Supporting Information

pH-responsive nanocomposite fibres allowing MRI monitoring of drug release

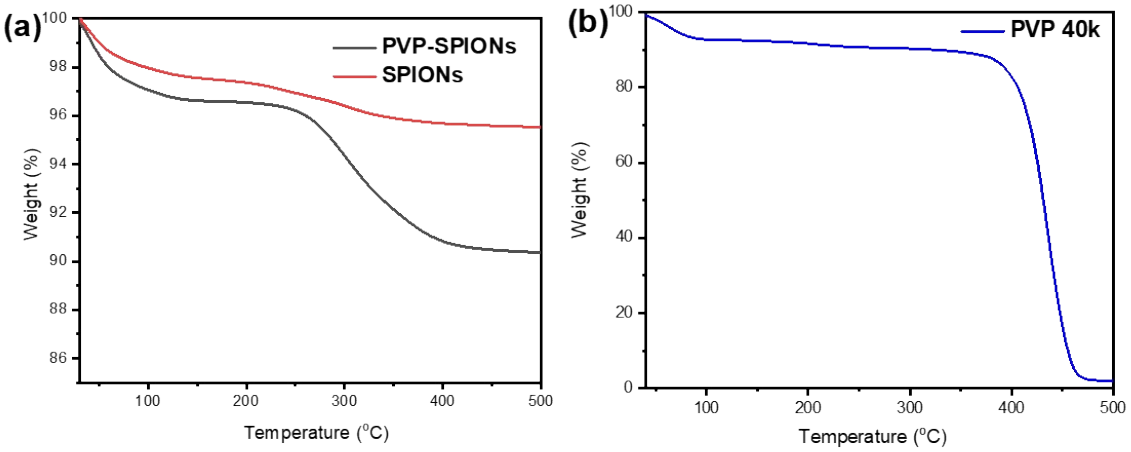

Figure S1. TGA curves of (a) SPIONs and PVP-SPIONs and (b) PVP 40k.

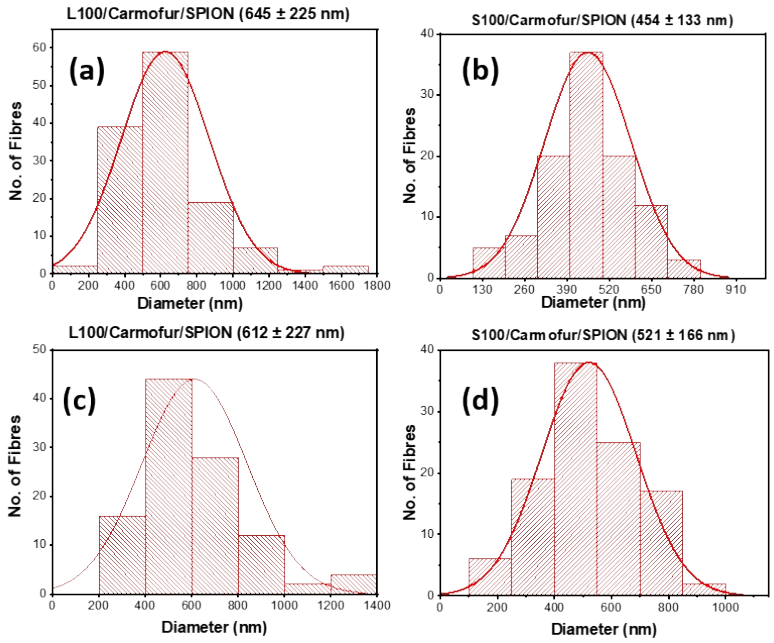

Figure S2. Fibre diameter distributions for (a) L100/Carmofur/SPION (645 ± 225 nm) and (b) S100/Carmofur/SPION fibres (454 ± 133 nm); and (c) the L100/Carmofur/SPION (612 ± 227 nm and (d) S100/Carmofur/SPION fibres (521 ± 166 nm) after suspension in pH 1.5 HCl for 2 h. Size data were obtained by size analysis of more than 100 fibres in SEM using the ImageJ software.

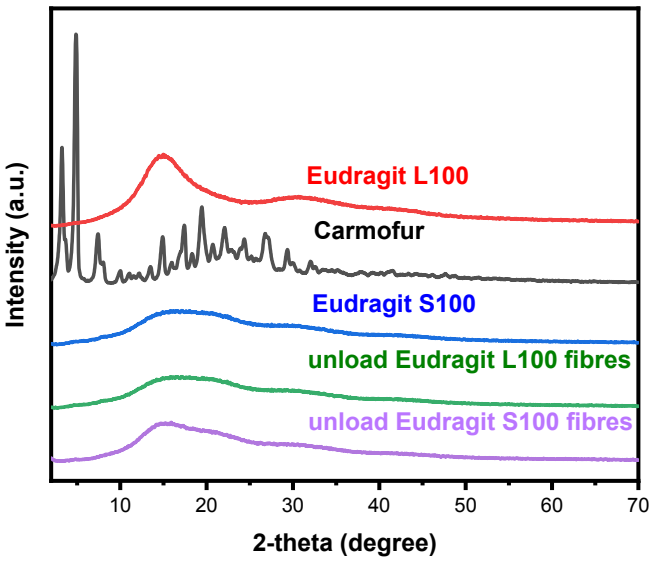

Figure S3. XRD patterns of the raw materials (carmofur, Eudragit L100 and S100) and drug-free L100 and S100 fibres.

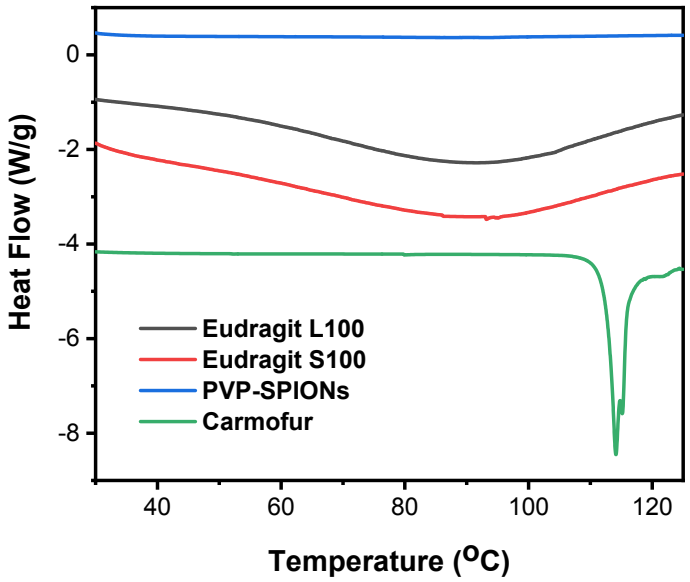

Figure S4. DSC thermograms of the Eudragit polymers, carmofur, and the PVP-SPIONs.

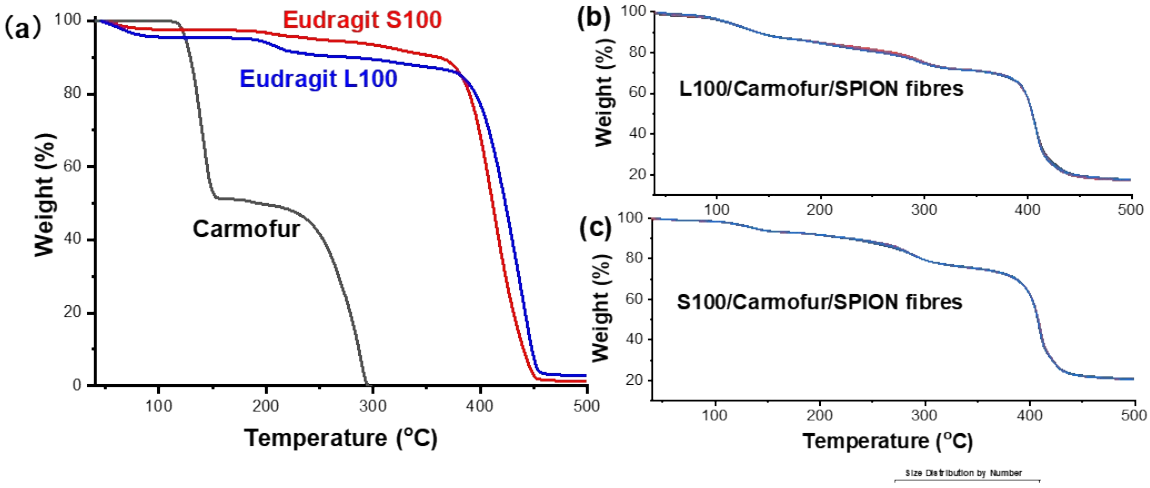

Figure S5. (a) TGA curves of Eudragit L100 and S100 and carmofur; repeat TGA profiles of (b) L100/Carmofur/SPION and (c) S100/Carmofur/SPION fibres.

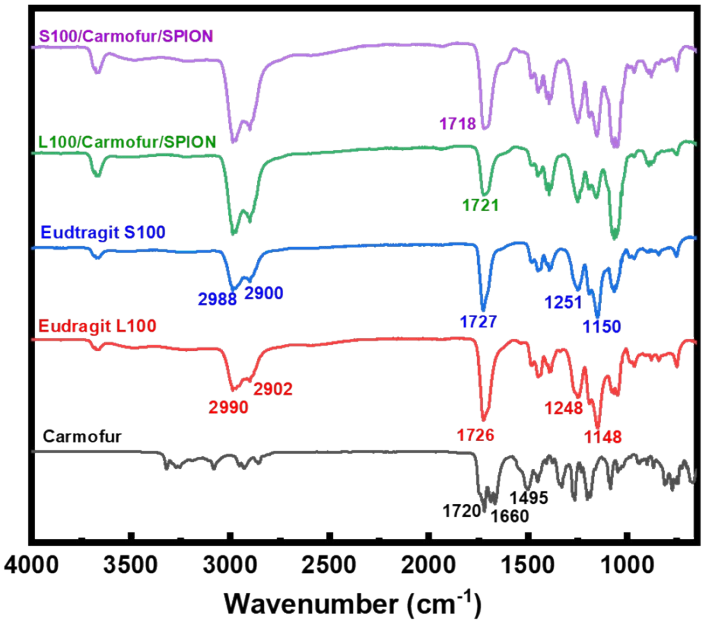

Figure S6. FTIR spectra of the raw materials (carmofur, Eudragit L100 and S100), and the L100/Carmofur/SPION and S100/Carmofur/SPION fibres.

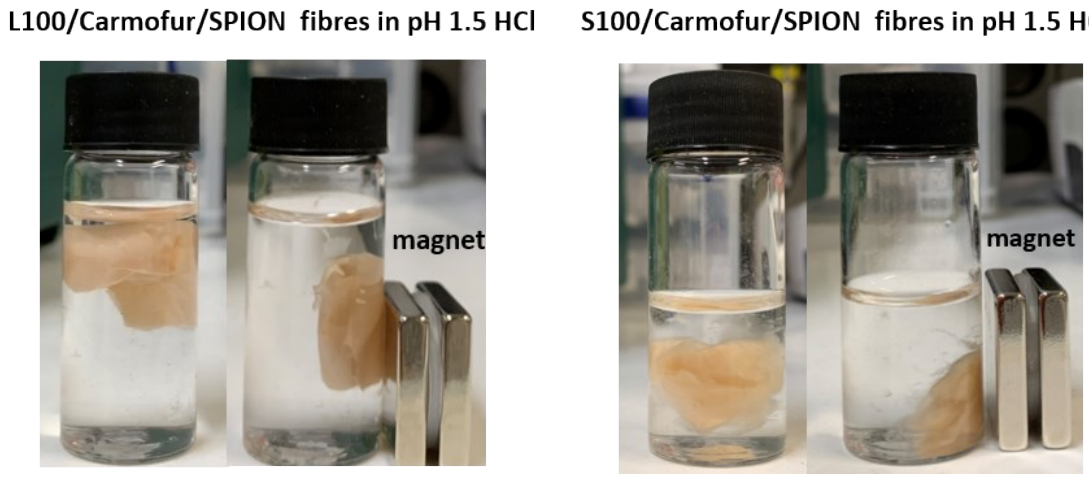

Figure S7. Photographs of suspensions of the fibres in pH 1.5 HCl solution (0.4 mg/mL) after two hours' immersion.

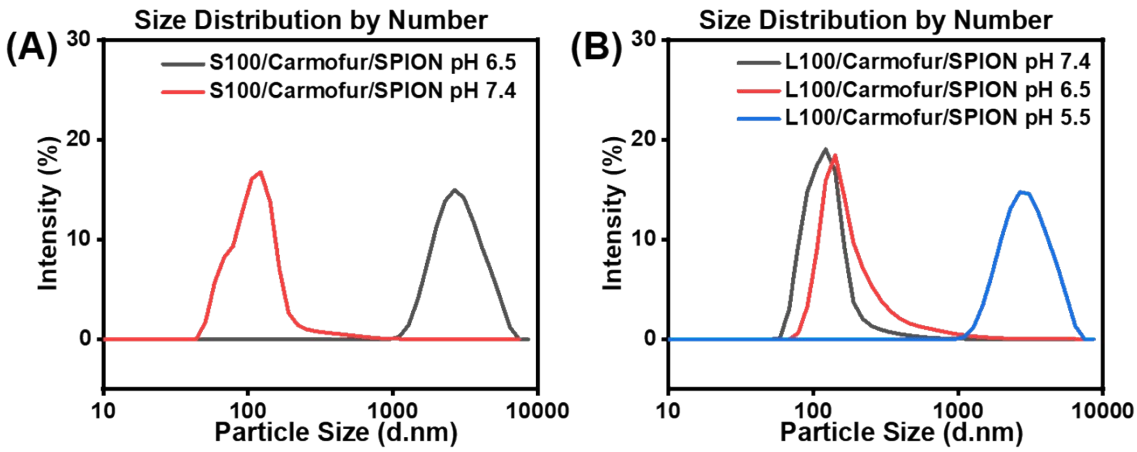

Figure S8. Particle size distribution curves (~ 0.5 mg/mL) of (a) S100/Carmofur/SPION (458 ± 31 nm at pH 7.4 and 2959 ± 176 nm at pH 6.5) and (b) L100/Carmofur/SPION fibres (380 ± 11 nm at pH 7.4, 591 ± 32 nm at pH 6.5, and 3049 ± 41 nm at pH 5.5). Measurements were taken after 24 h of shaking incubation (100 rpm, 37 °C).

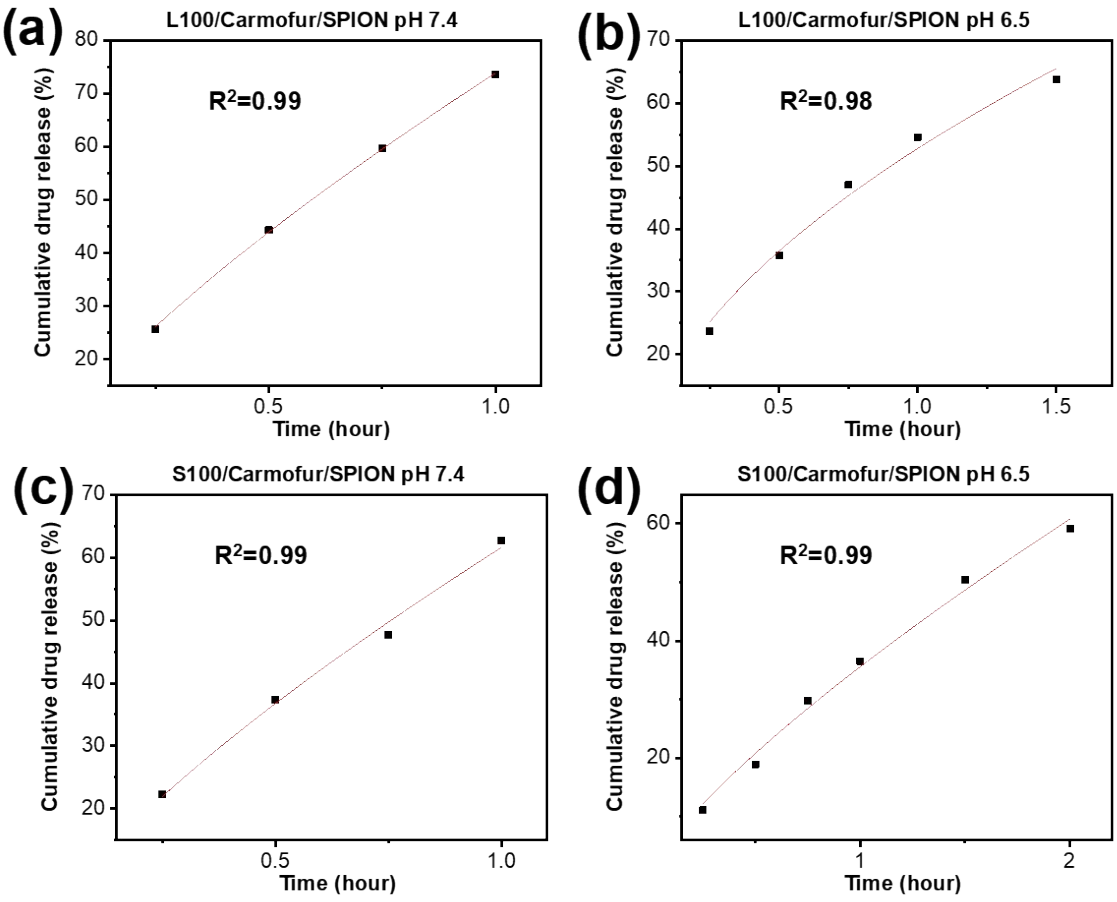

Figure S9. Fits of the Peppas model to the carmofur release data from L100/Carmofur/SPION fibres at (a) pH 7.4 and (b) 6.5, and S100/Carmofur/SPION fibres at (c) pH 7.4 and (d) 6.5.

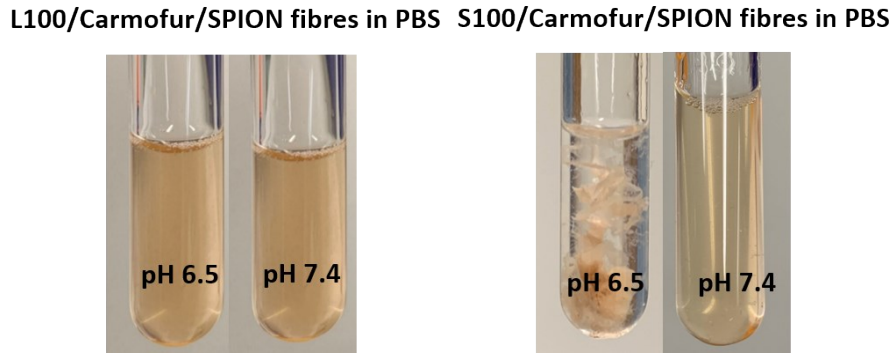

Figure S10. Photographs of the fibres in PBS buffer at different pH after 3 h of immersion. L100/Carmofur/SPIONs dissolve in PBS at both pH 6.5 or 7.4, while S100/Carmofur/SPION dissolves at pH 7.4 but not pH 6.5.

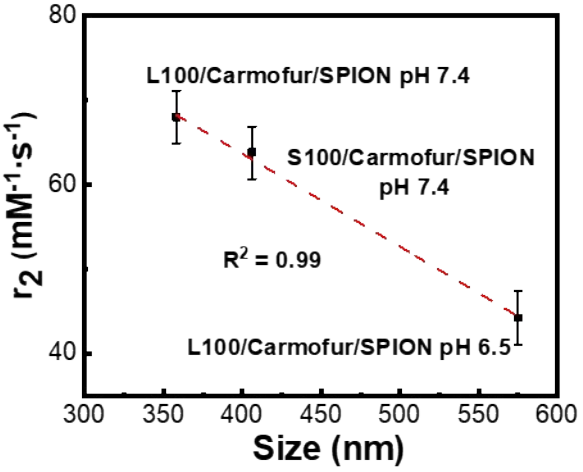

Figure S11. The relationship between the particle size of the dissolved L100/Carmofur/SPION and S100/Carmofur/SPION fibres and relaxivity.

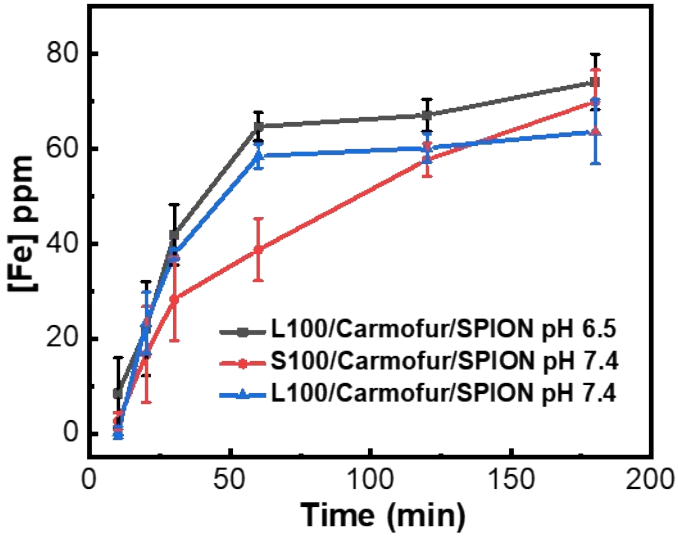

Figure S12. The release of SPIONs from the S100/Carmofur/SPION and L100/Carmofur/SPION formulations during relaxation monitoring experiments, measured by quantifying [Fe] release using ICP-MS (n=3).

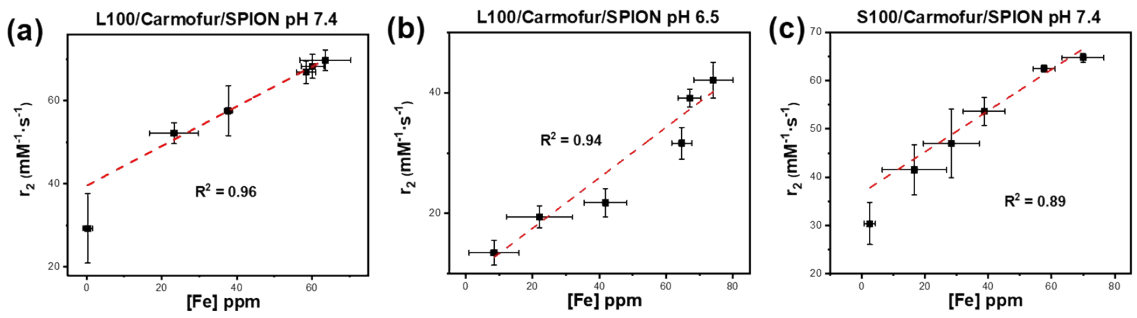

Figure S13. The relationship between Fe release and relaxation behaviour of the L100/Carmofur/SPION fibres at (a) pH 7.4 and (b) pH 6.5, and (c) the S100/Carmofur/SPION fibres at pH 7.4.

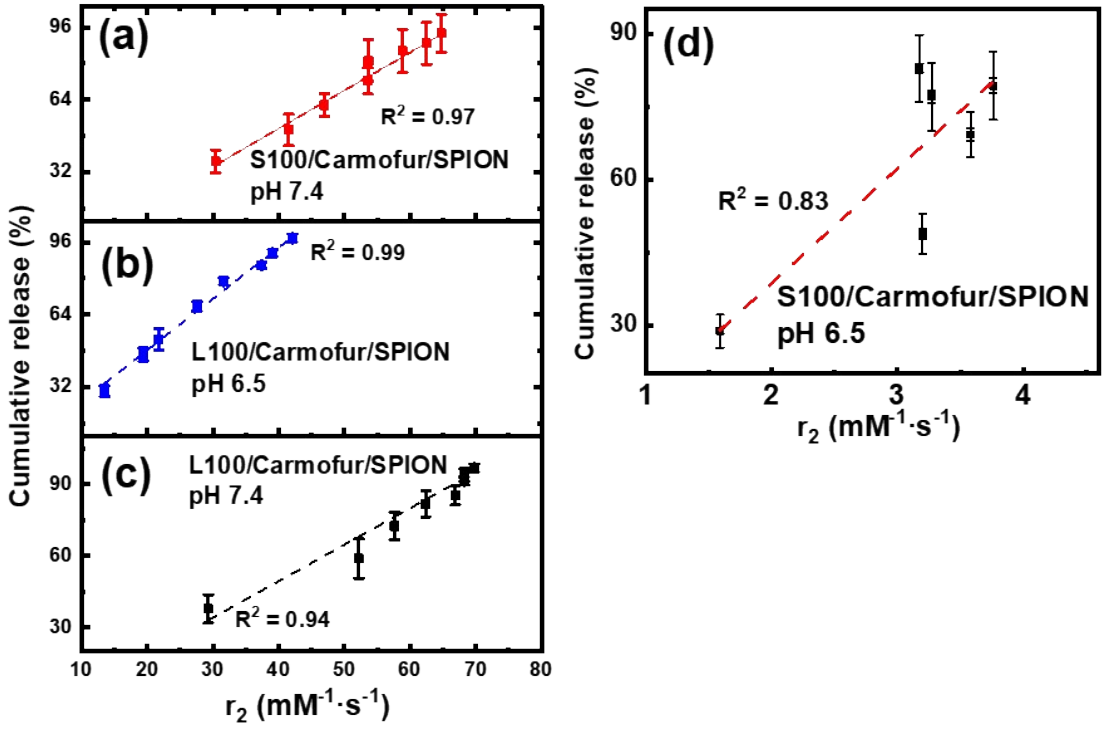

Figure S14. The relationship between relaxation behaviour (r_{2_t}) and cumulative carmofur release of L100/Carmofur/SPION fibres at (a) pH 7.4 and (b) pH 6.5, and S100/Carmofur/SPION fibres at (c) pH 7.4 and (d) 6.5.

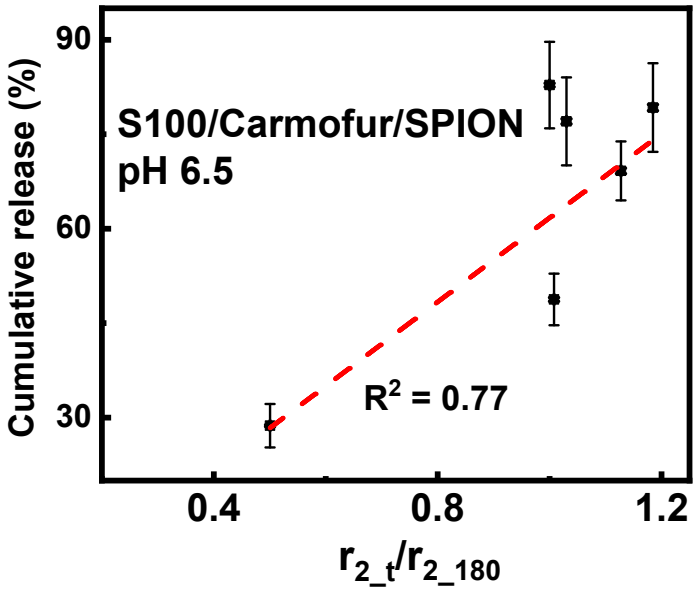

Figure S15. The relationship between relaxation behaviour (r_{2_t}/r_{2_180}) and cumulative carmofur release for the S100/Carmofur/SPION fibres at pH 6.5.

Table S1. The hydrodynamic size and PDI of the L100/Carmofur/SPION and S100/Carmofur/SPION samples at different pHs. Measurements were taken after 24 hours of shaking incubation (100 rpm, 37 °C). The fibre concentration was *ca*. 0.5 mg/mL.

Formulation	pH	Hydrodynamic diameter / nm (n=3)	PDI (n=3)
L100/Carmofur/SPION	**7.4**	380 ± 11	0.53 ± 0.02
	6.5	591 ± 32	0.64 ± 0.04
	5.5	3049 ± 41	0.20 ± 0.01
S100/Carmofur/SPION	**7.4**	458 ± 31	0.50 ± 0.02
	6.5	2959 ± 176	0.24 ± 0.05

Table S2. The linear relationship between cumulative drug release (%) and relaxation profile (r_{2_t}), constructed from the plots in Figure S14.

Formulation	pH	Fitting equation	R^2
S100/Carmofur/SPION	7.4	$Drug\ release\ (\%) = 1.68r_{2\_t} - 16.0$	0.98
L100/Carmofur/SPION	6.5	$Drug\ release\ (\%) = 2.31r_{2\_t} + 1.6$	0.99
	7.4	$Drug\ release\ (\%) = 1.53r_{2\_t} - 11.6$	0.94