High Strength Chitosan Hydrogels with Biocompatibility via New Avenue Based on Constructing Nanofibrous Architecture

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Keywords: chitosan nanofibers, alkali/urea solvent, high strength hydrogels, biocompatibility, smart drug release

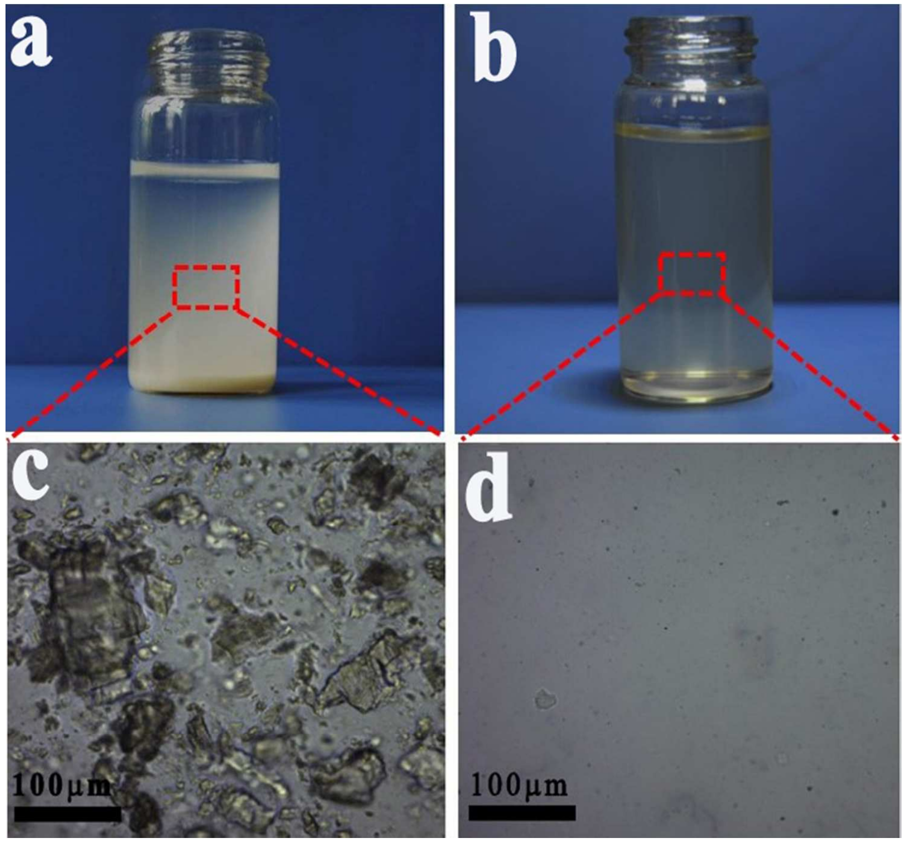

Figure S1. Chitosan powders dispersed in alkali/urea aqueous solution before (a, c) and after (b, d) freezing-thawing process.

Table S1. Chemical shift of chitosan powder and different chitosan solutions from ^{13}C NMR

Sample/Solvent	Chemical Shift (ppm)					
	C1	C4	C5	C3	C6	C2
CS powder[a]	105.53	83.26	75.76	75.76	62.29	57.98
Alkaline system[b]	101.78	78.42	76.42	73.79	60.34	57.09
Acidic system[b]	97.91, 96.83	76.42	74.87	70.38, 69.30	59.56	56.16, 54.92

[a] Results from solid state ^{13}C NMR. [b] Results from liquid ^{13}C NMR

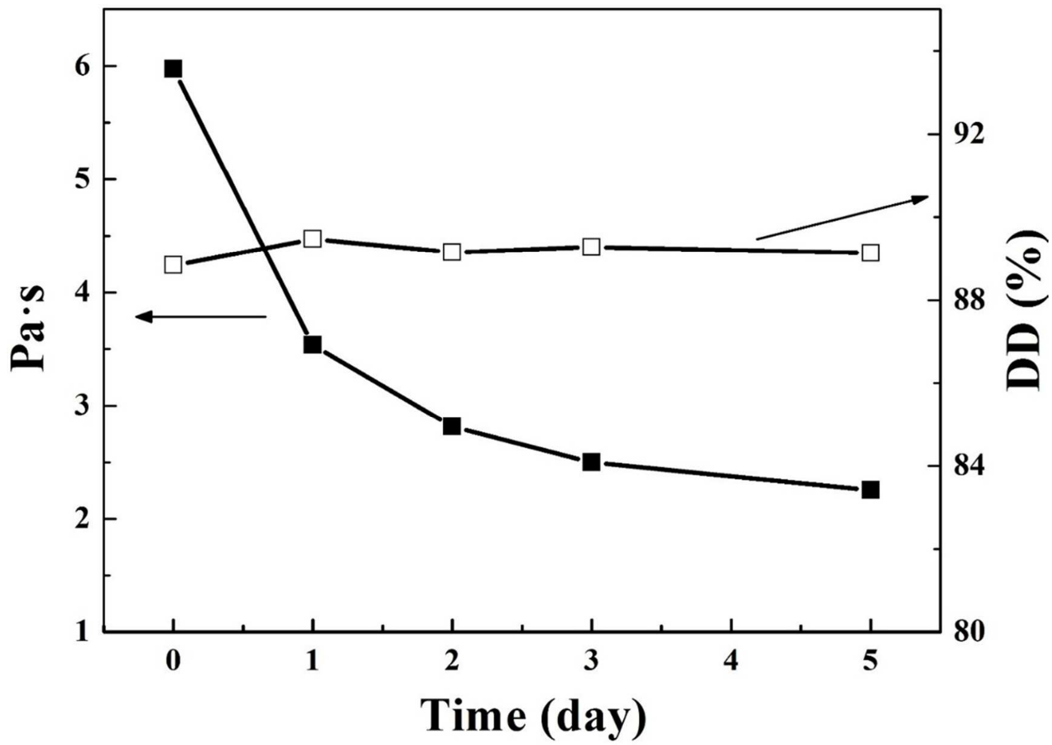

Figure S2. Dependence of zero-shear viscosity and DD of chitosan dissolved in alkaline solution on its storage time.

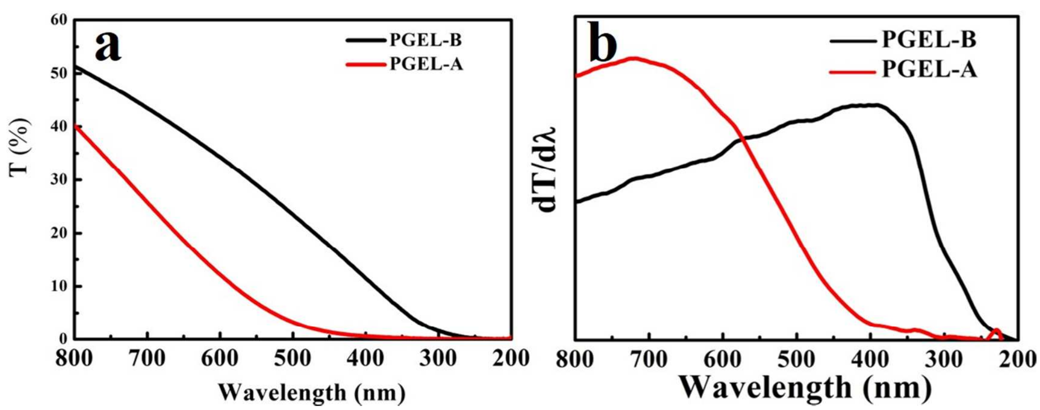

Figure S3. The light transmittance curves (a) and their differential curves (b) of the two kind of chitosan hydrogels in the range of visible light.

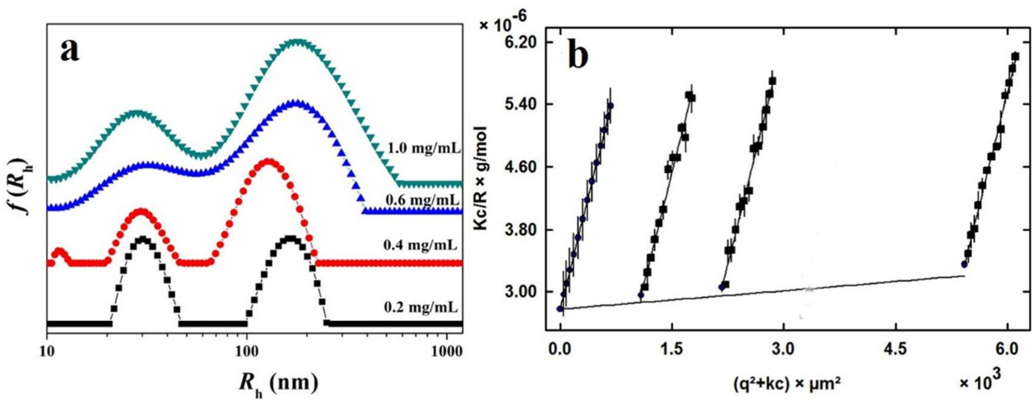

Figure S4. The hydrodynamic radius distributions (a) and Zimm plots (b) of chitosan alkaline solution at 10 ^{o}C.

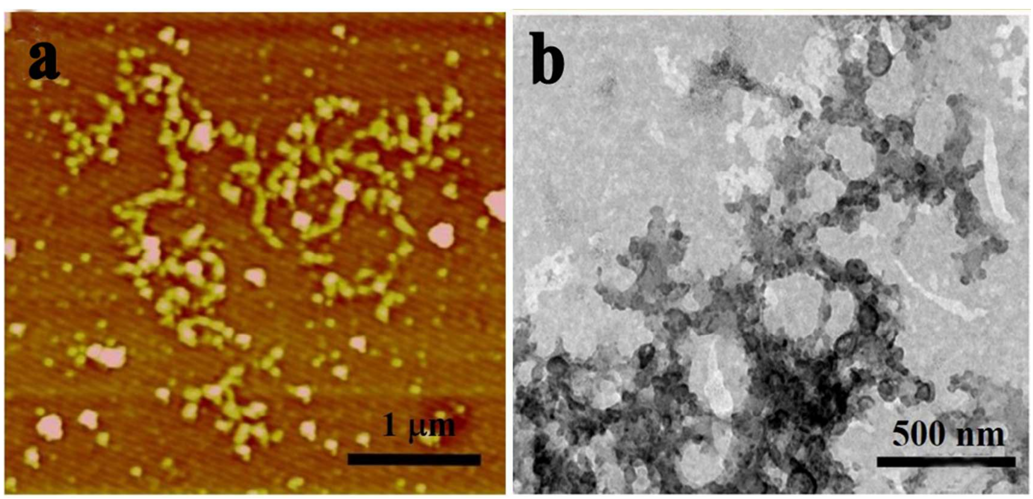

Figure S5. AFM image (a) and TEM image (b) of chitosan aggregates regenerated by sodium hydroxide from dilute chitosan acidic solution with a concentration of 1x10^{-4} g/mL.

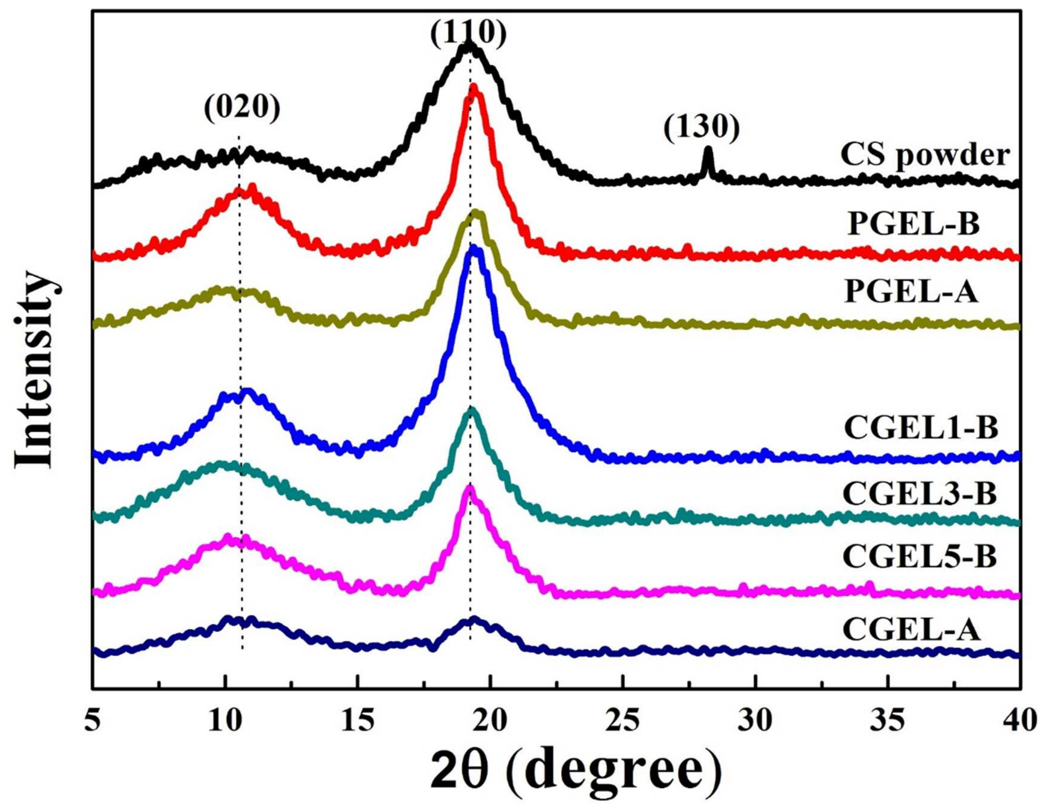

Figure S6. The XRD patterns of chitosan powders, chitosan physical and chemical hydrogel constructed form the two solvent systems.

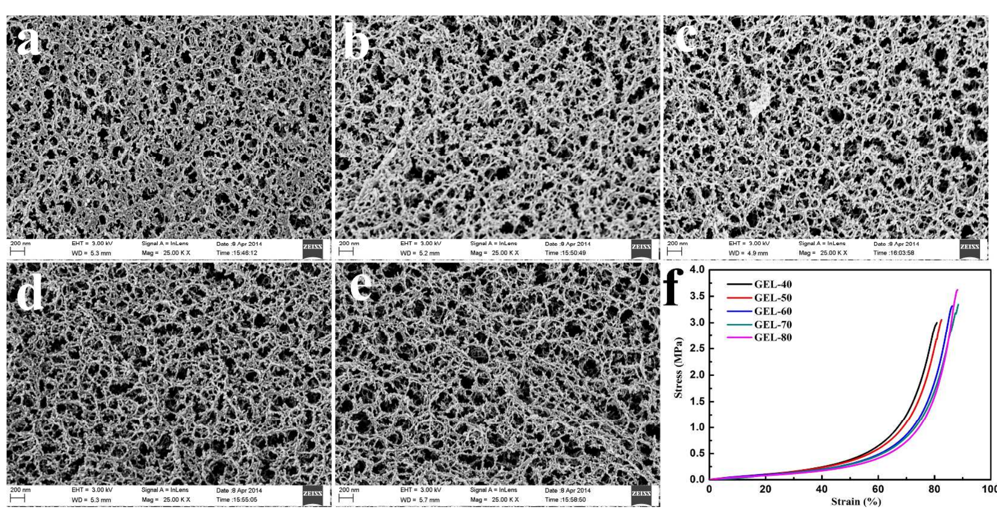

Figure S7. SEM images of chitosan physical hydrogel regenerated from hot water with different temperature (from 40, 50, 60, 70, to 80 ^{o}C for a ~ e). The compressive stress-strain curves (f) of chitosan hydrogels regenerated from hot water with different temperature.

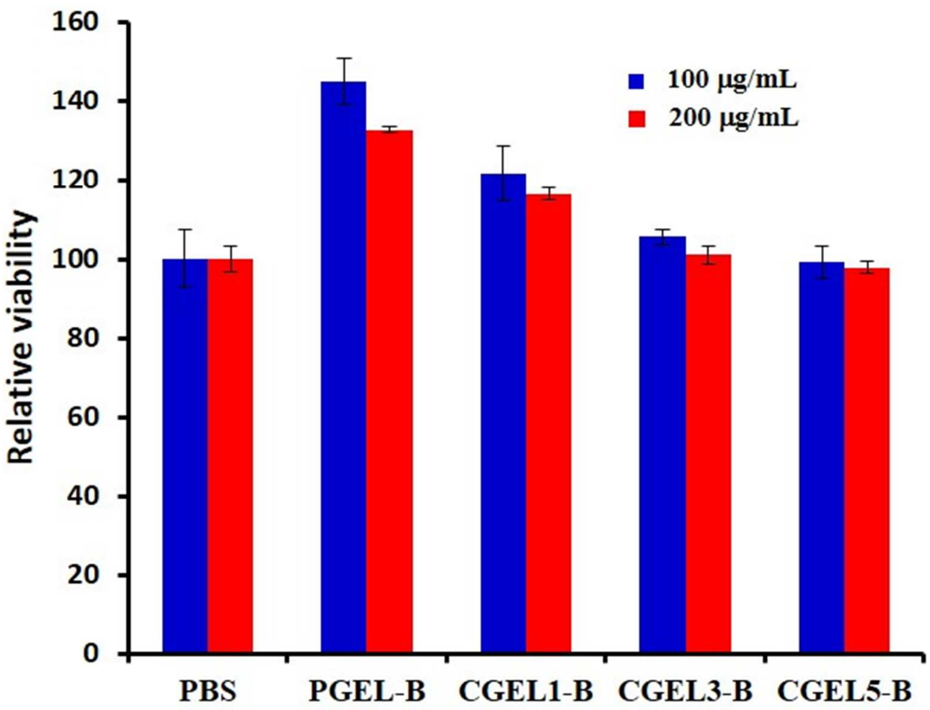

Figure S8. The C2C12 cells vitro cytotoxicity tests on different chitosan hydrogel samples.

Table S2. Comparison of compression mechanical properties of different high strength hydrogels

Samples	σ (MPa)	ε (%)	E (MPa)	Sources
PGEL-B	**3.31**	**85.9**	**0.60**	**This work**
CGEL3-B	**4.25**	**82.8**	**0.34**	**This work**
GO/PVA	**1.3**			***Ref. 1***
Tetra-PEG	**2.7**	**85**		***Ref. 2***
Agarose	**0.28**	**20**		***Ref. 2***
Cellulose	**0.44**	**55**		***Ref. 3***
Chitin/chitosan	**0.135**		**0.169**	***Ref. 4***
Cellulose nanofiber	**5.4**		**35.2**	***Ref. 5***
BC/Gelatin	**5.3**	**3.9**	**44**	***Ref. 6***

References

1. Zhang, L.; Wang, Z.; Xu, C.; Li, Y.; Gao, J.; Wang, W.; Liu, Y. *J. Mater. Chem.* *2011*, *21*, 10399-10406.

2. Sakai, T.; Matsunaga, T.; Yamamoto, Y.; Ito, C.; Yoshida, R.; Suzuki, S.; Sasaki, N.; Shibayama, M.; Chung, U. *Macromolecules* **2008**, *41*, 5379-5384

3. He, M.; Zhao, Y.; Duan, J.; Wang, Z.; Chen, Y.; Zhang, L. *ACS Appl. Mater. Interfaces*. **2014**, *6*, 1872−1878.

4. Araki, J.; Yamanaka, Y.; Ohkawa, K. *Polym. J.* **2012**, *44*, 713–717

5. Abe, K.; Yano, H. *Cellulose* **2012**, *19*, 1907–1912.

6. Nakayama, A.; Kakugo, A.; Gong, J. P. Osada, Y.; Takai, M.; Erata, T.; Kawano, S. Adv. Funct. Mater. **2004**, *14*, 1124-1128.