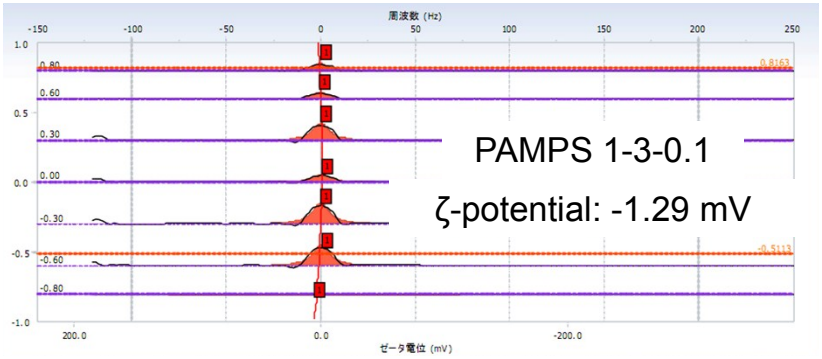
Electronic Supplementary Material (ESI) for Soft Matter.

Supporting information

(a)



(b)

	ζ-potential	Donnan potential
Potential/ mV	-1.30±0.55	-179.36±2.86

Fig. S1 (a) ζ-potential of single PAMPS (1-3-0.1) gel at 10^{-5} M NaCl solution; (b) Comparison of ζ-potential with Donnan potential measured at 10^{-5} M NaCl solution.

	Lysozyme			BSA		
ζ-potential / mV	34.07	30.12	40.23	-26.19	-23.96	-16.24
Average / mV	34.81±4.16			-22.13±4.26		

Table. S1. ζ-potential of BSA and lysozyme proteins measured at 10^{-5} M NaCl solution. The zeta-potential were measured by DelsaNano HC equipment at concentration of 1 mg/ml. The potential values of lysozyme and BSA proteins are 34.81±4.16 mV and -22.13±4.26 mV, respectively.

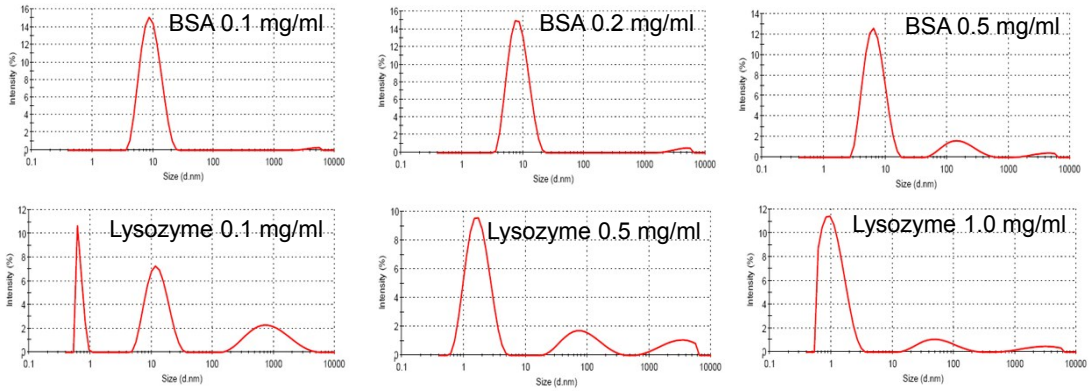


Fig. S2. Hydrodynamic diameter of protein are measured by DLS method at Malvern ZETASIZER Nano series. Firstly, BSA and lysozyme protein solution are prepared at concentration of 0.1, 0.2 and 1.0 mg/ml. Then, the hydrodynamic diameter of proteins are measured, and results were shown as above figures. The average diameter of BSA and lysozyme proteins are 8.17±0.85 nm and 1.23±0.56 nm, respectively. Because the concentration of protein solution is too concentrated, some proteins may aggregated to the multimer, the large size of protein multimer is found at DLS curves. On the other hand, an effect called multiple scattering may occur, because the light scattered by one particle is itself scattered by other particles prior to arriving at the detector, resulting a decrease of the apparent particle size. (Kaszuba, M., Connah, M. T., McNeil-Watson, F. K., & Nobbmann, U. (2007). Resolving concentrated particle size mixtures using dynamic light scattering. Particle & Particle Systems Characterization, 24(3), 159-162.).

	PNaAMPS					PNaSS
Code	1.0-4-0.1	1.5-4-0.1	2.0-4-0.1	2.5-4-0.1	3.0-4-0.1	1-8-0.1
Swelling ratio, Q	10.4±0.1	5.1±0.1	4.0±0.1	4.0±0.1	3.2±0.1	10.2±0.1
C_{ion}/ M	0.10	0.20	0.25	0.25	0.31	0.10

	PDMAEA-Q				
Code	2.0-5-0.1	2.0-6-0.1	2.0-7-0.1	2.0-8-0.1	2.0-9-0.1
Swelling ratio, Q	5.1±0.2	5.3±0.2	4.1±0.2	3.4±0.1	3.1±0.1
C_{ion}/ M	0.39	0.38	0.49	0.59	0.65

	PCDME	PDMAAm	PHEA	PNaAMPS	
Code	1.0-4-0.1	2.0-4-0.1	2.0-4-0.1	2.0-10-0.1	2.0-11-0.1
Swelling ratio, Q	3.8±0.1	1.6±0.2	1.2±0.2	2.6±0.1	1.7±0.1
C_{ion}/ M	0.26	1.25	1.67	0.77	1.18

Table S2. Swelling ratios of hydrogels and calculated the counterions concentration by using equation: $C_{ion} = C_0/Q$.

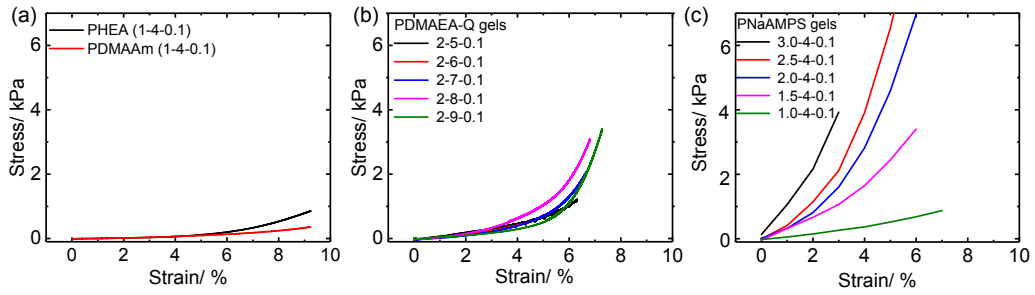


Fig. S3. Stress-strain curves of hydrogels at as-prepared states. (a) Neutral hydrogels; (b) Positive PDMAEA-Q hydrogels; (c) Negative PNaAMPS hydrogels.

	PNaAMPS					PNaSS
Code	1.0-4-0.1	1.5-4-0.1	2.0-4-0.1	2.5-4-0.1	3.0-4-0.1	1-8-0.1
Modulus/ kPa	5.3±3.0	26.1±9.9	36.7±4.3	54.1±5.0	64.8±25.8	101.6±35.3

	PDMAEA-Q				
Code	2.0-5-0.1	2.0-6-0.1	2.0-7-0.1	2.0-8-0.1	2.0-9-0.1
Modulus/ kPa	6.7±1.8	11.5±4.3	11.1±5.4	12.2±3.9	17.1±7.1

	PCDME	PDMAAm	PHEA	PNaAMPS	
Code	1.0-4-0.1	2.0-4-0.1	2.0-4-0.1	2.0-10-0.1	2.0-11-0.1
Modulus/ kPa	39.1±8.6	7.8±1.7	4.7±0.8	85.3±9.8	100.8±10.8

Table. S3. Summary the Young's modulus of as-prepared hydrogels measuring by compression test within a small strain (<5%).

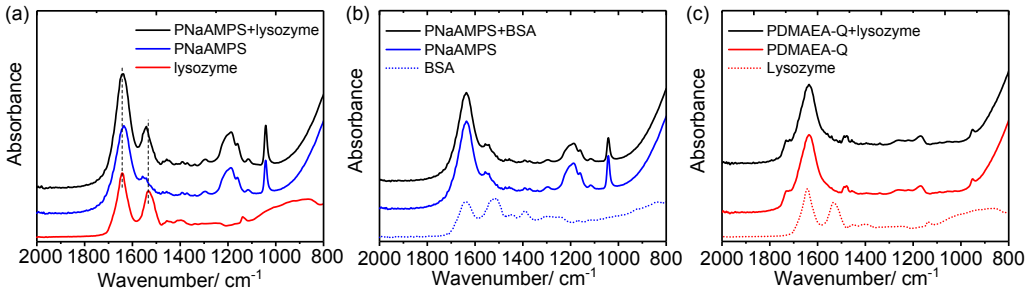


Fig. S4. (a) ATR-FTIR spectra of lysozyme, PNaAMPS gel and absorbed lysozyme protein on PNaAMPS gel. The locations of the Amide I and Amide II peaks at lysozyme proteins were recorded to be 1655 cm^{-1} and 1542 cm^{-1}, respectively from this spectrum. (b) ATR-FTIR spectra of BSA, PNaAMPS gel and absorbed BSA protein on PNaAMPS gel. (c) ATR-FTIR spectra of lysozyme, PDMAEA-Q gel and absorbed lysozyme protein on PDMAEA-Q gel.

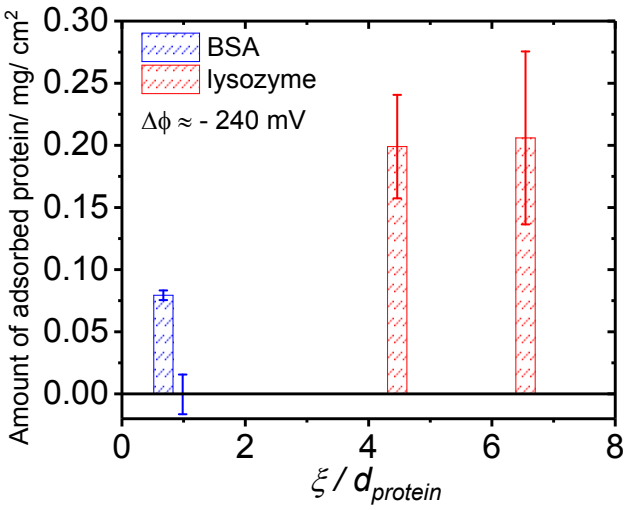


Fig. S5. The effect of the gel mesh size on the amount of protein adsorption. Even though the potential value are almost same, size ratio of mesh size to protein diameter is different. The amount of absorbed proteins are independent to mesh size of hydrogels.

Polyelectrolyte gels having same electrical potential value, i.e. the potential of PNaAMPS 2-11-0.1 gel and PNaAMPS 3-4-0.1 gel are -241.0 ± 9.6 and -238.0 ± 6.6 mV, are selected. After analyzing mesh sizes of those two gels, 5.49 nm of PNaAMPS 2-11-0.1 gel and 8.05 nm of PNaAMPS 3-4-0.1 gel are found respectively. That means the size ratio, which is defined as the ratio of mesh size to protein diameter, $\xi/d_{protein}$, is also various for above two gels. In spite of different size ratio, the protein adsorption of lysozyme and BSA proteins seems no significant difference, as shown in **Fig. S5**. Large protein adsorption was observed at oppositely charged hydrogels to protein, indicating an influence of oppositive charges in tuning protein adsorption.

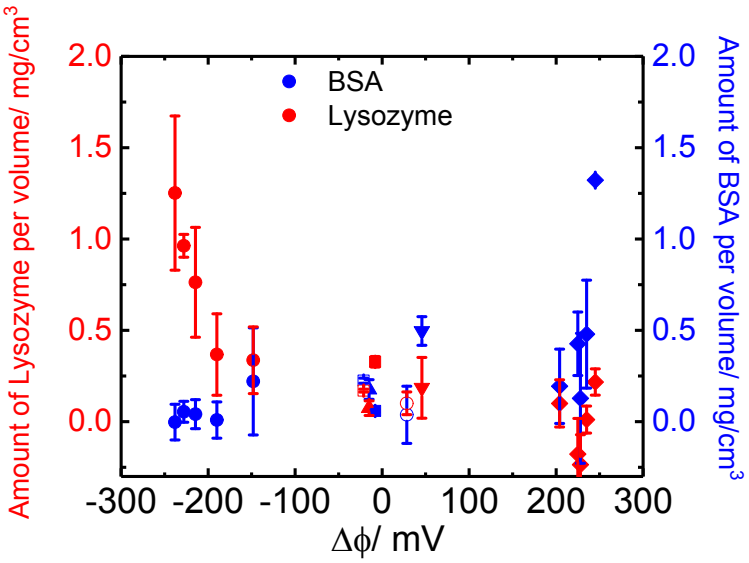


Fig. S6. Amount of protein adsorption per volume for various hydrogels, such as PDMAEA-Q (◆), PNaAMPS (●), PDMAAm (■), PHEA (▼), PCDME (▲) , PMPC (□), and PA (○) gels. The amount of adsorbed proteins on gel surface were calculated according to the equation, $m_{gel} = m_{total} - m_{solution} - m_{plate}$. The blue and red colors represent the protein adsorption behavior of BSA and lysozyme on hydrogels, respectively.

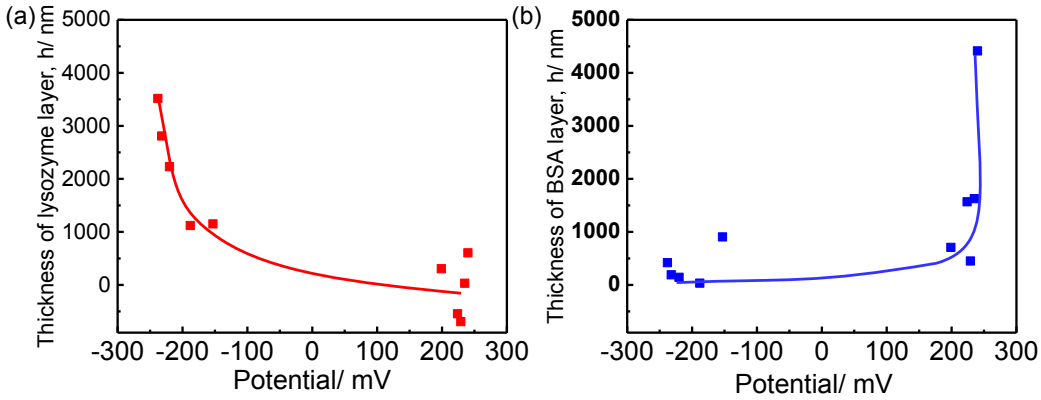


Fig. S7. Calculation of protein layer at gel surface. (a) The effect of gel potential on the thickness of lysozyme layer; (b) The effect of gel potential on the thickness of BSA layer.

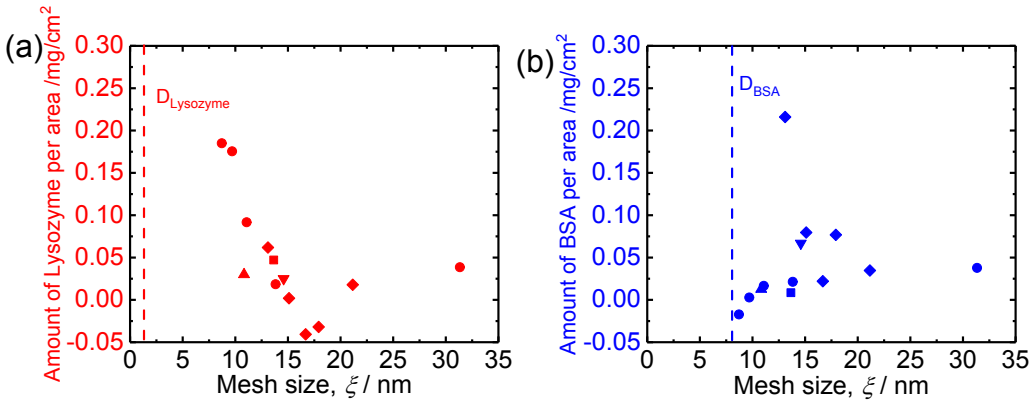


Fig. S8. The dependence of amount of absorbed proteins on mesh size of hydrogels. (a) Lysozyme proteins absorbed on hydrogels; (b) BSA proteins absorbed on hydrogels. The mesh sizes of all prepared hydrogels are much larger than hydrodynamic diameter of proteins. Symbols at figures represent PDMAEA-Q (◆), PNaAMPS (●), PDMAAm (■), PHEA (▼), PCDME (▲) , PMPC (□), and PA (○) gels.

From **Fig. S8 (a)**, even though the hydrogels have the same ξ_{real}, the lysozyme protein adsorption of PNaAMPS hydrogels is much larger than that of PDMAEA-Q gels; similarly to this phenomenon, PDMEAE-Q hydrogels with the same ξ_{real} of PNaAMPS gels show a large BSA protein adsorption.