

Electronic Supplementary Information

Biomimetic Spinning of Silk Fibers and *in situ* Cell Encapsulation

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Fig. S1-S6
Table S1-S2

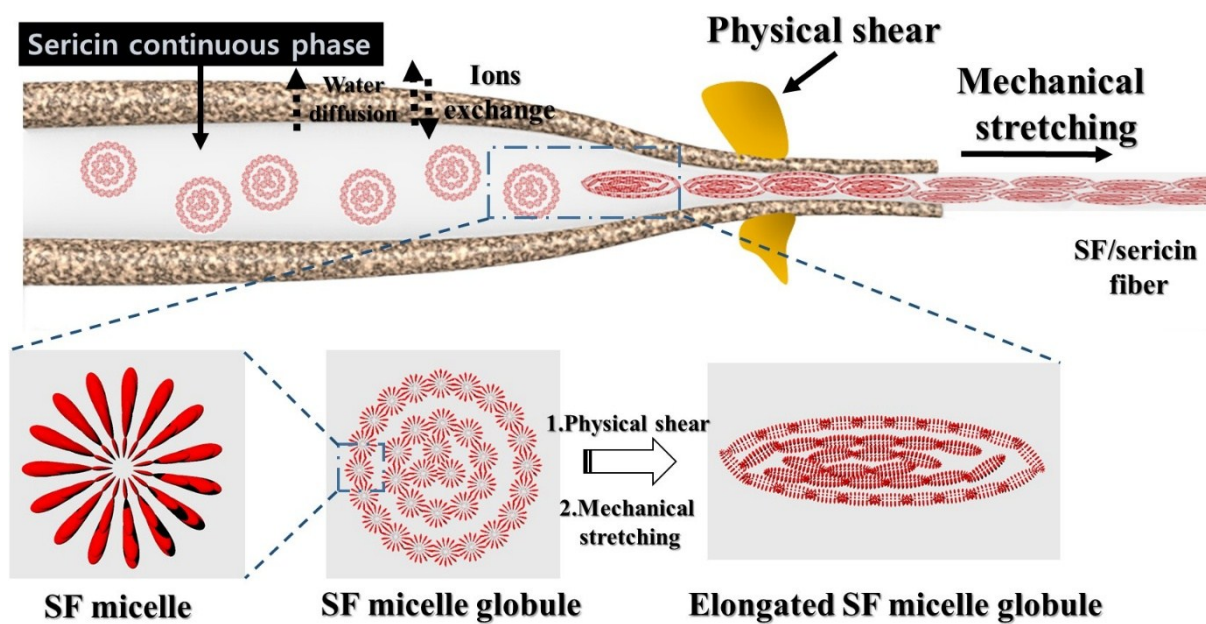
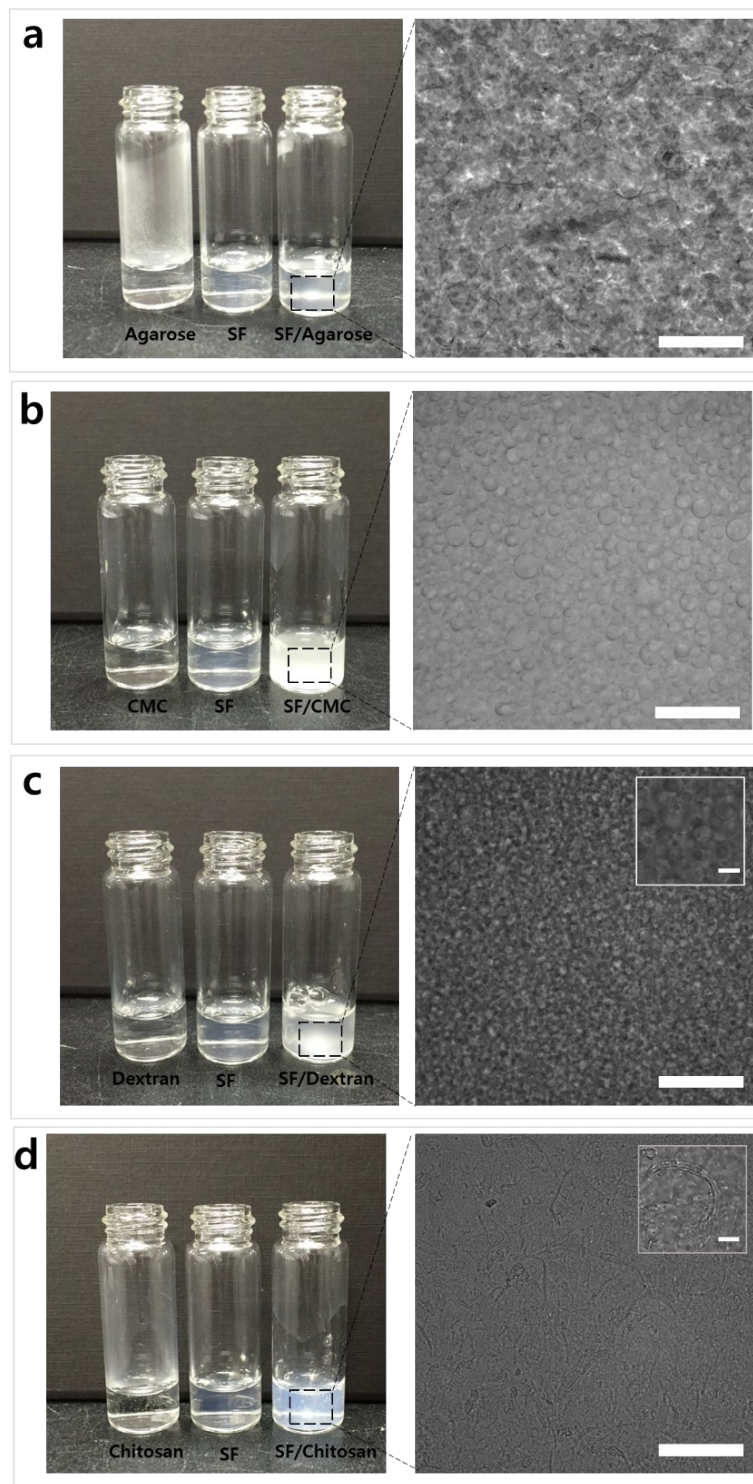


Fig. S1 Natural spinning process inside a *B. mori* duct. SF micelle globules were elongated and entangled each other under the combined action of the physical shearing force and mechanical stretching.



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2 **Fig.S2** Photographic and optical images show that phase separation occurred upon mixing of SF and
 3 various polysaccharides. a) Agarose/SF blend. b) CMC/SF blend. c) Dextran/SF blend. d) Chitosan/SF
 4 blend. The insets in (c and d) were the enlarged images. Scale bars: 200 μm (a, b, c, d) and 25 μm (insets
 5 in c and d).

Table S1 Comparison of polysaccharides possible to induce phase-separation of silk fibroin.

	Biocompatibility	Biodegradability (mechanism)	Mechanical property	Process to prepare hydrogel	Possibility of in situ cell encapsulation
Alginate ^[1, 2]	Good	Good (Enzymatic)	Weak	Neutral condition/ionic crosslinking	Yes
Chitosan ^[2, 3]	Good	Good (Enzymatic)	Weak	Acidic condition /ionic-chemical crosslinking	Possible, but need to modify polymer chain
Agarose ^[4]	Good	Good (not clear)	Weak	Neutral condition /heating-cooling crosslinking	Possible, but difficult
Dextran ^[5]	Good	Good (Enzymatic)	Weak	Neutral condition /chemical crosslinking	Possible, but need to modify polymer chain
Carboxy ^[6] methyl cellulose	Good	Good (Enzymatic)	Strong	Neutral condition /chemical crosslinking	Possible, but need to modify polymer chain

[1] J. L. Drury, D. J. Mooney, *Biomaterials* 2003, 24, 4337.

[2] K. Y. Lee, D. J. Mooney, *Chemical Reviews* 2001, 101, 1869.

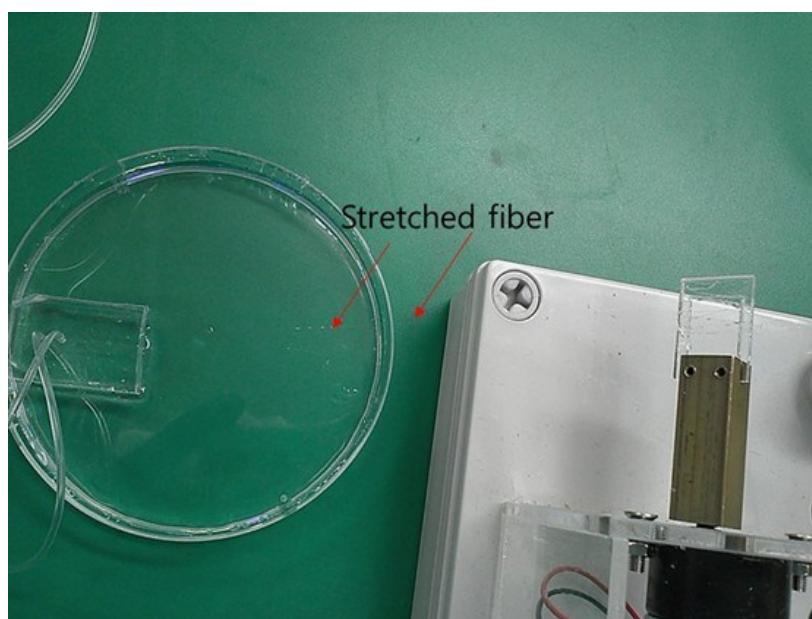
[3] J. Berger, M. Reist, J. M. Mayer, O. Felt, N. A. Peppas, R. Gurny, *European Journal of
Pharmaceutics and Biopharmaceutics* 2004, 57, 19.

[4] P. J. Emans, L. W. van Rhijn, T. J. M. Welting, A. Cremers, N. Wijnands, F. Spaapen, J. W.
Voncken, V. P. Shastri, *Proceedings of the National Academy of Sciences* 2010, 107, 3418; R. L.
Mauck, X. Yuan, R. S. Tuan, *Osteoarthritis and Cartilage* 2006, 14, 179.

[5] K. T. Nguyen, J. L. West, *Biomaterials* 2002, 23, 4307; Y. Liu, M. B. Chan-Park, *Biomaterials*
2009, 30, 196.

[6] C. Chang, L. Zhang, *Carbohydrate Polymers* 2011, 84, 40.

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Fig. S3 Fiber stretching by winding on the spool.

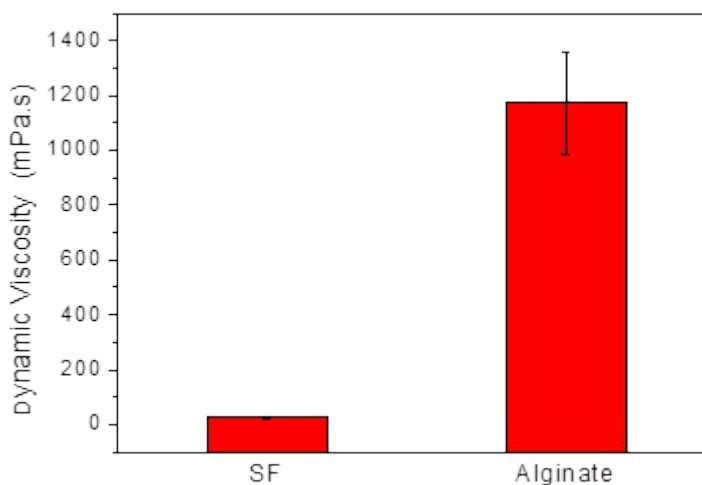
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1 **Table S2** Fabrication parameters for microfibers.

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Fiber code	Weight ratio (SF/alginate)	Flow conditions (ml/h)		Crosslinking method	Continuous fiber	Stretchable fiber	Continuous SF-only fiber
		Sheath	Core				
Alginate	1	30, 50,100	0.6, 1.2, 1.8	CaCl ₂ (3%, wt%)	Yes	Yes	No
SF1.25 /A1	1.25/1	30, 50,100	0.6, 1.2, 1.8	CaCl ₂ (3%, wt%)	Yes	Yes	No
SF3.5/A1	3.5/1	30, 50,100	0.6, 1.2, 1.8	CaCl ₂ (3%, wt%)	Yes	Yes	No
SF8/A1	8/1	30, 50,100	0.6, 1.2, 1.8	CaCl ₂ (3%, wt%)	Yes	Yes	No
SF10/A1	10/1	30, 50,100	0.6, 1.2, 1.8	CaCl ₂ (3%, wt%)	Yes	Yes	No
SF15/A1	15/1	30, 50,100	0.6, 1.2, 1.8	CaCl ₂ (3%, wt%)	Yes	Yes	Yes
SF30/A1	30/1	30, 50,100	0.6, 1.2, 1.8	CaCl ₂ (3%, wt%)	Yes	Yes	Yes
SF50/A1	50/1	30, 50,100	0.6, 1.2, 1.8	CaCl ₂ (3%, wt%)	Yes	No	Yes

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5 **Fig S4.** Viscosities of SF (5%) and alginate (5%) measured using a Brookfield viscometer.

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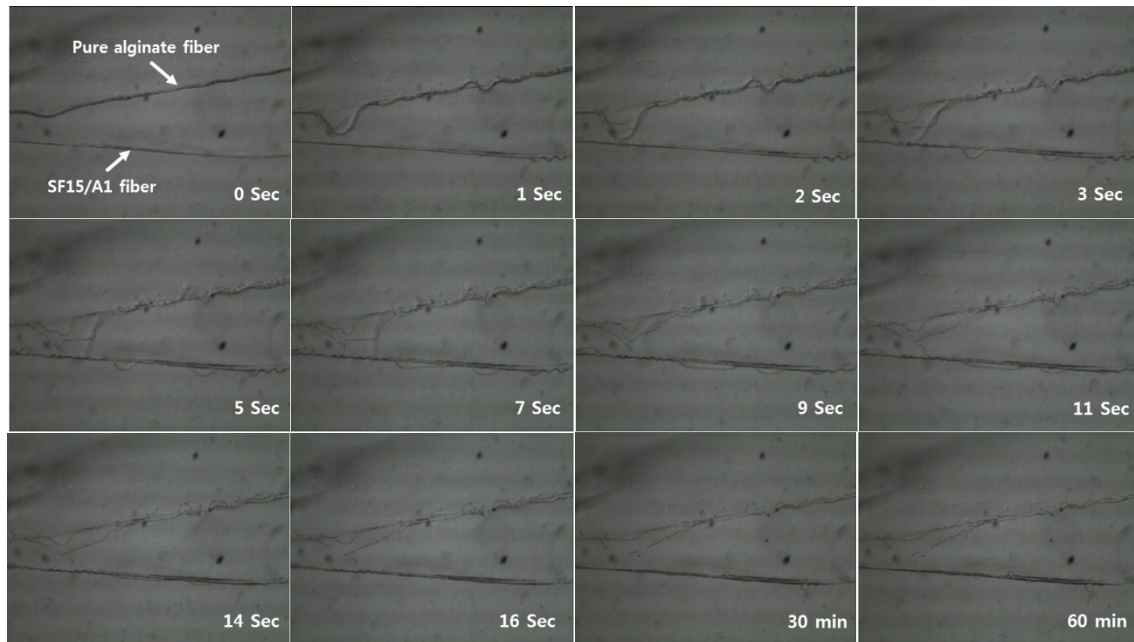


Fig. S5 Snap shots of microscopic images showing that pure alginate fiber rapidly degraded after addition of sodium citrate, on the other hand, SF15/A1 fiber remained intact.

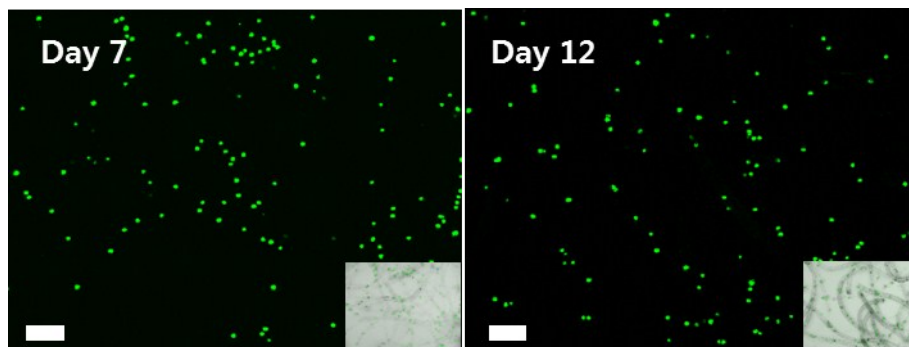


Fig. S6 Live/Dead stain of cells in SF1.25/A1 fibers after 7 and 12 days. Scale bars: 100 μm .