sup>, respectively.

Table S3. The inhibition of two film materials to *B. dothidea* and *Phomopsis* sp.

Fungal isolates	Materials	Toxic regression equation	Correlation coefficient (r)	EC <sub>50</sub> (mg L <sup>-1</sup> )	95% confidence region (mg L <sup>-1</sup> )
<i>B. dothidea</i>	Chitosan	y=5.1568+0.7069x	0.9941	600.11	531.59~677.42
	Dextrin	y=4.6559+1.0906x	0.9988	2067.57	1881.53~2272.20
<i>Phomopsis</i> sp.	Chitosan	y=5.2904+0.9250x	0.9865	485.41	402.14~585.77
	Dextrin	y=5.1848+0.7280x	0.9937	557.28	491.20~632.31

Table S4. The inhibition of different mixture ratios of chitosan-dextrin to *B. dothidea*

Chitosan : Dextrin(%)	Toxic regression equation	Correlation coefficient(r)	EC <sub>50</sub> (mg L <sup>-1</sup> )	95% confidence region(mg L <sup>-1</sup> )
4:10	y=5.0975+0.8514x	0.9778	768.21	564.00~1046.41
6:10	y=5.1424+0.9929x	0.9927	718.80	614.22~841.29
8:10	y=5.3454+0.9423x	0.9919	430.00	373.21~495.50
4:20	y=4.5466+0.4442x	0.9878	10484.21	6279.01~17505.49
6:20	y=4.5961+0.6476x	0.9937	4205.10	3215.18~550.00
8:20	y=4.8280+0.8139x	0.9564	1626.69	777.89~3401.71
4:30	y=4.2623+0.6724x	0.9677	12506.69	5548.45~28191.00
6:30	y=4.2844+0.9365x	0.9860	5809.41	3844.78~8.7778.11
8:30	y=4.6391+0.5601x	0.9838	4409.62	3001.89~6477.56

Table S5. The inhibition of different mixture ratios of chitosan-dextrin to *Phomopsis* sp.

Chitosan : Dextrin(%)	Toxic regression equation	Correlation coefficient(r)	EC <sub>50</sub> (mg L <sup>-1</sup> )	95% confidence region(mg L <sup>-1</sup> )
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4:10	$y=5.4524+1.0449x$	0.9985	369.00	347.11~392.33
6:10	$y=5.8436+1.4341x$	0.9888	258.11	214.82~310.14
8:10	$y=5.9211+1.5398x$	0.9912	252.32	215.78~294.84
4:20	$y=5.0372+1.3531x$	0.9965	938.61	855.78~1029.45
6:20	$y=5.0286+0.6983x$	0.9821	910.10	722.11~1147.00
8:20	$y=5.0682+0.7275x$	0.9885	805.83	634.33~1023.78
4:30	$y=4.6309+0.7253x$	0.9981	3228.44	2848.81~3658.45
6:30	$y=4.5874+0.8322x$	0.9941	3131.62	2572.89~3811.56
8:30	$y=4.6747+0.6812x$	0.9882	3002.71	2268.11~3975.35

(4) Firstly, we screened different antifungal substances, among which ferulic acid and natamycinpy had better inhibitory effect to pathogenic fungal (as shown in Table S6, S7). Then, we added them to 8:10 of chitosan composite film to test their inhibitory effect on pathogenic fungal (As shown in Table S8). Finally, considering the cost and safety, we selected ferulic acid as the synergistic substance of 8:10 of chitosan composite film, because it came from plants and was safe.

Table S6. The inhibition of different antifungal materials to *B. dothidea*

Antifungal materials	Toxic regression equation	Correlation coefficient(r)	EC <sub>50</sub> (mg L <sup>-1</sup> )	95% confidence region (mg L <sup>-1</sup> )
Natamycinpty	$y=7.5976+1.6267x$	0.9838	20.71	20.77~31.00
Ferulic acid	$y=11.0033+4.5315x$	0.9881	47.32	31.22~71.78
Antifungal peptide	$y=6.0308+1.0580x$	0.9809	106.13	85.12~132.33
Lysozyme	$y=5.4778+1.2953x$	0.9938	427.74	377.22~485.00
Glycine	$y=5.2978+1.0409x$	0.9952	517.42	463.89~577.11
Tea polyphenols	$y=5.1876+0.7540x$	0.9724	563.89	328.56~967.66
Citric acid		less effective		

Table S7. The inhibition of different antifungal materials to *Phomopsis* sp.

Antifungall materials	Toxic regression equation	Correlation coefficient(r)	EC <sub>50</sub> (mg L <sup>-1</sup> )	95% confidence region (mg L <sup>-1</sup> )
Natamycinpty	$y=7.2311+1.3893x$	0.9960	24.80	22.44~27.38
Ferulic acid	$y=6.8553+1.1438x$	0.9702	23.91	14.56~39.00
Antifungal peptide	$y=5.7350+0.9670x$	0.9979	173.83	160.11~188.56
Lysozyme	$y=6.0452+0.8915x$	0.9833	67.22	53.00~85.22
Glycine	$y=5.4886+0.8915x$	0.9833	283.00	223.13~358.18
Tea polyphenols		less effective		
Citric acid		less effective		

Table S8. The inhibition of two 19% film (Natamycinpty or Ferulic acid+8:10) to *B. dothidea* and *Phomopsis* sp.

Fungal isolates	Antimicrobial materials	Toxic regression equation	Correlation coefficient(r)	EC <sub>50</sub> (mg L <sup>-1</sup> )	95% confidence region (mg L <sup>-1</sup> )
<i>B. dothidea</i>	Natamycinpty	y=9.4433+5.8405x	0.9969	173.52	110.82~278.23
	Ferulic acid	y=6.0232+0.9754x	0.9775	89.31	68.93~115.67
<i>Phomopsis</i> sp.	Natamycinpty	y=6.0452+0.8915x	0.9833	67.20	53.00~85.22
	Ferulic acid	y=5.9994+0.9202x	0.9942	82.00	72.30~93.12

(5) We added calcium nitrate to the 19% chitosan composite film to test their inhibitory effect on pathogenic fungal (As shown in Table S9), 24% chitosan composite film caused the inhibition of mycelia growth in both *B. dothidea* and *Phomopsis* sp. with mycelial EC<sub>50</sub> values of 69.00 mg L<sup>-1</sup> and 50.41 mg L<sup>-1</sup>, respectively.

Table S9. The inhibition of two different film to *B. dothidea* and *Phomopsis* sp.

Fungal isolates	Films	Toxic regression equation	Correlatin coefficient(r)	EC <sub>50</sub> (mg L <sup>-1</sup> )	95% confidence region(mg L <sup>-1</sup> )
<i>B. dothidea</i>	24% (8:10+1+5)	y=6.0227+0.8807x	0.9995	69.00	66.56~71.5
	19% (8:10+1)	y=6.2927+1.2456x	0.9768	91.71	70.13~119.89
	18% (8:10)	y=5.1920+0.8183x	0.9954	582.62	521.78~650.45
<i>Phomopsis</i> sp.	24% (8:10+1+5)	y=5.8745+0.6742x	0.9880	50.41	42.33~60.12
	19% (8:10+1)	y=6.3408+1.1184x	0.9938	63.32	55.56~72.00
	18% (8:10)	y=5.4207+0.9403x	0.9872	356.90	296.78~429.34

Note: 24% (8% chitosan +10% dextrin +1% ferulic acid +5% calcium nitrate); 19% (8% chitosan +10% dextrin +1% ferulic acid); 18% (8% chitosan +10% dextrin).

(6) For the selection of auxiliaries, we screened out 3% glycerol, 1% organosilicon and 0.6% sodium benzoate with better results (as shown in Table S10, S11, S12). The 28.6% CCF (8% chitosan +10% dextrin +1% ferulic acid +5% calcium nitrate+5% glycerol+1% organosilicon+0.6% sodium benzoate) was finally prepared.

Table S10. The film-forming effects of different content glycol and glycerol to chitosan-dextrin film

Added concentration (%)			Cold storage effect	Film-forming effects
Film	Glycol	Glycerol		
24% CCF (8% chitosan +10% dextrin +1% ferulic acid +5% calcium nitrate)	1	-	Ivory, coagulated, unqualified	Bubbles increased and larger
	2	-	Ivory, semi coagulated, unqualified	Viscosity and the bubbles increased
	3	-	Ivory, gelatinous, restored at room temperature, basically qualified	Viscosity and the bubbles increased
	-	1	Ivory, gelatinous, restored at room temperature, basically qualified	No bubbles

-	2	Ivory, colloidal liquid, qualified	No bubbles
-	3	Ivory, colloidal liquid, qualified	Viscosity increased and no bubbles

Table S11. The film-forming effects of different content organosilicon to chitosan-dextrin film

Added concentration (%)		Film-forming effects
Film	Organosilicon	
27% CCF	0.5	Similar to control
(8% chitosan +10% dextrin +1% ferulic acid +5% calcium nitrate+3% glycerol)	1.0	Ductility increased
	1.5	Viscosity decreased
	2.0	Viscosity obviously decreased

Table S12. The storage period of different content sodium benzoate and salicylic acid to chitosan-dextrin film

Added concentration			Storage quality
Film	Sodium benzoate	Salicylic acid	
28% CCF	0.2	-	No fungal growth, slight change in smell after 60 d
(8% chitosan +10% dextrin +1% ferulic acid +5% calcium nitrate+5% glycerol+1% organosilicon)	0.4	-	No fungal growth, slight change in smell after 60 d
	0.6	-	No fungal growth, no change in smell after 60 d
	0.8	-	No fungal growth, no change in smell after 60 d
	-	0.2	No fungal growth, slight change in smell after 60 d
organosilicon)	-	0.4	No fungal growth, slight change in smell after 60 d
	-	0.6	No fungal growth, slight change in smell after 60 d
	-	0.8	No fungal growth, no change in smell after 60 d
	CK		Fungal growth, change in smell after 60 d

(7) In summary, through the selection of auxiliaries and the addition of calcium nitrate, the 28.6% CCF was finally prepared. The effects of CCFs on the mycelial growth of pathogenic fungi are shown in Table S13. 28.6% CCF had strong inhibitory effect to mycelia growth of *B. dothidea* and *Phomopsis* sp. with mycelial EC<sub>50</sub> values of 68.11 mg L<sup>-1</sup> and 50.34 mg L<sup>-1</sup>, respectively.

Table S13. The inhibition of different CCFs to mycelial growth of *Botryosphaeria dothidea* and *Phomopsis* sp.

Fungal isolates	Tested pharmacy	Toxic regression equation	Correlatin coefficient(r)	EC <sub>50</sub> (mg L <sup>-1</sup> )	95% confidence region (mg L <sup>-1</sup> )
<i>B. dothidea</i>	28.6% CCF	y=6.0206+0.8746x	0.9990	68.11	64.67~71.56
	24% CCF	y=6.0213+0.8795x	0.9993	69.00	65.78~72.32
	19% CCF	y=6.3511+1.2645x	0.9901	85.40	72.20~101.00
	18% CCF	y=5.1942+0.8207x	0.9960	579.89	523.14~643.00
<i>Phomopsis</i> sp.	28.6% CCF	y=5.8783+0.6764x	0.9878	50.34	42.13~60.10
	24% CCF	y=5.7794+0.6363x	0.9913	59.52	50.67~69.89
	19% CCF	y=5.9323+1.1281x	0.9949	149.11	127.78~174.13
	18% CCF	y=5.4267+0.9425x	0.9882	352.45	295.24~421.00