Supplementary Materials: Intact Fibrillated 3D-Printed Cellulose Macrofibrils/CaCO₃ for Controlled Drug Delivery

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Table S1. Volumetric flow rate, Q_v vs Extrusion Speed of CMF solution. The data are presented as mean ± standard deviation (n = 3).

Extrusion Speed (mm/s)	Volumetric Flow Rate, Q_v (mm³/s)
0.4174 ± 0.0125	7.2501 ± 0.2042
0.5146 ± 0.0134	8.5823 ± 0.3251
0.6091 ± 0.0113	9.5833 ± 0.2750
0.6862 ± 0.0206	11.1667 ± 0.3583
0.7855 ± 0.0186	13.0033 ± 0.4267

Table S2. Volumetric flow rate, Q_v vs Extrusion Speed of CMF solution. The data are presented as mean ± standard deviation (n = 3).

Print Speed, v (mm/s)	Print Line Width, x (mm)	Print Line Height, h (mm)	Print Cross-sectional Area, A (mm²)
5	1.68 ± 0.07	1.39 ± 0.06	1.84 ± 0.12
10	1.24 ± 0.09	0.95 ± 0.03	0.93 ± 0.15
15	0.92 ± 0.06	0.86 ± 0.04	0.62 ± 0.13
20	0.80 ± 0.05	0.78 ± 0.06	0.49 ± 0.12
25	0.71 ± 0.07	0.68 ± 0.04	0.37 ± 0.13
30	0.65 ± 0.09	0.63 ± 0.06	0.32 ± 0.14

Table S3. Mechanical Properties of 3D printed CMF. The data are presented as mean ± standard deviation (n = 3).

Samples	Tensile Strength (MPa)	Young's Modulus (GPa)	Strain %)
CMF 7%	48.96 ± 1.02	1.59 ± 0.06	7.54 ± 0.12
CMF 8%	58.89 ± 1.36	1.91 ± 0.07	8.05 ± 0.08
CMF 9%	66.70 ± 0.98	2.16 ± 0.06	8.76 ± 0.09

Table S4. 5-FU uptake data on CMF/CaCO₃ composite printed structure. The data are presented as mean ± standard deviation (n = 3).

Samples	C_0 (ppm)	C_e (ppm)	LE (%)	q_e (mg/g)
CMF		6.19	22.67 ± 0.53	0.91 ± 0.03
CMF:0.25CaCO₃		5.73	28.36 ± 0.41	1.13 ± 0.05
CMF:0.50CaCO₃	8	5.35	33.17 ± 0.62	1.33 ± 0.05
CMF:0.75CaCO₃		4.87	39.13 ± 0.56	1.57 ± 0.06
CMF:1.00CaCO₃		4.45	44.41 ± 0.47	1.78 ± 0.04
CaCO₃		1.40	82.49 ± 0.51	3.29 ± 0.07

C_0 and C_e, initial and equilibrium concentrations of the 5-FU (mg/L); LE, loading efficiency of 5-FU at final equilibrium; q_e, total uptake amount of 5-FU at final equilibrium.

Table S5. One way ANOVA results on kinetic release of 5-FU (%) for 24 h (Tukey Method; N: 3; α = 0.05; Equal variances were assumed for the analysis)

Samples	Time (h)	Mean	StDev	Grouping					
CaCO$_3$		29.493	0.300	A					
CMF:1.00CaCO$_3$		11.132	0.195		B				
CMF:0.75CaCO$_3$	1	9.687	0.230			C			
CMF:0.50CaCO$_3$		8.857	0.241				D		
CMF:0.25CaCO$_3$		7.345	0.115					E	
CMF		7.286	0.100					E	
CaCO$_3$		48.941	0.201	A					
CMF:1.00CaCO$_3$		14.468	0.331		B				
CMF:0.75CaCO$_3$	2	12.454	0.250			C			
CMF:0.50CaCO$_3$		11.187	0.248				D		
CMF:0.25CaCO$_3$		10.185	0.209					E	
CMF		9.314	0.187						F
CaCO$_3$		59.610	0.262	A					
CMF:1.00CaCO$_3$		18.157	0.431		B				
CMF:0.75CaCO$_3$	3	16.289	0.326			C			
CMF:0.50CaCO$_3$		14.257	0.348				D		
CMF:0.25CaCO$_3$		13.331	0.300					E	
CMF		12.398	0.180						F
CaCO$_3$		67.096	0.202	A					
CMF:1.00CaCO$_3$		22.081	0.431		B				
CMF:0.75CaCO$_3$	4	19.083	0.382			C			
CMF:0.50CaCO$_3$		17.456	0.379				D		
CMF:0.25CaCO$_3$		16.055	0.322					E	
CMF		15.191	0.220					E	
CaCO$_3$		75.943	0.221	A					
CMF:1.00CaCO$_3$		24.931	0.489		B				
CMF:0.75CaCO$_3$	5	22.685	0.287			C			
CMF:0.50CaCO$_3$		21.022	0.384				D		
CMF:0.25CaCO$_3$		19.648	0.274					E	
CMF		19.015	0.276					E	
CaCO$_3$		82.122	0.295	A					
CMF:1.00CaCO$_3$		30.764	0.188		B				
CMF:0.75CaCO$_3$	6	26.213	0.382			C			
CMF:0.50CaCO$_3$		22.888	0.430				D		
CMF:0.25CaCO$_3$		21.109	0.295					E	
CMF		19.929	0.289						F
CaCO$_3$		85.992	0.280	A					
CMF:1.00CaCO$_3$		34.391	0.214		B				
CMF:0.75CaCO$_3$	7	30.491	0.140			C			
CMF:0.50CaCO$_3$		24.269	0.283				D		
CMF:0.25CaCO$_3$		22.570	0.315					E	
CMF		20.386	0.296						F

CaCO$_3$		87.579	0.340	A			
CMF:1.00CaCO$_3$		38.623	0.245		B		
CMF:0.75CaCO$_3$	8	33.598	0.154			C	
CMF:0.50CaCO$_3$		27.465	0.333				D
CMF:0.25CaCO$_3$		24.793	0.278					E
CMF		21.152	0.256						F

CaCO$_3$		88.556	0.350	A			
CMF:1.00CaCO$_3$		40.995	0.262		B		
CMF:0.75CaCO$_3$	9	36.758	0.169			C	
CMF:0.50CaCO$_3$		30.592	0.262				D
CMF:0.25CaCO$_3$		27.200	0.305					E
CMF		22.214	0.322						F

CaCO$_3$		88.742	0.430	A			
CMF:1.00CaCO$_3$		44.717	0.290		B		
CMF:0.75CaCO$_3$	10	39.339	0.268			C	
CMF:0.50CaCO$_3$		32.404	0.282				D
CMF:0.25CaCO$_3$		28.366	0.318					E
CMF		22.763	0.330						F

CaCO$_3$		88.887	0.315	A			
CMF:1.00CaCO$_3$		47.461	0.311		B		
CMF:0.75CaCO$_3$	11	40.916	0.279			C	
CMF:0.50CaCO$_3$		34.905	0.310				D
CMF:0.25CaCO$_3$		30.554	0.343					E
CMF		23.677	0.343						F

CaCO$_3$		88.926	0.258	A			
CMF:1.00CaCO$_3$		49.709	0.445		B		
CMF:0.75CaCO$_3$	12	45.184	0.207			C	
CMF:0.50CaCO$_3$		37.468	0.340				D
CMF:0.25CaCO$_3$		32.085	0.360					E
CMF		24.285	0.275						F

CaCO$_3$		88.9741	0.118	A			
CMF:1.00CaCO$_3$		52.438	0.349		B		
CMF:0.75CaCO$_3$	13	47.385	0.357			C	
CMF:0.50CaCO$_3$		40.481	0.289				D
CMF:0.25CaCO$_3$		32.989	0.299					E
CMF		25.437	0.283						F

CaCO$_3$		89.0158	0.135	A			
CMF:1.00CaCO$_3$		55.974	0.377		B		
CMF:0.75CaCO$_3$	14	50.007	0.202			C	
CMF:0.50CaCO$_3$		42.354	0.306				D
CMF:0.25CaCO$_3$		34.883	0.316					E
CMF		25.811	0.275						F

$CaCO_3$		89.062	0.167	A				
$CMF:1.00CaCO_3$		58.166	0.386		B			
$CMF:0.75CaCO_3$	15	52.186	0.325			C		
$CMF:0.50CaCO_3$		45.413	0.335				D	
$CMF:0.25CaCO_3$		36.048	0.327					E
CMF		26.084	0.278						F
$CaCO_3$		89.092	0.240	A				
$CMF:1.00CaCO_3$		60.068	0.409		B			
$CMF:0.75CaCO_3$	16	54.754	0.221			C		
$CMF:0.50CaCO_3$		48.534	0.257				D	
$CMF:0.25CaCO_3$		37.213	0.337					E
CMF		26.358	0.281						F
$CaCO_3$		89.112	0.230	A				
$CMF:1.00CaCO_3$		63.324	0.435		B			
$CMF:0.75CaCO_3$	17	56.846	0.256			C		
$CMF:0.50CaCO_3$		51.089	0.274				D	
$CMF:0.25CaCO_3$		39.325	0.356					E
CMF		26.905	0.287						F
$CaCO_3$		89.131	0.205	A				
$CMF:1.00CaCO_3$		67.112	0.332		B			
$CMF:0.75CaCO_3$	18	58.427	0.263			C		
$CMF:0.50CaCO_3$		52.897	0.286				D	
$CMF:0.25CaCO_3$		41.409	0.332					E
CMF		27.270	0.291						F
$CaCO_3$		89.132	0.212	A				
$CMF:1.00CaCO_3$		70.210	0.490		B			
$CMF:0.75CaCO_3$	19	61.064	0.275			C		
$CMF:0.50CaCO_3$		54.705	0.298				D	
$CMF:0.25CaCO_3$		43.112	0.391					E
CMF		27.562	0.271						F
$CaCO_3$		89.159	0.226	A				
$CMF:1.00CaCO_3$		72.676	0.510		B			
$CMF:0.75CaCO_3$	20	62.667	0.440			C		
$CMF:0.50CaCO_3$		55.453	0.303				D	
$CMF:0.25CaCO_3$		45.078	0.408					E
CMF		27.918	0.286						F
$CaCO_3$		89.156	0.240	A				
$CMF:1.00CaCO_3$		74.196	0.376		B			
$CMF:0.75CaCO_3$	21	63.131	0.222			C		
$CMF:0.50CaCO_3$		56.201	0.308				D	
$CMF:0.25CaCO_3$		47.216	0.354					E
CMF		28.091	0.300						F

CaCO$_3$		89.162	0.193	A				
CMF:1.00CaCO$_3$		74.956	0.528		B			
CMF:0.75CaCO$_3$	22	64.228	0.290			C		
CMF:0.50CaCO$_3$		57.270	0.316				D	
CMF:0.25CaCO$_3$		48.064	0.436					E
CMF		28.201	0.277						F
CaCO$_3$		89.162	0.222	A				
CMF:1.00CaCO$_3$		75.701	0.534		B			
CMF:0.75CaCO$_3$	23	64.926	0.396			C		
CMF:0.50CaCO$_3$		57.276	0.316				D	
CMF:0.25CaCO$_3$		48.218	0.338					E
CMF		28.273	0.302						F
CaCO$_3$		89.172	0.229	A				
CMF:1.00CaCO$_3$		75.705	0.534		B			
CMF:0.75CaCO$_3$	24	64.914	0.293			C		
CMF:0.50CaCO$_3$		57.281	0.316				D	
CMF:0.25CaCO$_3$		48.379	0.321					E
CMF		28.393	0.267						F

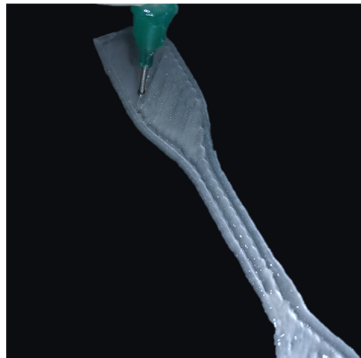*Means that do not share a letter are significantly different; $p < 0.05$

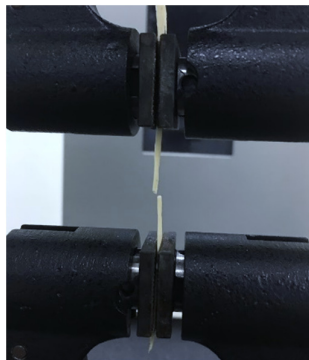**Figure S1.** Printing of CMF tensile sample according to ASTM D638 Type IV.

Figure S2. Tensile testing of regenerated and dried printed CMF tensile sample.
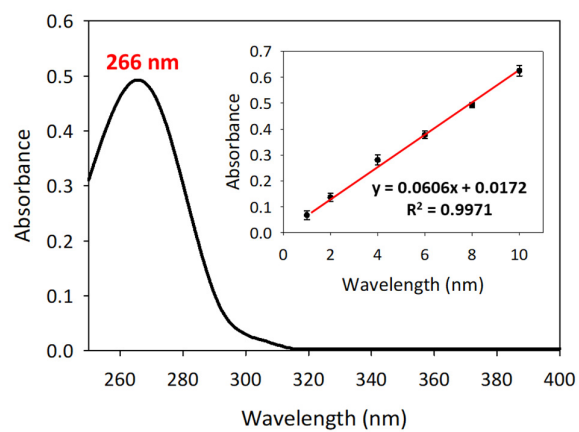
Figure S3. (a) UV Spectrum of 5-fluorouracil (5-FU) (inset shows linear calibration curve according to the Beer-Lambert law). The data are presented as mean ± standard deviation (n = 3).