## **Supporting Information**

pH-Responsive Supramolecular Hydrogels for Codelivery of Hydrophobic and Hydrophilic Anticancer Drugs

Jing Yu<sup>a, b</sup>, Wei Ha<sup>a</sup>, Juan Chen<sup>a</sup>, Yan-ping Shi<sup>a\*</sup>

<sup>a</sup> Key Laboratory of Chemistry of Northwestern Plant Resources and Key Laboratory for Natural Medicine of Gansu Province, Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, Lanzhou 730000, PR China

<sup>b</sup> University of Chinese Academy of Sciences, Beijing 100049, PR China

<sup>\*</sup>Correspondence: Prof Yan-ping Shi, Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, Lanzhou 730000, P. R. China; E-mail: shiyp@licp.cas.cn; Fax: +86-931-4968094; Tel: +86-931-4968028



Fig. S1 <sup>1</sup>H NMR (400MHz, CDCl<sub>3</sub>) spectrum of mPEG formyl benzoic acid ester.



**Fig. S2** <sup>1</sup>H NMR (a) (400MHz, CDCl<sub>3</sub>) and <sup>13</sup>C NMR (b) (100MHz, CDCl<sub>3</sub>) spectrum of NPOD-PEG.



Fig. S3 Calibration curve of DOX.



Fig. S4 The dependence of the viscoelastic moduli on frequency for various NPOD-PEG/ $\alpha$ -CD hydrogel samples. (a) NPOD-PEG = 10 mg mL<sup>-1</sup>, b) NPOD-PEG = 30 mg mL<sup>-1</sup>.



Fig. S5 (a) Dynamic and (b) steady rheological behaviors of the diluted HCl-treated NPOD-PEG/ $\alpha$ -CD hydrogel.

	_	NPOD		DOX		(5:1) NPOD:DOX			
Cell	$D_{\rm m}$								
type	(µg/m	g/m Linear equation		$D_{\rm m}$	Linear equation	r	$D_{\mathrm{m}}$	Linear equation	r
	L)								
A54	4.29	y=2.3402x-	0.9	4.4	y=1,4955x-	0.9	3.72+0.7	y=2.2122x-	0.9
9		1.4818	9	8	0.9739	6	4	1.4370	9

**Table S1.** Dose-effect relationship parameters for NPOD and DOX in cancer model

Shape (sigmoidicity) and conformity of dose-effect curve (linear correlation coefficient) are represented by  $D_{\rm m}$ , linear equation, r, respectively, where  $D_{\rm m}$  is the antilog of x-intercept in µg mL<sup>-1</sup>, r is the linear correlation coefficient of the median-effect plot.

**Table S2.** Interaction of NPOD and DOX combinations in cells at different stage of

 carcinogenesis: combination indices at different effect levels

Cell	Combination index (CI) at:										
type	<i>f</i> a0.1	fa0.2	fa0.3	fa0.4	fa0.5	fa0.6	<i>f</i> a0.7	fa0.8	<i>f</i> a0.9		
A549	1.09	1.06	1.05	1.04	1.03	1.03	1.02	1.02	1.02		

CI value <1, =1, >1 indicates synergism, additive effect, and antagonism, respectively. *f*a is the fraction effected.