Supporting Information

Facile Fabrication of Tea Tree Oil-Loaded Antibacterial Microcapsules by Complex Coacervation of Sodium Alginate/Quaternary Ammonium Salt of Chitosan

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Table S1. The independent variable and response value for the optimization of TTO-loaded microcapsules by using Box-Benhnken central composite design.

Treatment No. [a]	X_1 [b] Core-wall ratio (wt%)	X_2 pH value	X_3 Concentration of $CaCl_2$ (w/v %)	Response variable Actual EE (%)	Predict EE (%)
1	0[c] (1:1)	0 (6)	0 (0.6)	65.36 (±0.70)[d]	66.06
2	0 (1:1)	1 (7)	-1 (0.3)	47.08 (±0.23)	47.31
3	0 (1:1)	1 (7)	1 (0.9)	52.46 (±1.06)	51.40
4	1 (3:2)	0 (6)	1 (0.9)	49.28 (±0.04)	49.32
5	0 (1:1)	0 (6)	0 (0.6)	68.21 (±2.15)	66.06
6	0 (1:1)	-1 (5)	-1 (0.3)	49.55 (±1.06)	50.61
7	-1 (1:2)	-1 (5)	0 (0.6)	54.67 (±1.02)	53.65
8	0 (1:1)	0 (6)	0 (0.6)	67.72 (±1.66)	66.06
9	0 (1:1)	-1 (5)	1 (0.9)	56.37 (±0.23)	56.14
10	-1 (1:2)	0 (6)	1 (0.9)	53.04 (±1.25)	54.29
11	0 (1:1)	0 (6)	0 (0.6)	63.53 (±2.53)	66.06
12	0 (1:1)	0 (6)	0 (0.6)	65.48 (±0.58)	66.06
13	-1 (1:2)	1 (7)	0 (0.6)	48.28 (±0.19)	48.09
14	1 (3:2)	-1 (5)	0 (0.6)	46.14 (±0.19)	46.33
15	1 (3:2)	1 (7)	0 (0.6)	42.83 (±1.02)	43.85
16	1 (3:2)	0 (6)	-1 (0.3)	44.95 (±1.25)	43.70
17	-1 (1:2)	0 (6)	-1 (0.3)	50.33 (±0.04)	50.29

[a] The treatment were run as a stochastic order.

[b] The factors (X_1), (X_2) and (X_3) are variables.

[c] The values (-1), (0) and (1) are coded levels.

[d] The residual is actual encapsulation efficiency versus predict encapsulation efficiency.

Table S2. Analysis of variance of the regression parameters for TTO-loaded microcapsules.

Source	Sum of squares	Df	Mean square	F-value	p-value Prob > F
Model	1130.43	9	125.60	39.53	< 0.0001 ***
X_1	66.82	1	66.82	21.03	0.0025 **
X_2	32.32	1	66.82	10.17	0.0153 *
X_3	46.27	1	46.27	14.56	0.0066 **
$X_1 X_2$	2.37	1	2.37	0.75	0.4162 #
$X_1 X_3$	0.66	1	0.66	0.21	0.6633 #
$X_2 X_3$	0.52	1	0.52	0.16	0.6983 #
X_1^2	422.95	1	422.95	133.12	< 0.0001 ***
X_2^2	273.36	1	273.36	86.04	< 0.0001 ***
X_3^2	185.50	1	185.50	58.39	0.0001 ***
Residual	22.24	7	3.18		
Lack of Fit	7.63	3	2.54	0.70	0.6008 #
Pure Error	14.61	4	3.65		
Cor Total	1152.67	16			
R^2					0.9807
Adj R^2					0.9559
Pred R^2					0.8742
Adeq Precision					16.356

* ,**, *** and # indicate significant ($P < 0.05$), highly significant ($P < 0.01$), top significant ($P < 0.001$) and not significant, respectively.

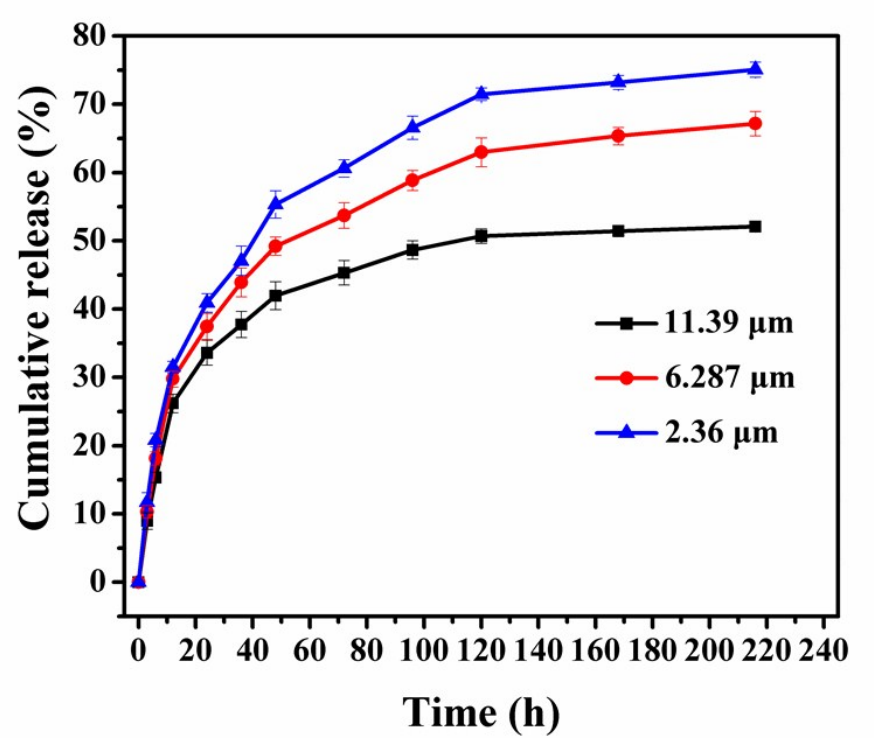

Fig. S1. In vitro release profiles of TTO from the TTO-loaded microcapsules with different sizes.

The TTO-loaded microcapsules with different sizes (2.36 μm, 6.287 μm and 11.39 μm) were successfully prepare by various shearing velocity. And then the vitro TTO release studies was implemented according to the means of 2.6 in the manuscript.

Table S3. The results of fitting the TTO release profiles by different kinetic models.

Test number		Kinetic models											
		Zero-order			First-order			Higuchi			Rigter-Peppas		
		$Q_t = K_0t + C_0$			$\ln(100-Q_t) = K_1t + C_1$			$Q_t = K_Ht^{1/2} + C_H$			$Q_t = K_pt^n$		
		K_0	C_0	R_0^2	K_1	C_1	R_1^2	K_H	C_H	R_H^2	K_p	n_p	R_p^2
T[a]	4°C	0.15	17.40	0.7257	-0.0021	4.41	0.7767	2.75	7.78	0.8968	1.57	0.44	0.9101
	25°C	0.19	24.05	0.7252	-0.0032	4.33	0.8020	3.56	11.59	0.8965	2.01	0.41	0.9286
	37°C	0.24	28.01	0.7408	-0.0045	4.28	0.8431	4.28	13.09	0.9069	2.17	0.42	0.9371
H[b]	24%	0.24	28.01	0.7408	-0.0045	4.28	0.8431	4.28	13.09	0.9069	2.17	0.42	0.9371
	35%	0.27	33.49	0.7027	-0.0061	4.19	0.8248	4.92	16.10	0.8818	3.36	0.40	0.9383
	50%	0.29	39.09	0.6892	-0.0088	4.11	0.8453	5.54	19.42	0.8730	2.53	0.39	0.9398

T[a]: The environmental temperature; H[b]: the environmental humidity;

Q_t : The % cumulative release rate of tea tree oil from the microcapsules at time t;

K_0 , C_0, K_1, C_1, K_H, C_H, K_p : Constants of the corresponding kinetic models;

n_p : The diffusion index of Rigter-Peppas models: Fickian diffusion ($n_p \leq 0.45$), non Fickian diffusion ($0.45 < n_p < 0.89$) or matrix erosion diffusion ($n_p \geq 0.89$);

R_0^2, R_1^2, R_H^2 , R_p^2: Correlation coefficients of the corresponding kinetic models.