## **Supporting Information**

## MRGSHHHHHHGS

MDTYKLILNGKTLKGETTTEAVDAATAEKVFKQYANDNGVDGEWTYDDATKTFTVTERS
MDTYKLILNGKTLKGETTTEAVDAATAEKVFKQYANDNGVDGEWTYDDATKTFTVTERS
MDTYKLILNGKTLKGETTTEAVDAATAEKVFKQYANDNGVDGEWTYDDATKTFTVTERS
MDTYKLILNGKTLKGETTTEAVDAATAEKVFKQYANDNGVDGEWTYDDATKTFTVTERS
MDTYKLILNGKTLKGETTTEAVDAATAEKVFKQYANDNGVDGEWTYDDATKTFTVTERS
MDTYKLILNGKTLKGETTTEAVDAATAEKVFKQYANDNGVDGEWTYDDATKTFTVTERS
MDTYKLILNGKTLKGETTTEAVDAATAEKVFKQYANDNGVDGEWTYDDATKTFTVTERS
MDTYKLILNGKTLKGETTTEAVDAATAEKVFKQYANDNGVDGEWTYDDATKTFTVTERS

Figure S1. The amino acid sequence of (GB1)<sub>8</sub> polyprotein. There are six lysine residues in each GB1 domain, providing great possibility for derivatizing (GB1)<sub>8</sub> using BH reagent to increase the phenolic content of (GB1)<sub>8</sub> and increase the crosslinking density of the resultant protein hydrogel.

Table S1. The molecular weight of (GB1)<sub>8</sub> and derivatized (GB1)<sub>8</sub> determined via MALDI-MS

Sample	Molecule weight (Dalton)					Number of
						modified K
	Calculated	Tested				
(GB1) <sub>8</sub>	53911.95	53876.1	53957.2			
(GB1) <sub>8</sub> -	54060.01	53994.6	54016.8			~ 1
Rx1						
(GB1) <sub>8</sub> -	55096.43	54749.9	54751.9	54759.8		5~6
Rx2						
(GB1) <sub>8</sub> -	56132.85	54990.4	55648.4	55459	55472.2	10~11
Rx3						

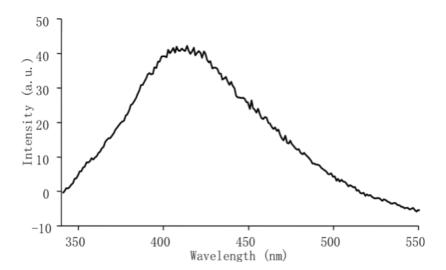


Figure S2. Fluorescence spectrum of acid hydrolysis product of (GB1)<sub>8</sub>-Rx2-based hydrogel in 100 mM NaHCO<sub>3</sub>-Na<sub>2</sub>CO<sub>3</sub> buffer (pH 9.9). The fluorescence emission peak at ~410 nm is identical to that of purified dityrosine, indicating the presence of dityrosine-like crosslinks in the hydrogel.

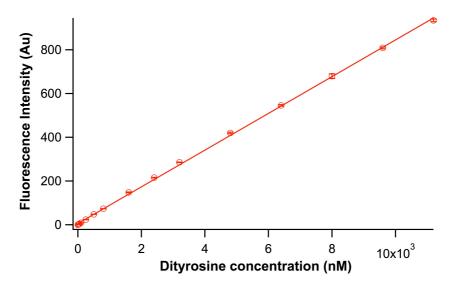


Figure S3. Standard fluorescence-concentration curve of dityrosine in 100 mM NaHCO<sub>3</sub>-Na<sub>2</sub>CO<sub>3</sub> buffer (pH 9.9). Error bars indicate standard deviation. The fluorescence intensity of dityrosine compound at 410 nm is linearly proportional to its concentration within the tested concentration range. This standard curve is used to determine the concentration of dityrosine-like crosslinks in the hydrogels.