

# Physico-chemical, antimicrobial and antioxidant properties of gelatin-chitosan based films loaded with nanoemulsions encapsulating active compounds

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DOI:

[10.1016/j.foodhyd.2017.12.012](https://doi.org/10.1016/j.foodhyd.2017.12.012)

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*Document Version*

Peer reviewed version

*Citation for published version (Harvard):*

Pérez-Córdoba, LJ, Norton, IT, Batchelor, HK, Gkatzionis, K, Spyropoulos, F & Sobral, PJA 2017, 'Physico-chemical, antimicrobial and antioxidant properties of gelatin-chitosan based films loaded with nanoemulsions encapsulating active compounds', *Food Hydrocolloids*. <https://doi.org/10.1016/j.foodhyd.2017.12.012>

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Checked for eligibility: 08/01/2018

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**Table 1.** Encapsulation efficiencies of cinnamaldehyde,  $\alpha$ -tocopherol and garlic oil and mean droplet sizes, polydispersity indices (PDI),  $\zeta$ -potential and pH values of the O/W nanoemulsions containing these encapsulated active compounds, as a function of storage time (all systems stored at  $4\pm 1^\circ\text{C}$ ).

Samples*	Time (Days)	Encapsulation efficiency			Droplet size (nm)	PDI	$\zeta$ -potential (mV)	pH
		Cinnamaldehyde (%)	$\alpha$ -tocopherol(%)	GO(%)				
Control ( $N_1$ )	0	----	---	----	$157.0 \pm 4.1^{\text{aA}}$	$0.19 \pm 0.02^{\text{bA}}$	$-17.3 \pm 0.6^{\text{aA}}$	$6.1 \pm 0.0^{\text{abA}}$
	30	----	---	----	$158.2 \pm 2.9^{\text{aA}}$	$0.20 \pm 0.02^{\text{bA}}$	$-16.0 \pm 1.4^{\text{bA}}$	$6.0 \pm 0.0^{\text{bcA}}$
	60	----	---	----	$161.2 \pm 5.9^{\text{aA}}$	$0.23 \pm 0.02^{\text{aA}}$	$-17.2 \pm 0.9^{\text{abA}}$	$6.1 \pm 0.0^{\text{aA}}$
	90	----	---	----	$156.9 \pm 4.7^{\text{aA}}$	$0.21 \pm 0.02^{\text{bA}}$	$-18.1 \pm 1.0^{\text{aA}}$	$6.0 \pm 0.0^{\text{cA}}$
$\alpha$ -t/Cin ( $N_2$ )	0	$100 \pm 0.0^{\text{aA}}$	$57.1 \pm 1.1^{\text{aA}}$	----	$123.1 \pm 1.5^{\text{aB}}$	$0.16 \pm 0.01^{\text{aB}}$	$-14.2 \pm 0.5^{\text{aC}}$	$5.3 \pm 0.0^{\text{aD}}$
	30	----	---	----	$121.5 \pm 0.6^{\text{aC}}$	$0.16 \pm 0.04^{\text{aB}}$	$-12.3 \pm 0.3^{\text{bC}}$	$5.1 \pm 0.0^{\text{bC}}$
	60	----	---	----	$121.9 \pm 2.7^{\text{aC}}$	$0.15 \pm 0.01^{\text{aB}}$	$-13.7 \pm 0.9^{\text{aC}}$	$4.7 \pm 0.1^{\text{cC}}$
	90	$99.7 \pm 0.3^{\text{aA}}$	$46.6 \pm 0.4^{\text{bB}}$	----	$122.4 \pm 2.5^{\text{aC}}$	$0.15 \pm 0.01^{\text{aBC}}$	$-13.7 \pm 1.0^{\text{aC}}$	$4.6 \pm 0.0^{\text{dB}}$
$\alpha$ -t/GO ( $N_3$ )	0	----	$52.4 \pm 0.4^{\text{aA}}$	$92.2 \pm 1.9^{\text{aA}}$	$111.0 \pm 2.0^{\text{aC}}$	$0.16 \pm 0.0^{\text{aB}}$	$-15.9 \pm 0.7^{\text{abB}}$	$6.0 \pm 0.0^{\text{aB}}$
	30	----	---	---	$112.5 \pm 2.4^{\text{aD}}$	$0.14 \pm 0.02^{\text{aB}}$	$-15.5 \pm 0.6^{\text{abA}}$	$5.4 \pm 0.0^{\text{bB}}$
	60	----	---	---	$111.2 \pm 1.9^{\text{aD}}$	$0.14 \pm 0.01^{\text{aB}}$	$-16.0 \pm 0.8^{\text{aB}}$	$4.9 \pm 0.0^{\text{cB}}$
	90	----	$47.8 \pm 0.2^{\text{bA}}$	$88.5 \pm 2.7^{\text{aA}}$	$111.5 \pm 1.9^{\text{aD}}$	$0.14 \pm 0.01^{\text{aC}}$	$-15.0 \pm 0.9^{\text{bB}}$	$4.3 \pm 0.0^{\text{dD}}$
$\alpha$ -t/Cin and GO ( $N_4$ )	0	$93.9 \pm 2.6^{\text{aA}}$	$56.7 \pm 3.1^{\text{aA}}$	$70.9 \pm 2.0^{\text{aB}}$	$124.8 \pm 1.4^{\text{bB}}$	$0.15 \pm 0.01^{\text{bB}}$	$-14.4 \pm 0.7^{\text{aC}}$	$5.5 \pm 0.0^{\text{aC}}$
	30	---	---	---	$126.4 \pm 2.2^{\text{bB}}$	$0.16 \pm 0.02^{\text{bB}}$	$-13.7 \pm 0.4^{\text{bB}}$	$4.9 \pm 0.0^{\text{bD}}$
	60	---	---	---	$130.0 \pm 1.1^{\text{aB}}$	$0.20 \pm 0.01^{\text{aC}}$	$-13.8 \pm 0.5^{\text{abC}}$	$4.7 \pm 0.0^{\text{cC}}$
	90	$89.6 \pm 0.6^{\text{aB}}$	$45.2 \pm 0.3^{\text{bC}}$	$61.6 \pm 0.1^{\text{bB}}$	$126.6 \pm 3.0^{\text{bB}}$	$0.16 \pm 0.02^{\text{bB}}$	$-14.5 \pm 0.6^{\text{aBC}}$	$4.5 \pm 0.0^{\text{dC}}$

Mean values  $\pm$  standard deviation (n = 3). Different lower case letters in the same column indicate significant differences ( $p < 0.05$ ) for the same sample over different days and different capital letters indicate significant differences ( $p < 0.05$ ) among different samples measured at the same time interval (day).

Cin: cinnamaldehyde;  $\alpha$ -t:  $\alpha$ -tocopherol; GO: garlic oil.

\* $N_1$ : Control nanoemulsion (no encapsulated species);  $N_2$ :  $\alpha$ -tocopherol/cinnamaldehyde;  $N_3$ :  $\alpha$ -tocopherol/garlic oil;  $N_4$ :  $\alpha$ -tocopherol/cinnamaldehyde and garlic oil-loaded nanoemulsion.

**Table 2.** Physical/Mechanical properties of gelatin-chitosan films loaded with O/W nanoemulsions containing encapsulated active compounds.

<b>Sample *</b>	<b>Thickness (mm)</b>	<b>Moisture content (%)</b>	<b>Solubility in water (%)</b>	<b>Swelling (g/g)</b>	<b>TS (MPa)</b>	<b>EB (%)</b>	<b>EM (MPa)</b>
<i>Films N<sub>0</sub></i>	0.082 ± 0.001 <sup>a</sup>	18.2 ± 0.8 <sup>a</sup>	50.8 ± 0.7 <sup>a</sup>	26.9 ± 2.8 <sup>b</sup>	19.0 ± 2.1 <sup>a</sup>	89.1 ± 6.4 <sup>d</sup>	101.4 ± 4.5 <sup>a</sup>
<i>Films N<sub>1</sub></i>	0.081 ± 0.002 <sup>a</sup>	17.8 ± 1.8 <sup>a</sup>	47.5 ± 0.6 <sup>b</sup>	30.6 ± 0.6 <sup>a</sup>	10.0 ± 1.1 <sup>bc</sup>	123.3 ± 1.3 <sup>a</sup>	29.0 ± 3.6 <sup>c</sup>
<i>Films N<sub>2</sub></i>	0.081 ± 0.002 <sup>a</sup>	18.2 ± 1.8 <sup>a</sup>	44.0 ± 2.1 <sup>c</sup>	27.4 ± 2.5 <sup>ab</sup>	11.4 ± 1.0 <sup>b</sup>	108.7 ± 2.2 <sup>c</sup>	37.3 ± 2.5 <sup>b</sup>
<i>Films N<sub>3</sub></i>	0.081 ± 0.002 <sup>a</sup>	17.3 ± 2.1 <sup>a</sup>	43.1 ± 2.3 <sup>c</sup>	30.3 ± 1.5 <sup>a</sup>	8.9 ± 0.9 <sup>c</sup>	111.7 ± 4.8 <sup>bc</sup>	30.1 ± 4.9 <sup>c</sup>
<i>Films N<sub>4</sub></i>	0.082 ± 0.001 <sup>a</sup>	18.1 ± 2.0 <sup>a</sup>	48.9 ± 0.9 <sup>ab</sup>	25.3 ± 0.8 <sup>b</sup>	9.8 ± 3.7 <sup>bc</sup>	113.2 ± 2.1 <sup>b</sup>	39.2 ± 3.6 <sup>b</sup>

Mean values ± standard deviation (n = 3). Different letters in the same column indicate significant differences (p<0.05).

\* *N<sub>0</sub>*- Control 1: film without nanoemulsion; *N<sub>1</sub>*- Control 2: film with control nanoemulsion (no encapsulated species); *N<sub>2</sub>*:  $\alpha$ -tocopherol/cinnamaldehyde; *N<sub>3</sub>*:  $\alpha$ -tocopherol/garlic oil; *N<sub>4</sub>*:  $\alpha$ -tocopherol/cinnamaldehyde and garlic oil-loaded nanoemulsion.

TS: Tensile strength; EB: Elongation at break; EM: Elastic modulus.

**Table 3.** Light transmittance (%) and transparency of gelatin-chitosan films loaded with O/W nanoemulsions containing encapsulated active compounds.

Sample*	Light transmittance (%)						Transparency value
	Wavelength (nm)						
	250	280	350	450	600	800	
<i>Films N<sub>0</sub></i>	22.2 ± 0.3 <sup>a</sup>	23.9 ± 0.6 <sup>a</sup>	80.0 ± 0.4 <sup>a</sup>	90.9 ± 0.2 <sup>a</sup>	94.7 ± 0.2 <sup>a</sup>	97.6 ± 0.2 <sup>a</sup>	0.29 ± 0.01 <sup>c</sup>
<i>Films N<sub>1</sub></i>	5.0 ± 0.7 <sup>b</sup>	9.6 ± 0.3 <sup>b</sup>	66.3 ± 0.7 <sup>bc</sup>	77.6 ± 0.5 <sup>d</sup>	83.7 ± 0.4 <sup>d</sup>	88.8 ± 0.4 <sup>e</sup>	0.96 ± 0.02 <sup>a</sup>
<i>Films N<sub>2</sub></i>	0.0 ± 0.0 <sup>c</sup>	0.1 ± 0.0 <sup>d</sup>	63.7 ± 1.4 <sup>d</sup>	85.7 ± 0.6 <sup>b</sup>	91.4 ± 0.4 <sup>b</sup>	95.4 ± 0.4 <sup>b</sup>	0.48 ± 0.02 <sup>d</sup>
<i>Films N<sub>3</sub></i>	0.1 ± 0.0 <sup>c</sup>	0.7 ± 0.0 <sup>c</sup>	67.5 ± 1.7 <sup>b</sup>	83.8 ± 1.7 <sup>c</sup>	87.7 ± 1.4 <sup>c</sup>	91.2 ± 1.4 <sup>d</sup>	0.71 ± 0.09 <sup>b</sup>
<i>Films N<sub>4</sub></i>	0.0 ± 0.0 <sup>c</sup>	0.1 ± 0.0 <sup>d</sup>	65.0 ± 1.0 <sup>cd</sup>	84.5 ± 0.6 <sup>bc</sup>	88.9 ± 0.5 <sup>c</sup>	92.5 ± 0.6 <sup>c</sup>	0.63 ± 0.03 <sup>c</sup>

Mean values ± standard deviation (n = 3). Different letters in the same column indicate significant differences (p<0.05).

\* *N<sub>0</sub>*- Control 1: film without nanoemulsion; *N<sub>1</sub>*- Control 2: film with control nanoemulsion (no encapsulated species); *N<sub>2</sub>*:  $\alpha$ -tocopherol/cinnamaldehyde; *N<sub>3</sub>*:  $\alpha$ -tocopherol/garlic oil; *N<sub>4</sub>*:  $\alpha$ -tocopherol/cinnamaldehyde and garlic oil-loaded nanoemulsion.

**Table 4.** Thermal properties and roughness characteristics of gelatin-chitosan films loaded with O/W nanoemulsions containing encapsulated active compounds.

Sample*	1 <sup>st</sup> Scan			2 <sup>nd</sup> Scan	Roughness	
	T <sub>g</sub> (°C)	ΔH <sub>g</sub> (J/g)	T <sub>m</sub> (°C)	T <sub>g</sub> (°C)	R <sub>a</sub> (nm)	R <sub>q</sub> (nm)
Films N <sub>0</sub>	45.6 ± 0.6 <sup>a</sup>	12.1 ± 0.8 <sup>b</sup>	54.9 ± 0.8 <sup>a</sup>	8.6 ± 2.2 <sup>ab</sup>	7.5	11.1
Films N <sub>1</sub>	46.2 ± 0.8 <sup>a</sup>	9.3 ± 0.9 <sup>a</sup>	53.5 ± 0.2 <sup>a</sup>	12.1 ± 1.1 <sup>a</sup>	44.1	58.6
Films N <sub>2</sub>	45.5 ± 0.4 <sup>a</sup>	9.0 ± 0.2 <sup>a</sup>	54.4 ± 0.2 <sup>a</sup>	8.2 ± 2.7 <sup>ab</sup>	31.4	40.5
Films N <sub>3</sub>	46.6 ± 2.3 <sup>a</sup>	9.6 ± 0.8 <sup>a</sup>	56.6 ± 5.2 <sup>a</sup>	10.4 ± 3.1 <sup>ab</sup>	39.5	53.9
Films N <sub>4</sub>	45.6 ± 0.4 <sup>a</sup>	9.3 ± 0.5 <sup>a</sup>	54.6 ± 0.2 <sup>a</sup>	6.7 ± 1.4 <sup>b</sup>	32.3	42.6

Mean values ± standard deviation (n = 3). Different letters in the same column indicate significant differences (p<0.05).

\* N<sub>0</sub>- Control 1: film without nanoemulsion; N<sub>1</sub>- Control 2: film with control nanoemulsion (no encapsulated species); N<sub>2</sub>: α-tocopherol/cinnamaldehyde; N<sub>3</sub>: α-tocopherol/garlic oil; N<sub>4</sub>: α-tocopherol/cinnamaldehyde and garlic oil-loaded nanoemulsion.

T<sub>g</sub>: Glass transition temperature; T<sub>m</sub>: Melting temperature; ΔH<sub>g</sub>: Melting enthalpy; R<sub>a</sub>: average roughness; R<sub>q</sub>: root-mean-square roughness.

**Table 5.** Inhibition halos against *P. aeruginosa* and *L. monocytogenes* and Trolox Equivalent Antioxidant Capacity (TEAC) of gelatin-chitosan films loaded with O/W nanoemulsions containing encapsulated active compounds.

Sample*	Zone of inhibition (mm <sup>2</sup> )		TEAC ( $\mu\text{mol TE/g dried film}$ )		
	<i>P. aeruginosa</i>	<i>L. monocytogenes</i>	DPPH* method	ABTS*+ method	FRAP assay
<i>Films N<sub>0</sub></i>	0.0 $\pm$ 0.0 <sup>c</sup>	0.0 $\pm$ 0.0 <sup>a</sup>	0.0 $\pm$ 0.0 <sup>c</sup>	0.0 $\pm$ 0.0 <sup>d</sup>	6.9 $\pm$ 0.4 <sup>e</sup>
<i>Films N<sub>1</sub></i>	0.0 $\pm$ 0.0 <sup>c</sup>	0.0 $\pm$ 0.0 <sup>a</sup>	0.0 $\pm$ 0.0 <sup>c</sup>	1.3 $\pm$ 0.0 <sup>c</sup>	39.8 $\pm$ 0.2 <sup>d</sup>
<i>Films N<sub>2</sub></i>	138.2 $\pm$ 2.4 <sup>a</sup>	0.0 $\pm$ 0.0 <sup>a</sup>	0.2 $\pm$ 0.0 <sup>a</sup>	2.6 $\pm$ 0.1 <sup>a</sup>	49.9 $\pm$ 1.2 <sup>c</sup>
<i>Films N<sub>3</sub></i>	138.2 $\pm$ 0.0 <sup>a</sup>	0.0 $\pm$ 0.0 <sup>a</sup>	0.1 $\pm$ 0.0 <sup>b</sup>	2.5 $\pm$ 0.0 <sup>a</sup>	81.5 $\pm$ 2.2 <sup>a</sup>
<i>Films N<sub>4</sub></i>	65.4 $\pm$ 1.4 <sup>b</sup>	0.0 $\pm$ 0.0 <sup>a</sup>	0.1 $\pm$ 0.0 <sup>b</sup>	2.3 $\pm$ 0.1 <sup>b</sup>	68.3 $\pm$ 2.9 <sup>b</sup>

Mean values  $\pm$  standard deviation (n = 3). Different letters in the same column indicate significant differences (p<0.05).

\* *N<sub>0</sub>*- Control 1: film without nanoemulsion; *N<sub>1</sub>*- Control 2: film with control nanoemulsion (no encapsulated species); *N<sub>2</sub>*:  $\alpha$ -tocopherol/cinnamaldehyde; *N<sub>3</sub>*:  $\alpha$ -tocopherol/garlic oil; *N<sub>4</sub>*:  $\alpha$ -tocopherol/cinnamaldehyde and garlic oil-loaded nanoemulsion.