Electronic Supplementary Material (ESI) for Green Chemistry.

Supporting Information

Fabrication of robust protein-based foams with multifunctionality by manipulating intermolecular interactions

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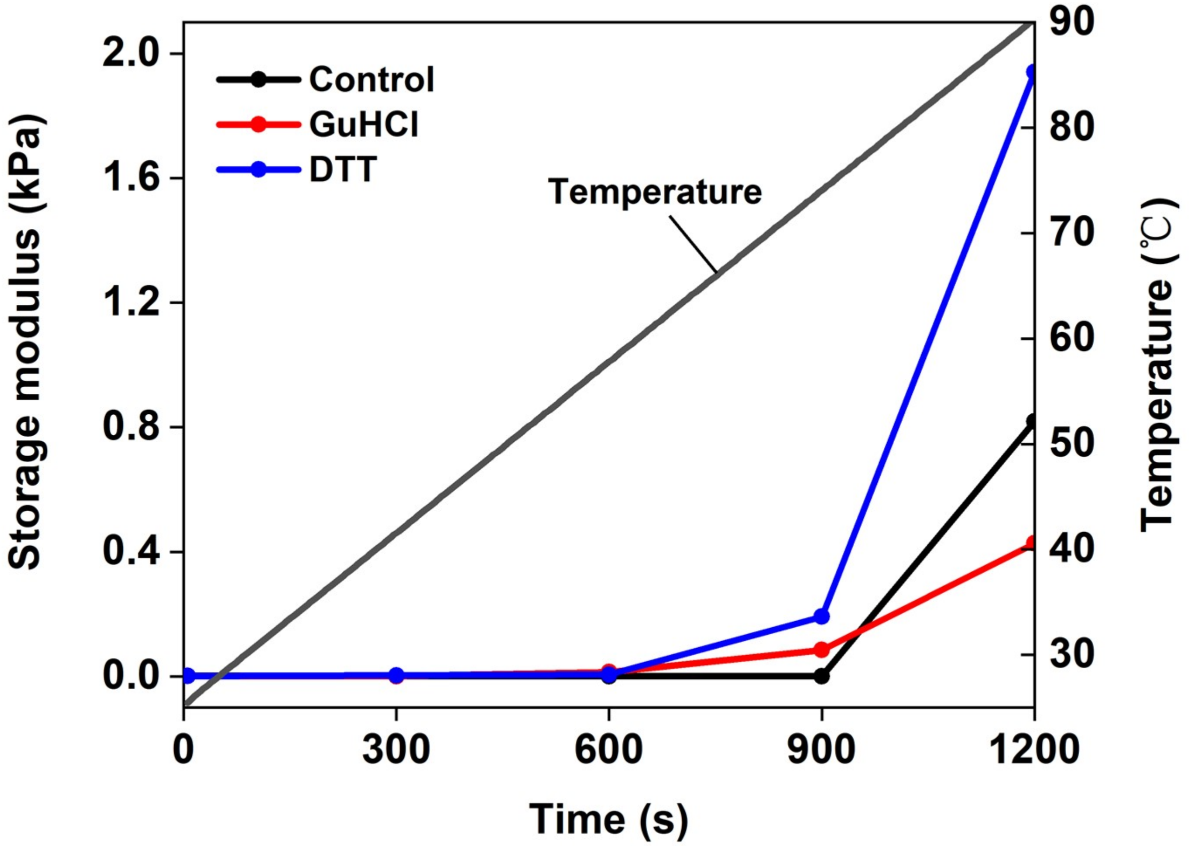

Fig. S1 Shear modulus as a function of time during the heating process (25–90 °C) of whey protein emulsions. The control, guanidinium hydrochloride (GuHCl), and dithiothreitol (DTT) samples contained 200 mM NaCl, 200 mM NaCl + 2 M GuHCl, and 200 mM NaCl + 10 mM DTT, respectively.

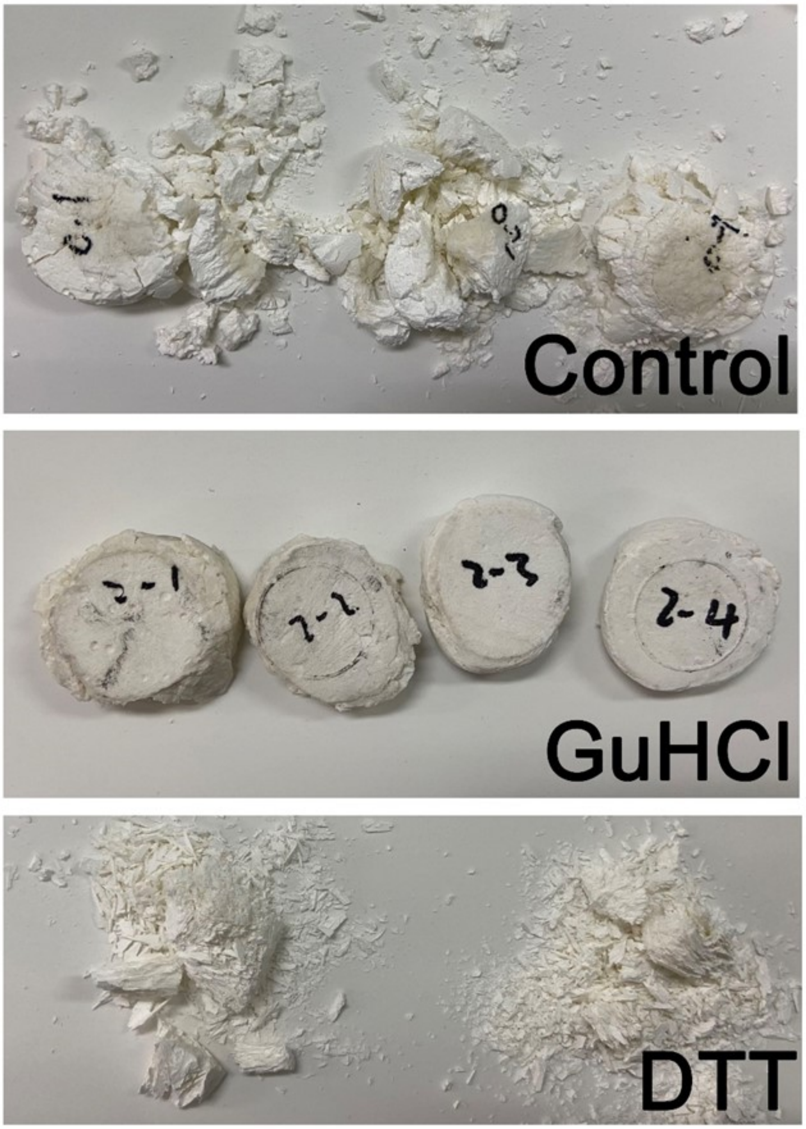

Fig. S2 Photographs of foams upon 50% deformation during the compression test. The control, guanidinium hydrochloride (GuHCl), and dithiothreitol (DTT) samples contained 200 mM NaCl, 200 mM NaCl + 2 M GuHCl, and 200 mM NaCl + 10 mM DTT, respectively.

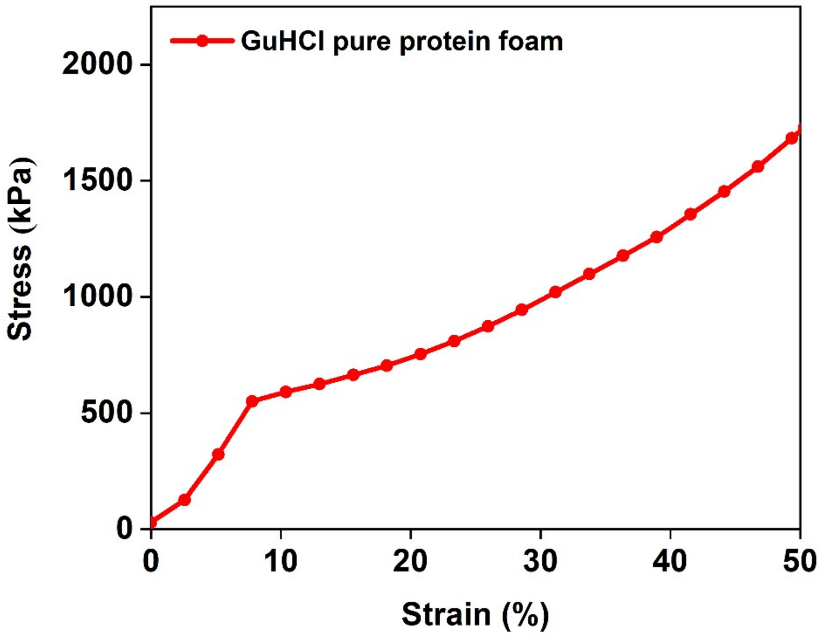

Fig. S3 Stress–strain curve of the guanidinium hydrochloride (GuHCl) pure protein foam during the compression test. The sample contained 200 mM NaCl + 2 M GuHCl.

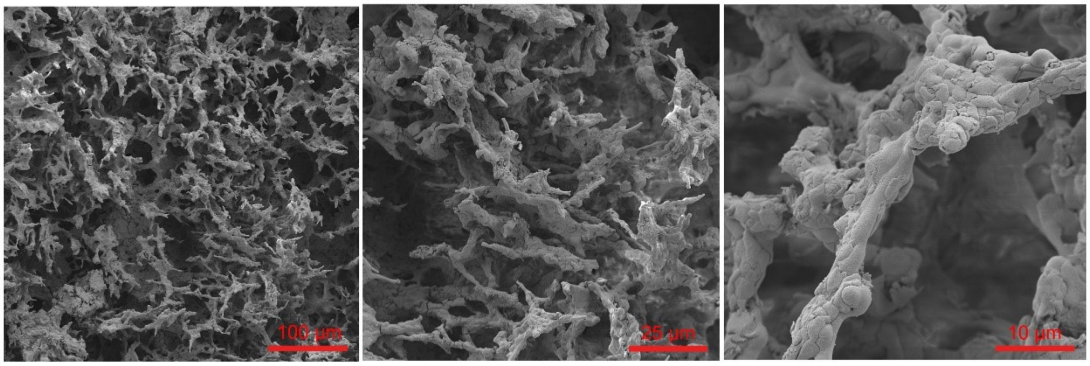

Fig. S4 Scanning electron micrographs of the guanidinium hydrochloride (GuHCl) pure protein foam at different magnifications.

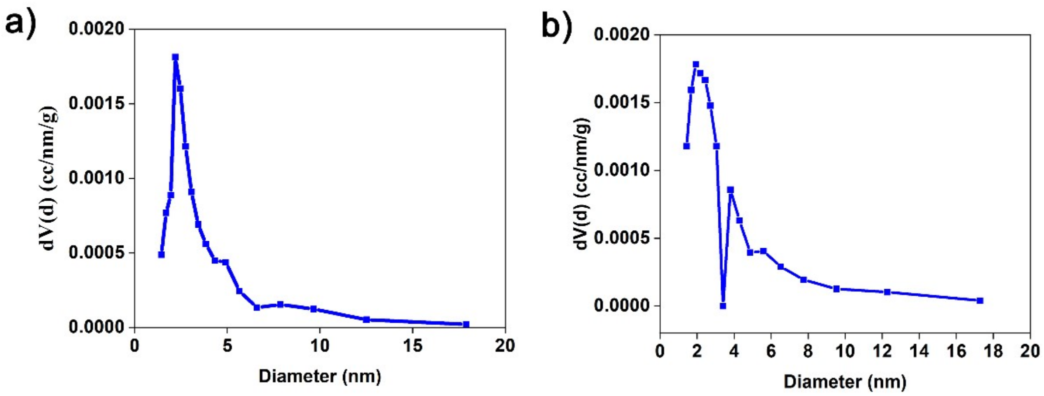

Fig. S5 Pore size distribution curves of the guanidinium hydrochloride (GuHCl) foam obtained from (a) the adsorption process and (b) the desorption process.

Fig. S6 Photographs of 2 mL of various dye solutions (100 mg/L) before and after filtration. MB, methylene blue; MG, methylene green; CV, crystal violet.

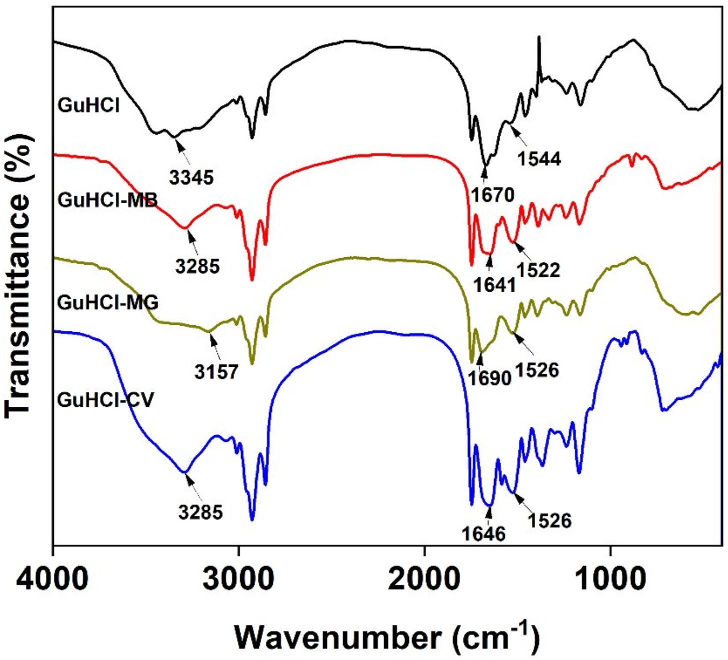

Fig. S7 Fourier transform infrared spectra of the guanidinium hydrochloride (GuHCl) foam before and after MB, MG, and CV adsorption, respectively. MB, methylene blue; MG, methylene green; CV, crystal violet.

Fig. S8 Photographs of 1 mL of Indigo Carmine solution (100 mg/L) before and after filtration.

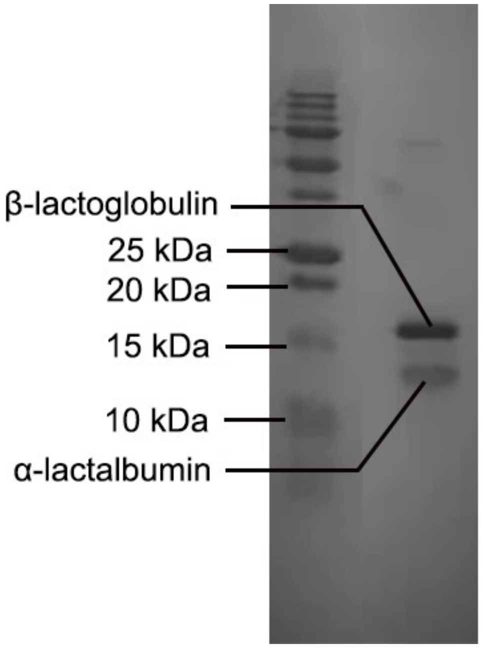

Fig. S9 Sodium dodecyl sulfate-polyacrylamide gel electrophoresis profiles of whey protein isolate. α-lactalbumin and β-lactoglobulin correspond to the bands at molecular weights of 14.4 and 18.1 kDa, respectively.

Table S1 Comparison of mechanical properties of various emulsion-filled protein gels

Materials	Testing methods	Modulus (kPa)	Fracture stress (kPa)	Fracture strain (%)	Ref.
9 wt% whey protein, 25 wt% sunflower oil	Uniaxial compression test	50	26	57	[1]
9 wt% casein micelle and sodium caseinate, 25 wt% sunflower oil	Uniaxial compression test	20	7	39	[1]
13 w/v% soybean protein isolate, 10 v/v% soybean oil	Uniaxial compression test	Not measured	14	70	[2]
6 w/v% soybean protein isolate, 20 v/v% soybean oil	Uniaxial compression test	104	8.6	32	[3]
4.5 w/v% egg white protein, 0.72 w/v% glucono delta-lactone, 10 v/v% grape seed oil	Uniaxial compression test	Not measured	18.5	72	[4]
10 wt% gelatin, 20 wt% medium-chain triglycerides oil	Uniaxial compression test	43	38	58	[5]
10 wt% whey protein, 5 wt% soybean oil droplets	Tensile test	42	55	147	This work (GuHCl gel)

Table S2 Mechanical properties of the emulsion-filled whey protein foam during the compression test

Sample[1]	Young's modulus (MPa)	Compressive strength[2] (MPa)
Control	2.23 ± 0.39	0.53 ± 0.06
GuHCl	16.86 ± 3.62	1.84 ± 0.07
DTT	0.35 ± 0.04	0.51 ± 0.04

[1] The control, guanidinium hydrochloride (GuHCl), and dithiothreitol (DTT) samples contained 200 mM NaCl, 200 mM NaCl + 2 M GuHCl, and 200 mM NaCl + 10 mM DTT, respectively.

[2] The value is the stress at 50% strain.

Table S3 Comparison of preparation and mechanical properties of various protein-based aerogels/foams

Materials	Hydrogel preparation complexity	Foam preparation method	Mechanical properties	Ref.
10% (w/v) whey protein	Simple	Freeze drying	Modulus: 2.4 MPa; yield stress: 0.3 MPa; yield strain: 15%	[6]
10% (w/v) whey protein, 2.5% (w/v) alginate, 2.5 wt% clay	Moderate	Freeze drying	Modulus: 2.5 MPa Yield stress: < 0.1 MPa; yield strain: < 5%	[6]
4 w/v% silk fibroin, 2 w/v% hyaluronic acid, 1 w/v% heparin, EDC (13.6 mg/mL)[1], NHS (5 mg/mL)[2]	Complicated	Freeze drying, annealing with ethanol vapor	Modulus: 13.1 kPa	[7]
5.3% soybean protein, 2.7% nanofibrillar cellulose	Simple	Freeze drying	Modulus: 4.4 MPa	[8]
10 w/w% protein extracts from canola seed meal	Simple	Freeze drying	Yield stress: < 0.1 MPa; yield strain: < 6%	[9]
8 w/v% whey protein, 1 w/v% cellulose particles	Moderate	Freeze drying	Modulus: ≈ 400 kPa; yield stress: 50 kPa; yield strain: < 25%	[10]
10 wt% whey protein, 5 wt% soybean oil droplets	Simple	Freeze drying	Modulus: 16.9 MPa; yield stress: 1.4 MPa; yield strain: 16.5%	This work (GuHCl foam)

[1] EDC, 1-ethyl-3-(3-dimethylaminopropyl)carbodiimide hydrochloride.

[2] NHS, N-hydroxysuccinimide.

Mercury porosimetry. Mercury intrusion porosimetry was conducted on an AutoPore V 9600 Mercury Porosimeter (Micromeritics Instruments, USA). A 0.12 g sample was put into measurement cells and was measured under the experimental conditions of 27 K, 0.1-61000 psia, and 130° contact angle of mercury on the foam.

Table S4 Pore characterization results of the GuHCl foam[1]

Sample	Surface area and pore size analyzer			Mercury porosimetry		
	Average pore size of skeleton (nm)	Specific surface area (m^2/g)	Total pore volume (cm^3/g)	Average pore diameter (μm)	Bulk density (g/cm^3)	Porosity (%)
GuHCl[2]	6.04	2.60	3.90×10^{-3}	16.05	0.31	69.53

[1] The GuHCl samples contained 200 mM NaCl + 2 M GuHCl.

[2] GuHCl, guanidinium hydrochloride.

Table S5 Comparison of precursors, crosslinkers, and organic solvents used in various biopolymer-based aerogels/foams, and their preparation complexity

Precursors	Crosslinkers	Organic solvents	Preparation complexity	Ref.
Soy flour	Formaldehyde and tannin	Ethylene glycol, methanol	Complicated	[11]
Gelatin powders/TiO_2/branched polyethyleneimine	Glutaraldehyde	None	Moderate	[12]
Cellulose nanofibril	Methylene diphenyl diisocyanate and triethylamine	*tert*-butanol, acetone	Complicated	[13]
Chitosan	Formaldehyde	Methanol, acetone, acetic acid	Moderate	[14–16]
Periodate-oxidized cellulose	None	Octylamine	Complicated	[17]
Gelatin and attapulgite	Glutaraldehyde	Ethanol	Moderate	[18]
Cellulose	3-(glycidyloxypropyl) trimethoxysilane and branched-polyethyleneimine	None	Simple	[19]
Nanocellulose	Polyethyleneimine and hexamethylenediamine	None	Complicated	[20]
Gelatin and cellulose nanofibers	Epichlorohydrin	None	Simple	[21]
Whey protein	Soybean oil droplets	None	Simple	This work (GuHCl foam)

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