

**Biodegradable films based on alfalfa cellulosic residue and carrageenan
blends for sustainable food packaging**

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**Supplementary file
Sustainable Food Technology**

Equations

Tensile strength (TS)

$$TS \text{ (MPa)} = \frac{\text{Force used to break film (N)}}{\text{Film's surface area (mm}^2\text{)}} \dots \text{(S1)}$$

Elongation at break (EB)

$$EB \text{ (\%)} = \frac{\text{Elongation at break (mm)}}{\text{Initial length (mm)}} \times 100 \dots \text{(S2)}$$

Water vapor permeability (WVP)

$$WVP = \frac{WVTR}{P (R_1 - R_2)} \times t \dots \text{(S3)}$$

wherein,

P: saturation vapor pressure of water (Pa) at 25 °C,

R₁: RH in the desiccator,

R₂: RH in the cup,

t: film thickness (m).

The driving force [$P (R_1 - R_2)$] in the experimental condition was calculated as 3073.93 Pa.

Color

$$WI = 100 - [(100 - L^*)^2 + a^{*2} + b^{*2}]^{0.5} \dots \text{(S4)}$$

$$YI = 142.86 \frac{b^*}{L^*} \dots \text{(S5)}$$

$$TCD = [(L^* - L_b^*)^2 + (a^* - a_b^*)^2 + (b^* - b_b^*)^2]^{0.5} \dots \text{(S6)}$$

Wherein,

L*, a*, and b*: Film's color in the Hunter scale

L_b*, a_b*, and b_b*: Background color in the Hunter scale

WI: Whiteness index

YI: Yellowness index

TCD: Total color difference

Transparency and absorption coefficient

$$Transparency = \frac{\log (T_{600})}{t} \dots (S7)$$

$$\alpha = \frac{1}{t} \ln \left(\frac{1}{T\%} \right) \dots (S8)$$

Wherein,

T₆₀₀: transmittance at 600 nm (%)

t: thickness of the film (mm)

α: absorption coefficient

T%: transmittance

Moisture content

$$Moisture (\%) = \left(\frac{Initial\ weight - Weight\ after\ drying}{Initial\ weight} \right) \times 100 \dots (S9)$$

Water solubility

$$Water\ solubility (\%) = \frac{(Initial\ dried\ weight - Final\ dried\ weight)}{Initial\ dried\ weight} \times 100 \dots (S10)$$

Water absorption

$$\text{Water absorption (\%)} = \frac{\text{Water absorbed weight} - \text{Initial dried weight}}{\text{Initial dried weight}} \times 100 \quad \dots(\text{S11})$$

$$\frac{t}{m_t} = K_2 t + K_1 \quad \dots(\text{S12})$$

$$mr_t = a + \frac{b \times t}{c \times t + 1} \dots(\text{S13})$$

$$mr_t = a + b \times \left(1 - \frac{1}{1 + b \times c \times t}\right) \dots(\text{S14})$$

$$mr_t = a + \frac{b \times t}{c + t} \dots(\text{S15})$$

$$mr_t = a \times t^m \quad \dots(\text{S16})$$

$$\frac{mr_t / mr_\infty}{t^{1/2}} = K_1 t^{1/2} + K_2 \quad \dots(\text{S17})$$

$$mr_t = a \times \exp\left[-\frac{b}{(1+t)^\alpha}\right] \dots(\text{S18})$$

$$mr_t = 1 - \exp\left[\left(-\frac{t}{a}\right)^b\right] \dots(\text{S19})$$

$$mr_t = m_e \times (m_o - m_e) \exp\left(-t/\beta\right)^\alpha \dots(\text{S20})$$

$$R^2 = 1 - \frac{\sum (y_i - \hat{y})^2}{\sum (y_i - \bar{y})^2} \quad \dots(\text{S21})$$

$$RMSE = \sqrt{\frac{\sum (y_i - \hat{y})^2}{N}} \quad \dots(\text{S22})$$

Herein,

Equations S12-S20 correspond to Peleg, Singh, Gornicki, Pilosof, Czel, and Czigany, Peppas, Vega-Galvez, Garcia-Pascual, and Weibull models, respectively.

m_t :	moisture content (%) at time 't'
mr_t :	moisture ratio at time 't'
mr_∞ and m_e :	moisture ratio at 120 min
m :	slope of curve fitting in Czel and Czigany model
K_1 and K_2 :	intercept and slope of curve fitting in Peleg model, respectively
K_2 and K_1 :	intercept and slope of curve fitting in Peppas model, respectively
a, b, c, α , and β :	constants ($\alpha=0.9$, a fixed value for the Vega-Galvez's model only).
R^2 :	coefficient of determination
RMSE:	root mean square error

Antioxidant activity

$$\%Radical\ scavenging = \frac{(Absorbance\ of\ control - Absorbance\ of\ sample)}{Absorbance\ of\ control} \times 100\% \quad \dots (S23)$$

Biodegradation

$$\%biodegradation = \frac{Loss\ in\ film\ weight \times 100}{Initial\ weight} \quad \dots (S24)$$

$$Ln(y) = mx + c \quad \dots (S25)$$

$$Ln(y) = a + bx + cx^2 \quad \dots (S26)$$

Wherein,

$Ln(y)$:	natural logarithm of %biodegradation
x :	biodegradation days
m and c :	slope and intercept, respectively, in Equation S25
a, b , and c :	constants in Equation S26.

Figure and Tables

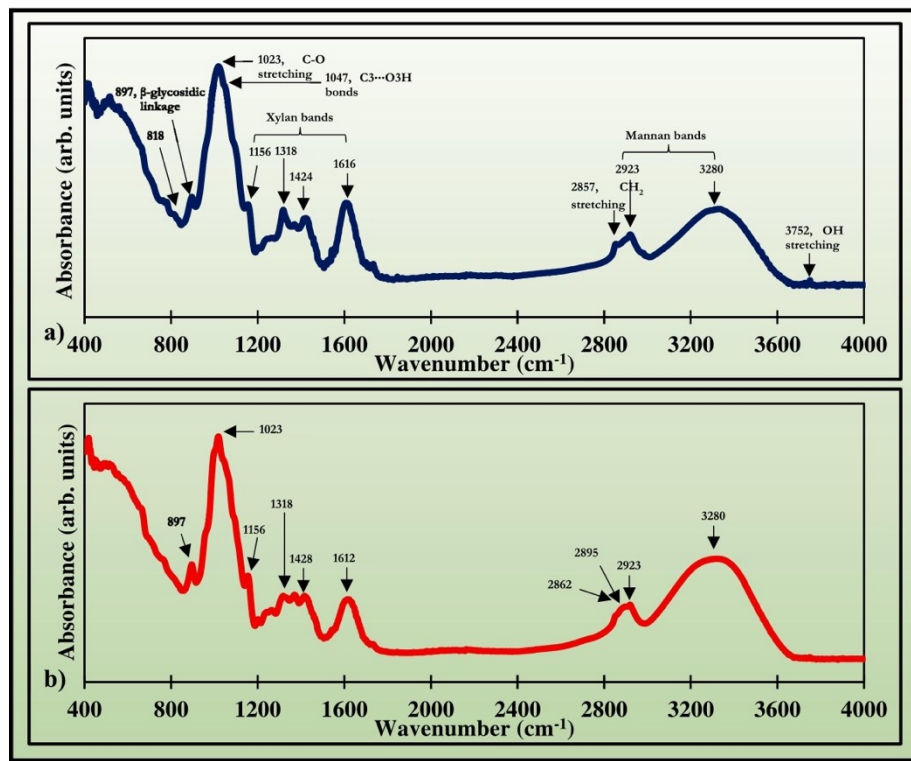


Fig. S1. FTIR spectra of a) alfalfa cellulosic residue and b) film. The functional groups are the same for both graphs if the peak exists. After the cellulosic solution preparation and subsequent film formation, some peaks have shifted, and a few have disappeared. These peaks are also correlated to Fig. 4.

Table S1. Water absorption kinetics model fittings of ACR and ACR-carrageenan films.

Model		ACR	A-t0.5%	A-t1.0%	A-t1.5%	A-κ0.5%	A-κ1.0%	A-κ1.5%	A-λ0.5%	A-λ1.0%	A-λ1.5%
Peleg $t/m_t=K_1+K_2 \times t$	K_1	0.0093	0.0084	0.0080	0.0077	0.0098	0.0069	0.0084	0.0075	0.0056	0.0091
	K_2	0.0066	0.0063	0.0062	0.0062	0.0064	0.0063	0.0060	0.0062	0.0060	0.0057
	R^2	0.9998	0.9999	0.9999	0.9999	0.9998	0.9999	0.9997	0.9998	0.9999	0.9998
	RMSE	0.0042	0.0020	0.0017	0.0021	0.0033	0.0017	0.0042	0.0038	0.0023	0.0035
Singh $m_r=a+(b/(ct+1))$	a	1.3036	1.2972	1.3109	1.3232	1.3058	1.009	1.3749	1.3305	1.3734	1.4732
	b	0.0084	0.0163	0.0210	0.0232	0.0112	0.1669	0.0199	0.0271	0.0393	0.0095
	c	0.0320	0.0529	0.0634	0.0748	0.0369	0.2941	0.0681	0.0977	0.1274	0.0278
	R^2	0.9917	0.9980	0.9973	0.9953	0.9968	0.9847	0.9608	0.9601	0.9807	0.9969
	RMSE	0.0057	0.0032	0.0039	0.0048	0.0041	0.0091	0.0134	0.0118	0.0083	0.0045
Pilosof $m_r=a+(b/(t+c))$	a	1.3266	1.2969	1.6427	1.6329	1.6053	1.5782	1.6591	1.606	1.6817	1.8012
	b	0.3127	0.3079	-3.3202	-2.5328	-4.5215	-1.3381	-2.1915	-1.6029	-1.4997	-5.9367
	c	76.2186	18.7913	12.9409	10.6037	20.2345	2.8097	8.9257	7.466	6.2891	25.998
	R^2	0.9733	0.998	0.9973	0.9953	0.9958	0.9841	0.9572	0.9572	0.9807	0.9950
	RMSE	0.0104	0.0032	0.004	0.0048	0.0048	0.0093	0.014	0.0122	0.0083	0.0057
Gornicki $m_r=a+b(1-1/(1+bct))$	a	1.3266	1.2969	1.3109	1.3228	1.3055	1.0081	1.3746	1.3298	1.3728	1.4729
	b	0.3128	0.3078	0.3310	0.3101	0.3051	0.5684	0.2921	0.2785	0.3091	0.3404
	c	0.0419	0.1728	0.1917	0.2423	0.1214	0.5187	0.2339	0.3533	0.4135	0.0822
	R^2	0.9733	0.9980	0.9973	0.9953	0.9968	0.9847	0.9608	0.9601	0.9807	0.9969
	RMSE	0.0104	0.0032	0.0039	0.0048	0.0041	0.0091	0.0134	0.0118	0.0083	0.0045
Gracia-Pascual $m_r=1-\exp(-(t/a)^b)$	a	-5.4×10 ⁷	-3.3×10 ⁷	-3.3×10 ⁷	-3.3×10 ⁷	-3.3×10 ⁷	-3.3×10 ⁷	-3.3×10 ⁷	-3.3×10 ⁷	-3.3×10 ⁷	-3.3×10 ⁷
	b	1.9×10 ⁷	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷
	R^2	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	RMSE	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Vega-Galvez $m_r=a \times \exp(-b/(1+t)^\alpha)$ $\alpha=0.9$	a	1.8145	1.5573	1.6005	1.6017	1.5429	1.5760	1.6336	1.5892	1.6680	1.7165
	b	0.2411	0.7708	0.7967	0.7244	0.7754	0.802	0.6799	0.6125	0.5948	0.7531
	R^2	0	0.9173	0.9325	0.9497	0.8924	0.9833	0.9227	0.9357	0.9565	0.863
	RMSE	0.0637	0.0208	0.0198	0.0155	0.0240	0.0095	0.0188	0.0150	0.0125	0.0297
Peppas $m_t/m_\infty=K_1 \times t+K_2 \times t^{1/2}$	K_1	-0.0305	-0.0303	-0.0302	-0.0306	-0.0301	-0.0304	-0.0306	-0.0312	-0.0315	-0.0301
	K_2	0.3877	0.3872	0.3866	0.3900	0.3844	0.3886	0.3893	0.3943	0.3980	0.3839
	R^2	0.8439	0.8550	0.8581	0.8590	0.8521	0.8740	0.8581	0.8581	0.8587	0.8482
	RMSE	2.6546	0.0385	0.0379	0.0383	0.0387	0.0356	0.0384	0.0391	0.0394	0.0393
Weibull $m_r=m_\infty+(m_0-m_\infty) \times \exp(-(t/\beta)^\alpha)$	α	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷	5.4×10 ⁷
	β	48.5200	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000	1.0000
	R^2	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
	RMSE	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND
Czel and Czigany $m_r=a \times t^m$	a	1.247	1.2696	1.2979	1.3244	1.2538	1.2862	1.3655	1.3534	1.4299	1.4005
	m	0.0396	0.0447	0.0457	0.0414	0.0455	0.0437	0.0437	0.0350	0.0334	0.0448
	R^2	0.9588	0.9902	0.9846	0.9883	0.9980	0.9042	0.9735	0.9639	0.9460	0.9956
	RMSE	0.013	0.0072	0.0094	0.0075	0.0033	0.0229	0.0282	0.0113	0.0139	0.0053

Table S2. ACR and ACR-carrageenan films' biodegradation kinetics model fittings.

Film	First-order		R ²	RMSE	HL	
	m	c				
ACR	0.1312	0.3957	0.9468	0.0305	26.8	
A-ι0.5%	0.0622	2.6222	0.8101	0.2778	20.7	
A-ι1.0%	0.0652	2.4869	0.8777	0.2384	21.9	
A-ι1.5%	0.0577	2.5670	0.8663	0.2351	23.3	
A-κ0.5%	0.0684	2.3106	0.8713	0.2575	23.4	
A-κ1.0%	0.0694	2.3032	0.8859	0.2440	23.2	
A-κ1.5%	0.0591	2.5517	0.9142	0.1879	23.0	
A-λ0.5%	0.0556	2.6122	0.8963	0.1854	23.4	
A-λ1.0%	0.0611	2.5889	0.8971	0.2028	21.6	
A-λ1.5%	0.0647	2.4125	0.9107	0.2101	23.2	
Second-order						
	a	b	c	R ²	RMSE	HL
ACR	-0.3137	0.2330	-0.0027	0.9768	0.2015	25.8
A-ι0.5%	1.9704	0.1605	-0.0027	0.9146	0.1862	17.0
A-ι1.0%	1.8283	0.1597	-0.0025	0.9747	0.1085	18.2
A-ι1.5%	1.9430	0.1432	-0.0021	0.9676	0.1158	19.3
A-κ0.5%	1.7912	0.1429	-0.0020	0.9257	0.1956	20.8
A-κ1.0%	1.6539	0.1626	-0.0025	0.9698	0.1254	19.8
A-κ1.5%	2.0564	0.1270	-0.0017	0.9783	0.0946	19.9
A-λ0.5%	2.3246	0.0970	-0.0011	0.9222	0.1605	21.6
A-λ1.0%	2.0855	0.1334	-0.0019	0.9630	0.1217	18.7
A-λ1.5%	1.8271	0.1449	-0.0020	0.9852	0.0854	19.8