## **Supplementary Information**

## Investigation of the influence of Natural Deep Eutectic Solvents (NaDES) in the properties of chitosan-stabilised films.

Antonella Rozaria Nefeli Pontillo<sup>a</sup>, Spyridon Koutsoukos<sup>b</sup>, Tom Welton<sup>b</sup> and Anastasia Detsi\*<sup>a</sup>

<sup>1</sup> Laboratory of Organic Chemistry, Department of Chemical Sciences, School of Chemical Engineering, National Technical University of Athens, Zografou, Greece

<sup>2</sup> Department of Chemistry, Molecular Sciences Research Hub, Imperial College London, White City Campus, London W12 0BZ, UK.



Figure S1. Curve of pH Vs concentration of the bet:LA NaDES and physical mixture (top) and the ChCl:LA NaDES and physical mixture (bottom).



Figure S2. FT-IR spectra of the bet:LA NaDES (top) and the ChCl:LA NaDES (bottom).



Figure S3. Deconvolution of 1200-840 cm<sup>-1</sup> range for F/AA.

Model	Gauss								
Equation		$y = y_0 + \left(\frac{A}{w * \sqrt{\frac{\pi}{2}}}\right) * e^{\left(-2 * \left(\frac{x - x_c}{w}\right)^2\right)}$							
	Peak 1	Peak 2	Peak 3	Peak 4	Peak 5	Peak 6			
<b>y</b> <sub>0</sub>	$101.1 \pm 0.1$								
X <sub>c</sub>	$1150.2 \pm 0.1  1076.4 \pm 0.3  999.9 \pm 0.2  936.8 \pm 0.3  890 \pm 0.2  855.2$					$855.2 \pm 0.6$			
W	$19.3 \pm 0.2 \qquad 97.2 \pm 0.5 \qquad 57.0 \pm 0.5 \qquad 45.1 \pm 0.6 \qquad 21.4 \pm 0.5 \qquad 36.43 = 1000 \qquad 3000 \qquad 30000 \qquad 3000 \qquad 300$					$36.6 \pm 1.1$			
А	$-153 \pm 3$ $-4122 \pm 31$ $-1650 \pm 32$ $-719 \pm 15$ $-161 \pm 6$ $-172 =$								
R-Square	0.99929								
Adj. R-Square	0.99928								

Table S1. Accuracy of deconvolution model for F/AA.



**F**igure S4. Deconvolution of 1200-840 cm<sup>-1</sup> range for F/Tween.

Model	Gauss										
Equation	$y = y_0 + \left(\frac{A}{w * \sqrt{\frac{\pi}{2}}}\right) * e^{\left(-2 * \left(\frac{x - x_c}{w}\right)^2\right)}$										
	Peak 1	Peak 1         Peak 2         Peak 3         Peak 4         Peak 5         Peak 6         Peak 7         Peak 8									
y_0		$99.31 \pm 0.05$									
Xc	1149.4 ± 0.1	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$									
W	$17.7 \pm 0.2$	$2 \begin{array}{ c c c c c c c c c c c c c c c c c c c$						$\begin{array}{c} 28.4 \pm \\ 0.3 \end{array}$			
А	$ \begin{array}{ c c c c c c c } -118 \pm 3 & -784 \pm & -3056 \pm \\ 27 & 349 & -702 \pm 65 & -534 \pm \\ 16 & -874 \pm 8 & -138 \pm 3 & -249 \pm 4 \end{array} $										
R-Square	0.99974										
Adj. R- Square	0.99974										

Table S2. Accuracy of deconvolution model for F/Tween.



Figure S5. Deconvolution of 1200-840 cm<sup>-1</sup> range for F/0.5LA.

Model	Gauss									
Equation	$y = y_0 + \left(\frac{A}{w * \sqrt{\frac{\pi}{2}}}\right) * e^{\left(-2 * \left(\frac{x - x_c}{w}\right)^2\right)}$									
	Peak 1	Peak 1         Peak 2         Peak 3         Peak 4         Peak 5         Peak 6         Peak								
y_0		$99.0 \pm 0.1$								
Xc	$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$						$\begin{array}{c} 894.5 \pm \\ 0.6 \end{array}$			
W	$26.6 \pm 0.1$	$42.2 \pm 0.6$	$35.7 \pm 0.9$	33.3±1.2	54.2 ± 4.5	19.8 ± 2	$14.1 \pm 1$			
А	-975 ± 10	$-1865 \pm 36$	$-1574 \pm 77$	$-805 \pm 87$	-758 ± 69	$-70 \pm 19$	$-52.9 \pm 8.9$			
R-Square	0.99939									
Adj. R- Square	0.99938									

Table S3. Accuracy of deconvolution model for F/0.5LA.



Figure S6. Deconvolution of 1200-920 cm<sup>-1</sup> range for F/bet:LA.

Model	Gauss										
Equation	$y = y_0 + \left(\frac{A}{w * \sqrt{\frac{\pi}{2}}}\right) * e^{\left(-2 * \left(\frac{x - x_c}{w}\right)^2\right)}$										
	Peak 1	Peak 1         Peak 2         Peak 3         Peak 4         Peak 5         Peak 6         Peak									
y_0		$100 \pm 0$									
Xc	1126.4 ± 0.1	$1081.5 \pm 0.1$	$1032.8 \pm 0.1$	$1001.8 \pm 0.1$	978.5 ± 0.1	954.3 ± 0.1	931.7 ± 0.1				
W	$27.9 \pm 0.1$	$43.7 \pm 0.3$	$29.9 \pm 0.3$	21.8 ± 0.5	$\begin{array}{c} 17.5 \pm \\ 0.3 \end{array}$	$\begin{array}{c} 15.6 \pm \\ 0.3 \end{array}$	$\begin{array}{c} 17.3 \pm \\ 0.1 \end{array}$				
А	$-743 \pm 4$	-1181 ± 8	$-706 \pm 9$	$-220 \pm 8$	-196 ± 5	$-113 \pm 2$	-229.1 ±				
R-Square	0.99959										
Adj. R- Square	0.99959										

Table S4. Accuracy of deconvolution model for F/bet:LA.



Figure S7. Deconvolution of 1200-840 cm<sup>-1</sup> range for F/bet:LA mix.

Model	Gauss									
Equation	$y = y_0 + \left(\frac{A}{w * \sqrt{\frac{\pi}{2}}}\right) * e^{\left(-2 * \left(\frac{x - x_c}{w}\right)^2\right)}$									
	Peak 1	Peak 1         Peak 2         Peak 3         Peak 4         Peak 5         Peak 6         Peak 7         Peak 8								
y_0		$100 \pm 0$								
Xc	$1127.7 \pm 0.0$	$1079.3 \pm 0.1$	$1030.5 \pm 0.1$	$1001.1 \pm 0.2$	978.7 ± 0.2	$\begin{array}{c} 954.9 \pm \\ 0.1 \end{array}$	932.7 ± 0.1	$\begin{array}{c} 893.1 \pm \\ 0.1 \end{array}$		
w	$25.6 \pm 0.1$	$43.9 \pm 0.3$	33.7 ± 0.3	$19.8 \pm 0.5$	$\begin{array}{c} 20.0 \pm \\ 0.4 \end{array}$	15.7± 0.3	18.6±0.1	17.9 ± 0.1		
А	$-762 \pm 4$	-1654 ± 10	-1125 ± 15	$-292 \pm 14$	-401 ± 12	-170 ± 5	$-364 \pm 3$	$-424 \pm 1$		
R-Square	0.99971									
Adj. R- Square		0.99971								

Table S5. Accuracy of deconvolution model for F/bet:LA phys. mix.



Figure S8. Deconvolution of 1200-840 cm<sup>-1</sup> range for F/ChCl:LA.

Model	Gauss								
Equation	$y = y_0 + \left(\frac{A}{w * \sqrt{\frac{\pi}{2}}}\right) * e^{\left(-2 * \left(\frac{x - x_c}{w}\right)^2\right)}$								
	Peak 1 Peak 2 Peak 3 Peak 4 Peak					Peak 6			
y <sub>0</sub>	$100 \pm 0$								
X <sub>c</sub>	$1126.5 \pm 0.0 \qquad 1082.8 \pm 0.1 \qquad 1043.2 \pm 0.0 \qquad 1004.0 \pm 0.0 \qquad 952.9 \pm 0.0 \qquad 922.1 = 0.0 \qquad 922.1 \pm 0.0 \qquad 922.1 = 0.$								
W	$24.6 \pm 0.1$ $26.5 \pm 0.1$ $32.1 \pm 0.2$ $23.2 \pm 0.2$ $19.1 \pm 0.0$ $16.6 \pm 0.0$								
А	$-891 \pm 3 \qquad -948 \pm 5 \qquad -972 \pm 7 \qquad -333 \pm 4 \qquad -669 \pm 2 \qquad -134 \pm 2$								
R-Square	0.99921								
Adj. R-Square	0.99920								

Table S6. Accuracy of deconvolution model for F/ChCl:LA .



Figure S9. Deconvolution of 1200-840 cm<sup>-1</sup> range for F/ChCl:LA mix.

Model	Gauss								
Equation	$y = y_0 + \left(\frac{A}{w * \sqrt{\frac{\pi}{2}}}\right) * e^{\left(-2 * \left(\frac{x - x_c}{w}\right)^2\right)}$								
	Peak 1 Peak 2 Peak 3 Peak 4 Peak 5 Peal								
y <sub>0</sub>	$98.9 \pm 0$								
X <sub>c</sub>	$1126.5 \pm 0.0 \qquad 1082.8 \pm 0.1 \qquad 1040.7 \pm 0.1 \qquad 998.6 \pm 0.3 \qquad 952.7 \pm 0.0 \qquad 922.2 \pm 0.0$								
W	$26.6 \pm 0.1$ $28.9 \pm 0.2$ $44.4 \pm 0.9$ $28.0 \pm 0.3$ $20.4 \pm 0.1$ $20.1 \pm 0.3$								
А	$-883 \pm 5 \qquad -1123 \pm 23 \qquad -1865 \pm 39 \qquad -522 \pm 19 \qquad -744 \pm 3 \qquad -179 \pm 3$								
R-Square	0.99905								
Adj. R-Square	0.99903								

Table S7. Accuracy of deconvolution model for F/ChCl:LA mix.



Figure S10. Gauss Curve equation parameters explanation.



Figure S11. TGA graphs of the bet:LA NaDES (top) and the ChCl:LA NaDES (down).



Figure S12. TGA-MS of F/AA.



Figure S13. TGA-MS of F/Tween.



Figure S14. TGA-MS of F/0.5LA



Figure S15. TGA-MS of F/bet:LA NaDES



Figure S16. TGA/MS of F/bet:LA mix



Figure S17. TGA-MS of F/ChCl:LA NaDES



Figure S18. TGA-MS of F/ChCl:LA mix



Figure S19. TGA-MS of chitosan



Figure S20. TGA of acetic acid.



Figure S21. TGA-MS of lactic acid.



Figure S22. TGA-MS of betaine.



Figure S23. TGA-MS of choline chloride.