Electronic Supplementary Information

Complex calcium carbonate/polymer microparticles as carriers for aminoglycoside antibiotics

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Abstract

Composite microparticles of CaCO₃ and two pectin samples (which differ by the functional group ratio) or corresponding nonstoichiometric polyelectrolyte complexes with different molar ratios (0.5, 0.9 and 1.2) are obtained, characterized and tested for loading and release of streptomycin (STR) and kanamycin (KAN) sulphate. The synthesized carriers were characterized before and after drug loading in terms of morphology (by SEM using secondary electron and energy selective backscattered electron detectors), porosity (by water sorption isotherms) and elemental composition (by elemental mapping using the energy dispersive X-Ray and FTIR spectroscopy). The kinetic of the release mechanism from microparticles was investigated using Higuchi and Korsmeyer-Peppas mathematical models.

Keywords: calcium carbonate, pectin, polyelectrolyte complexes, streptomycin, kanamycin

Scheme 1S. Chemical structure of polymers [pectins – PCTxx, and poly(allyamine hydrochloride) – PAH] and drugs [streptomycin (STR) and *kanamycin* (KAN)] used in this study



Table 1S. Diffusion coefficients for pectin-CaCO₃ and NPEC-CaCO₃ composite microparticles, determined from sorption-desorption experimental data

Pectin type	NPEC	$10^{-5} K_1^*,$	$10^{-4} K_2^*$,	l	D_1	D_2
	n⁺/n⁻	$M_t/M_\infty < 0.5$	$M_t/M_\infty > 0.5$	(cm)	(10 ⁻⁸ cm ² /s)	(10 ⁻⁷ cm ² /s)
PCT49	0	3.00	-5.26	0.1	5.89	5.33
	0.5	2.76	-6.88	0.1	5.41	6.98
	0.9	2.44	-6.33	0.1	4.79	6.41
	1.2	2.49	-6.99	0.1	4.90	7.09
PCT53_A21	0	3.71	-5.08	0.1	7.28	5.15
	0.5	2.94	-6.44	0.1	5.76	6.53
	0.9	5.18	-8.92	0.1	5.18	9.04
	1.2	2.06	-5.38	0.1	4.04	5.45

* K_1 is slope of linear regression between $(t-t_R)$ and $(M_t/M_{\infty})^2$ for $(t-t_R) \ge 0$ and $(M_t/M_{\infty})^2 < 0.2$ where t_R is time correlation (minus intercept/slope) for $M_t/M_{\infty}=0$; K_2 is slope of linear regression between t and $ln(1-M_t/M_{\infty})$ for -1.2 > ln > -3.0



Figure 1S. Sorption/desorption isotherms as relative humidity (RH) vs. weight variation



Figure 2S. SEM micrograph of pectin-CaCO₃ and NPEC-CaCO₃ microparticles after STR retention



Figure 3S. SEM micrograph of pectin-CaCO₃ and NPEC-CaCO₃ microparticles after KAN retention





Figure 4S. STR and KAN in-vitro release



Figure 5S. SEM micrograph of composite microparticles after KAN release



Figure 6S. FTIR spectra of STR loaded sample: (1) CaCO₃/(PCT49/PAH=0.5), (2) CaCO₃/(PCT49/PAH=0.9), (3) CaCO₃/(PCT49/PAH=1.2) and after STR release: (4) CaCO₃/(PCT49/PAH=0.5), (5) CaCO₃/(PCT49/PAH=0.9), (6) CaCO₃/(PCT49/PAH=1.2)



Figure 7S. FTIR spectra of KAN loaded sample: (1) CaCO₃/(PCT49/PAH=0.5), (2) CaCO₃/(PCT49/PAH=0.9), (3) CaCO₃/(PCT49/PAH=1.2) and after KAN release: (4) CaCO₃/(PCT49/PAH=0.5), (5) CaCO₃/(PCT49/PAH=0.9), (6) CaCO₃/(PCT49/PAH=1.2)



Figure 8S. FTIR spectra of STR loaded sample: (1) CaCO₃/(PCT53_A21/PAH=0.5), (2) CaCO₃/(PCT53_A21/PAH=0.9), (3) CaCO₃/(PCT53_A21/PAH=1.2) and after STR release: (4) CaCO₃/(PCT53_A21/PAH=0.5), (5) CaCO₃/(PCT53_A21/PAH=0.9), (6) CaCO₃/(PCT53_A21/PAH=1.2)



Figure 9S. FTIR spectra of KAN loaded sample: (1) CaCO₃/(PCT53_A21/PAH=0.5), (2) CaCO₃/(PCT53_A21/PAH=0.9), (3) CaCO₃/(PCT53_A21/PAH=1.2) and after KAN release: (4) CaCO₃/(PCT53_A21/PAH=0.5), (5) CaCO₃/(PCT53_A21/PAH=0.9), (6) CaCO₃/(PCT53_A21/PAH=1.2)



Figure 10S. Graphical representation of Higuchi equation



Figure 11S. Graphical representation of Korsmeyer-Peppas equation