

Biocompatible, Injectable and Self-healable MOF-Based Anti-freezing Eutectogel for Higher Encapsulation and Sustained Release of Anticancer Drug Curcumin

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Table S1: Effect of different composition on eutectogel formation.

DES (1 ml)	MOF % (w/v)	Suspension	SA (w/v)	Gel
CG DES, CF DES	0.2	Evenly distributed particles	–	–
	0.4	Evenly distributed particles	–	–
	0.6	Evenly distributed particles	–	–
	0.8	Evenly distributed particles	–	–
	1.0	Evenly distributed particles	1 %	Viscous sol
			2 %	Loose gel
			3 %	Gel
	1.2	Particles began to settle down	–	–
Beyond 1.2	Particles settled down	–	–	

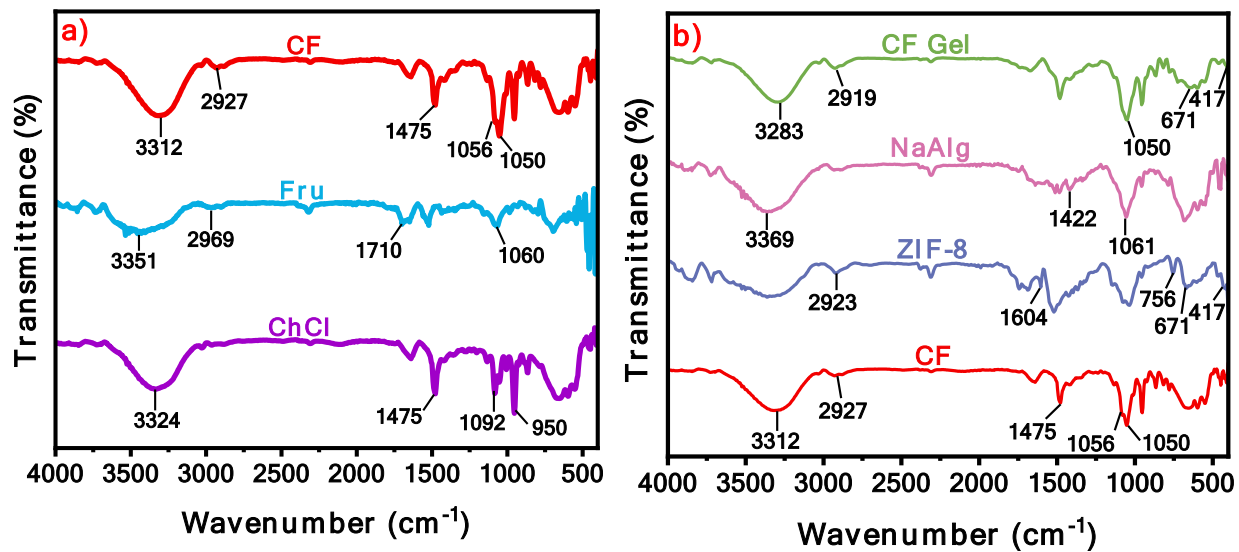


Figure S1: FT-IR spectra of a) ChCl, Fru and, CF; b) CF, SA, ZIF-8, CF Gel.

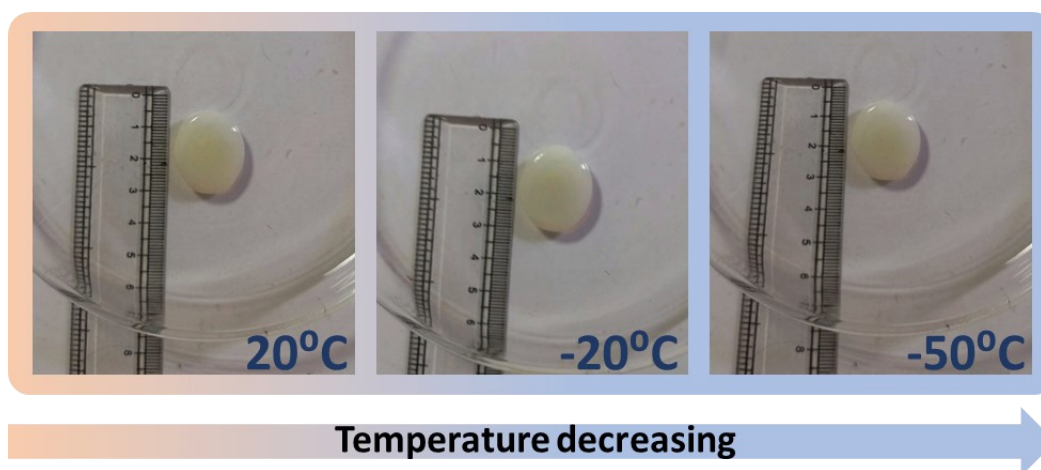


Figure S2: Digital photographs of representative eutectogel at different temperature (20°C, -20°C, and -50°C).

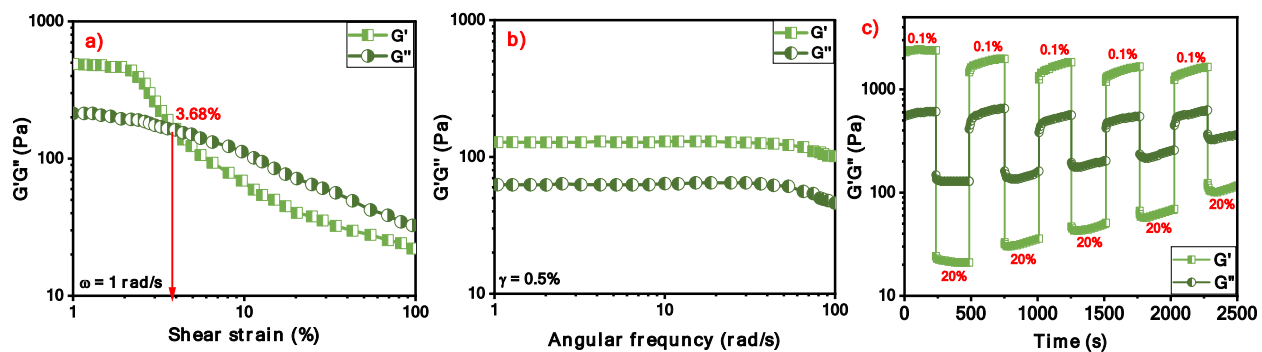


Figure S3: a) Strain sweep measurement (CF Gel); b) Frequency sweep measurement (CF gel); c) Thixotropic behavior (CF gel).

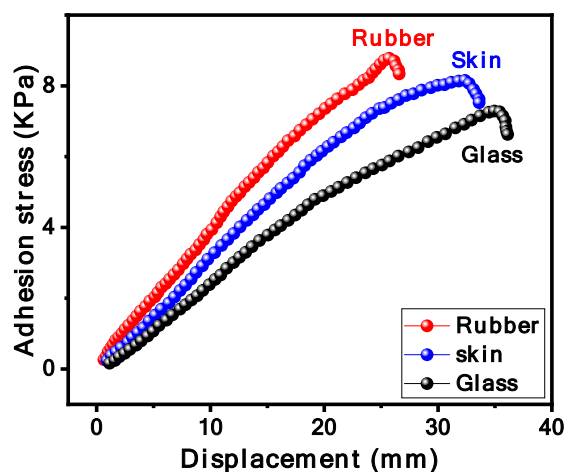


Figure S4: Shear stress-displacement profiles of representative eutectogel on different substrates.

Table S2: Hemolysis (%) level of CG and CF gel at range of increasing concentration. (Place in ESI)

Gel	Concentration (mg/mL)	Hemolysis (%) \pm 0.05
CG Gel	10	0.98
	20	1.02
	30	1.80
	40	1.50
	50	1.70
CF Gel	10	0.92

	20	1.34
	30	1.41
	40	1.30
	50	1.60

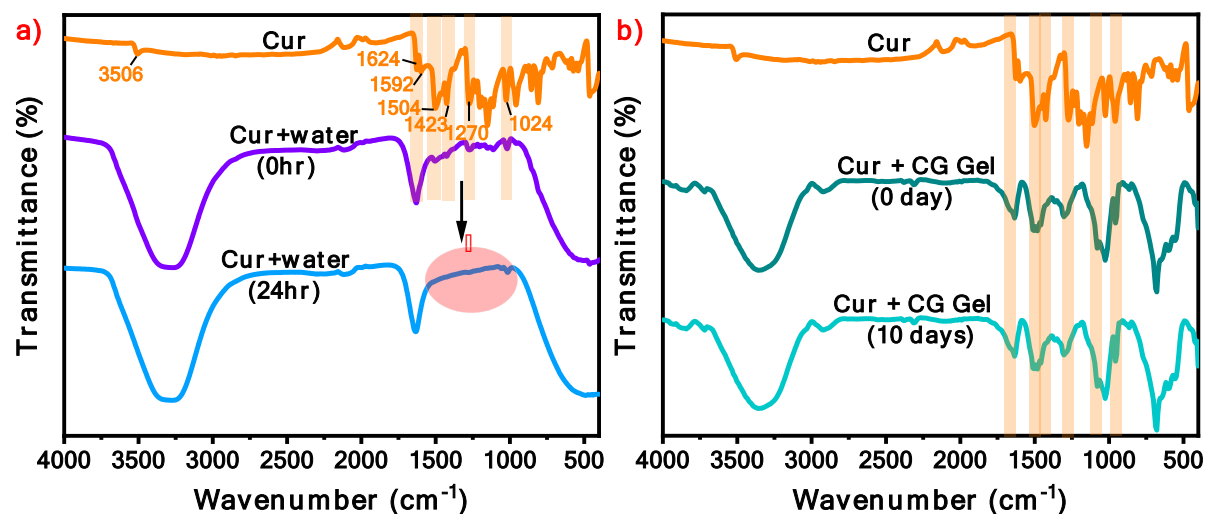


Figure S5: Comparative FTIR spectra of **a)** Cur, cur in water (0hr) and cur in water (24hr), **b)** Cur, cur loaded CG gel (0 day) and cur loaded CG gel (10 days).

Table S3: Comparative encapsulation efficiency and degradation of curcumin in our studied eutectogels with other systems reported in the literature.

System	Curcumin encapsulation	Stability (With time)	Reference
CG Gel	45.60 mg/g	30 days	Present work
CF Gel	42.34 mg/g	30 days	Present work
(TBAC: DA)	5.9 (± 0.8) mg/ml	15 days	1
Choline chloride:sorbitol (1:1)	1.29 mg/ml	NA	2
Choline chloride:Fructose	3.11 mg/ml	NA	2

(1:1)			
Choline chloride:glucose (1:1)	2.90 mg/ml	NA	2
Choline chloride:sucrose (1:1)	0.27 mg/ml	NA	2
Choline chloride:maltose (1:1)	1.17 mg/ml	NA	2
Choline chloride: glycerol (1:1)	7.15 mg/ml	NA	2
CUR-ME-G	14.9 mg/g	NA	3
Car/Alg beads	10.16 ± 0.15 mg/g	NA	4
Cur-loaded SA hydrogel beads.	7.25 ± 3.16 mg/g	NA	5
CUR/CS-Alg-g-PF127	5mg/ml	NA	6
Alginate aldehyde–gelatin	72%*	NA	7
Alg-Ccm conjugare	NA	6h	8
CG/Alg nanoparticles	69%*	NA	9
ZIF-8@PCNF composite hydrogel	82%*	NA	10
CCM-ZIF-8-GA	90%*	NA	11
CCMZIF 0.008	87.1%*	4.2 time higher stable then only CCM (4 hour)	12
Cur-ZIF8-HA	96.11%*	48 hours	13
ZIF@CCM	82.76%*	NA	14



* No actual loading values are provided in the literature.

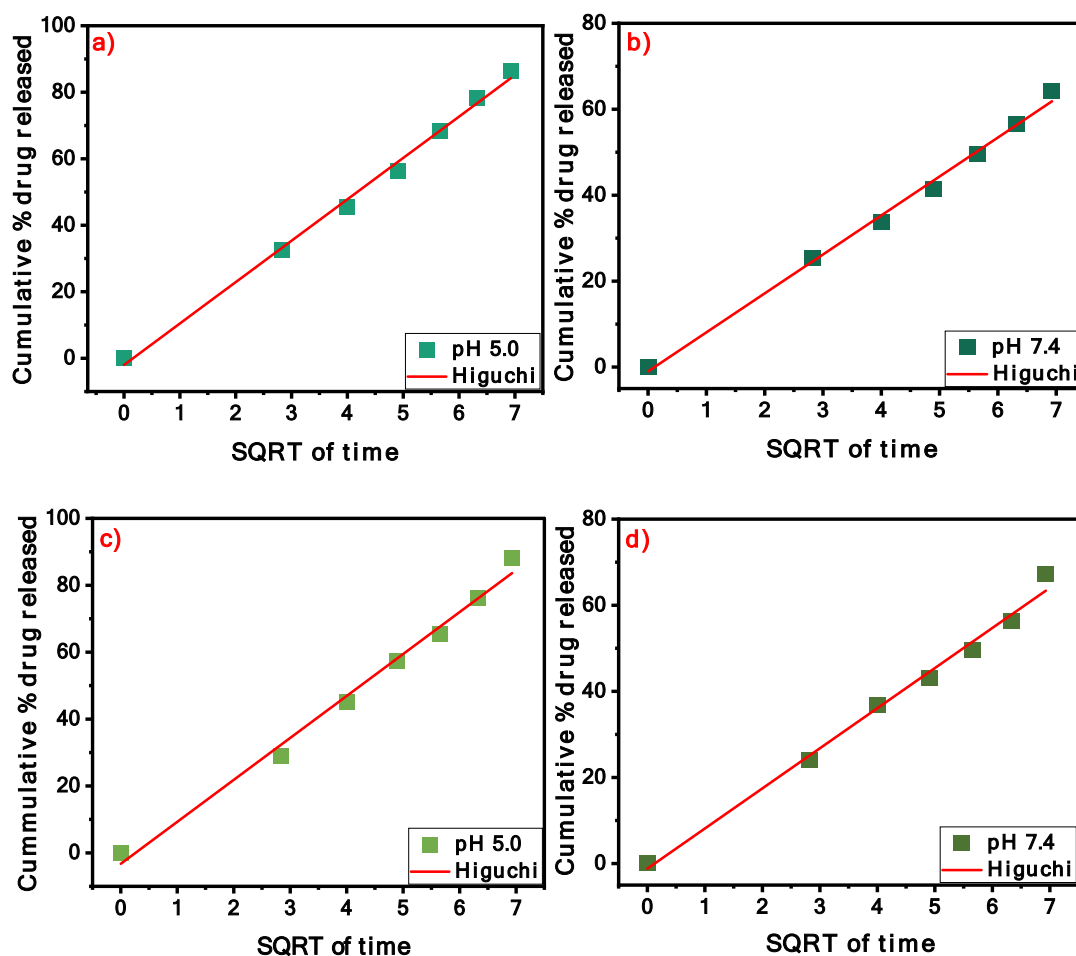


Figure S6: Linear fitting of curcumin release data for **a)** CG gel at pH 5.0, **b)** CG gel at pH 7.4, **c)** CF gel at pH 5.0 and, **d)** CF gel at pH 7.4.

Table S4: Kinetic model fitting in curcumin loaded CG and CF Gel at pH 7.4 and pH 5.0.

Kinetic model	CG Gel (pH 7.4)	CG Gel (pH 5.0)	CF Gel (pH 7.4)	CF Gel (pH 5.0)
Zero-order	$y = 1.2119x + 9.6207$ $R^2 = 0.9382$	$y = 1.6668x + 12.482$ $R^2 = 0.9432$	$y = 1.2441x + 9.7414$ $R^2 = 0.9344$	$y = 1.6932x + 11.001$ $R^2 = 0.954$
First-order	$y = -0.0086x +$	$y = -0.0171x +$	$y = -0.0091x +$	$y = -0.0176x +$

	1.9722 $R^2 = 0.9852$	2.0061 $R^2 = 0.983$	1.9737 $R^2 = 0.9755$	2.0225 $R^2 = 0.9597$
Higuchi	$y = 9.0707x - 0.9943$ $R^2 = 0.9946$	$y = 12.448x - 1.9965$ $R^2 = 0.9954$	$y = 9.3158x - 1.1727$ $R^2 = 0.9914$	$y = 12.542x - 3.2561$ $R^2 = 0.9905$
Hixcon-Crowell	$y = 0.0259x + 0.1229$ $R^2 = 0.9757$	$y = 0.0444x + 0.0944$ $R^2 = 0.9913$	$y = 0.0269x + 0.1212$ $R^2 = 0.9697$	$y = 0.0454x + 0.0549$ $R^2 = 0.9846$
Korsmeyer-Peppas	$y = 36.057x - 3.9235$ $R^2 = 0.9477$	$y = 49.418x - 5.9402$ $R^2 = 0.946$	$y = 37.064x - 4.2199$ $R^2 = 0.9464$	$y = 49.581x - 6.9811$ $R^2 = 0.9334$

Table S5: Antimicrobial activities of DES and their respective gel with and without curcumin.

System	B. subtilis	E. coli	S. aureus	S. typhi
CG system	6.5 ± 0.13	6.9 ± 0.13	6.6 ± 0.11	6.9 ± 0.14
	10.5 ± 0.21	10.5 ± 0.22	12 ± 0.24	12.5 ± 0.23
	24 ± 0.15	23.5 ± 0.15	25.5 ± 0.3	25 ± 0.48
CF system	6.5 ± 0.13	7 ± 0.14	6.5 ± 0.13	6.8 ± 0.12
	12.1 ± 0.24	11.5 ± 0.23	11.5 ± 0.23	11 ± 0.24
	24 ± 0.48	23.1 ± 0.19	22.5 ± 0.35	22 ± 0.45

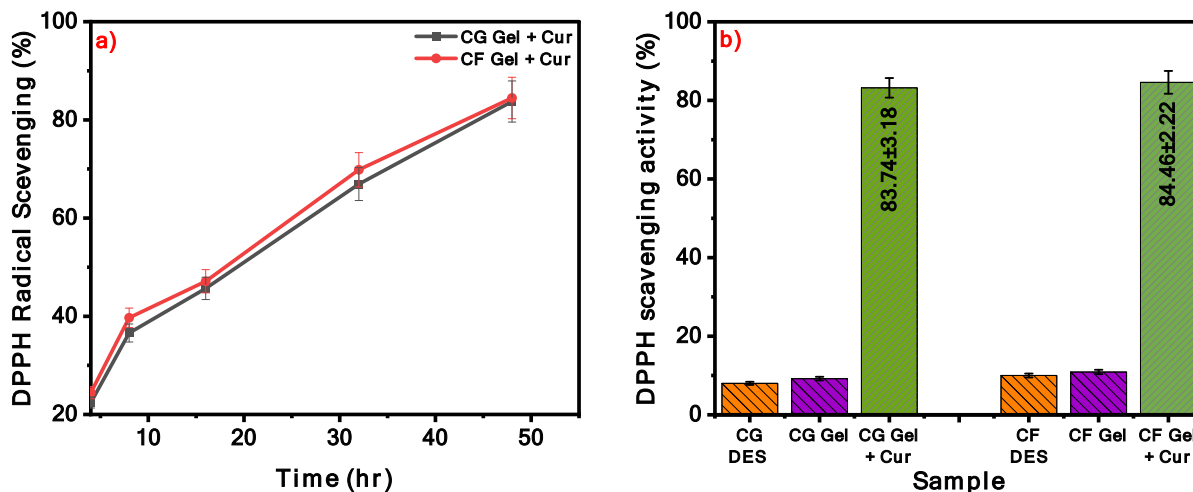


Figure S7: a) Time dependent DPPH radical scavenging (%) activities of Cur loaded gels (CG gel, CF gel) and; **b)** DPPH radical scavenging (%) activities of DESs and their respective gel with and without drug.

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