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Author(s)	Frauenlob, Martin; Guo, Honglei; Kurokawa, Takayuki; Gong, Jian Ping
Citation	ACS Macro Letters, 12(7), 860-865 https://doi.org/10.1021/acsmacrolett.3c00160
Issue Date	2023-07-18
Doc URL	http://hdl.handle.net/2115/92799
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1 Origin of surface charge of double network hydrogels prepared by
2 sequential polymerization

3 Martin Frauenlob^{1,2}, Honglei Guo³, Takayuki Kurokawa^{3,*} and Jian Ping Gong^{3,4*}
4

5 ¹Graduate School of Life Science, Hokkaido University, N21W11, Kita-ku, Sapporo, Hokkaido 001-
6 0021, Japan

7 ²Group of Biomaterials and Microfluidics Core Facility, Institut Pasteur, 25-28 Rue du Docteur Roux,
8 75015 Paris, France

9 ³Faculty of Advanced Life Science, Hokkaido University, N21W11, Kita-ku, Sapporo 001-10 0021,
10 Japan

11 ⁴Institute for Chemical Reaction Design and Discovery (WPI-ICReDD), Hokkaido University, 8
12 N21W10, Kita-ku, Sapporo 001-0021, Japan

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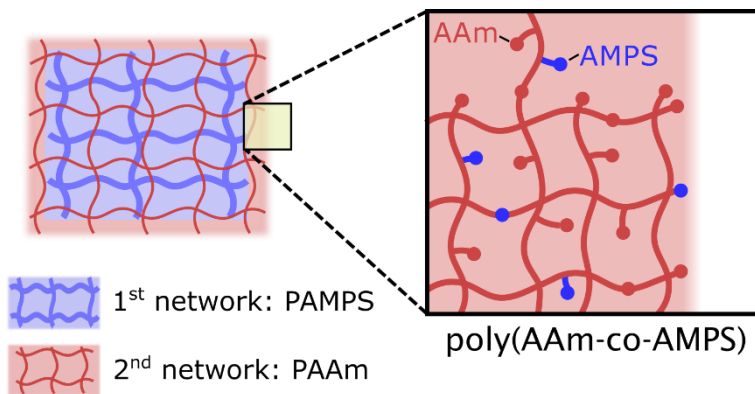
14 *Corresponding authors (E-mail: kurokawa@sci.hokudai.ac.jp; gong@sci.hokudai.ac.jp)
15

16 **Abstract**

17 Understanding the physicochemical properties of hydrogel surfaces and their molecular origins is
18 important for their applications. In this paper, we elucidate the molecular origin of surface charges in
19 double-network hydrogels synthesized by two-step sequential polymerization. Synthesis of hydrogels
20 by free-radical polymerization does not fully complete the reaction, leaving a small amount of
21 unreacted monomers. When this approach is used to synthesize double network (DN) hydrogels by
22 two-step sequential polymerization from charged monomers for the first network and neutral
23 monomers for the second network, the unreacted 1st network monomers are incorporated into the
24 2nd network. Since the surface of such DN hydrogels is covered with a μm -thick layer of the neutral
25 second network, the incorporation of a small amount of charged monomers into the 2nd network
26 increases the surface charge and, thereby, their repulsive/adhesive properties. Therefore, we propose
27 a method to remove unreacted monomers and modulate the surface charge density of DN hydrogels.

28

29 Keywords: double-network hydrogel, co-polymer, free-radical polymerization, surface charge
30 density, Donnan potential



31

32

33 Double network (DN) hydrogels are macromolecules with high water storage and load-bearing
34 capacity¹⁻³, applied in industrial^{4,5} and biomedical applications^{6,7} because of their excellent mechanical
35 performance and their similarity to biological tissue. The DN hydrogel has an interpenetrating structure
36 of two separate hydrophilic polymer networks of contrasting architecture and mechanical properties;
37 typically, a 1st network that is a highly crosslinked polyelectrolyte and at a low molar concentration
38 functions as a rigid and brittle network, and a 2nd network consisting of a loosely crosslinked electrically
39 neutral polymer that is soft and ductile⁸. The DN hydrogel is synthesized via a two-step sequential free-
40 radical polymerization where in the first step, the rigid 1st network is synthesized from an electrolyte
41 monomer such as 2-acrylamido-2-methylpropanesulfonic acid (AMPS) of high-concentration in the
42 presence of crosslinker and photo-initiator. In the second step, the as-prepared poly 2-acrylamido-2-
43 methylpropanesulfonic acid (PAMPS) hydrogel is immersed in an aqueous precursor solution
44 containing the 2nd network monomers such as acrylamide (AAm), low amount of crosslinker and a
45 photo-initiator. After the 1st network hydrogel reaches swelling equilibrium in the precursor solution
46 of the second network, the 2nd polymerization is performed in the presence of the 1st polyelectrolyte
47 network⁹.

48 Having a charged polyelectrolyte as the 1st network allows the synthesis of tough hydrogels but limits
49 the regulation of the surface charge concentration¹⁰. This is an obstacle to overcome because these
50 charges of hydrogels play a crucial role in surface applications such as lubrication and adhesion¹¹. E.g.
51 when hydrogels are used as implants, charged surfaces lead to the accumulation of proteins, which in
52 consequence leads to cell adhesion and an inflammatory response^{12,13}. Therefore, the regulation of
53 the surface properties of DN hydrogels is crucial for their applicability.

54 Previous approaches to control the DN hydrogel surface properties deploy the
55 hydrophobicity/hydrophilicity of polymerization molding substrate¹⁴, the interfacial modification, and
56 subsequent grafting of polymer brushes¹⁵ or micro-patterning^{16,17}. In our previous study, we described
57 the formation of a surface layer of the 2nd network polymer on top of the DN hydrogel bulk based on
58 the electrostatic repulsion between the glass molding substrate and 1st network polyelectrolyte

59 hydrogel¹⁴. The surface of such DN hydrogels, however, exhibits a charge density higher than expected
60 from the neutral polymer network for the DN hydrogels.

61 An explanation is that the increased charge density is related to the incorporation of unreacted
62 monomers from the 1st network polymerization step in the neutral 2nd network. In most cases of free-
63 radical polymerization, like the 1st network synthesis, the reaction does not run to completion^{18,19}.
64 Since high monomer concentration (\sim M) was used for the first network formation, the unreacted
65 residual monomer as high as sub-mM order could be present even for a reaction of 99.99% completion.
66 For single network (SN) hydrogels, these residual monomers are removed to an undetectable level by
67 washing the hydrogels in water after polymerization in multiple cycles, and the amount of residual
68 monomers is at a minimum at use. However, in the DN hydrogel synthesis, the 1st network hydrogel is
69 immediately after synthesis immersed in the 2nd network precursor solution (**Scheme 1A**). This is
70 performed to reach the high molar ratio of 20/1 – 30/1 between the 2nd and the 1st network required
71 for excellent mechanical performance⁹. Because of this two-step sequential polymerization, the
72 unreacted 1st network monomers could freely equilibrate in the 2nd network precursor solution. With
73 the subsequent synthesis of the 2nd network, the unreacted 1st network monomers could be
74 incorporated in the 2nd network so that the 2nd network is a co-polymer with a small fraction of the 1st
75 charged monomers.

76 To verify this assumption, we deployed electric conductivity measurements on monomer solution and
77 electric potential measurements on DN hydrogels and corresponding SN hydrogels. We deployed the
78 microelectrode technique (MET) to precisely quantify the charge concentration at the surface layer of
79 DN hydrogels. MET measures the Donnan potential of a hydrogel equilibrated in a bath solution
80 through the insertion of an extremely thin capillary electrode into the hydrogel^{20,21}.

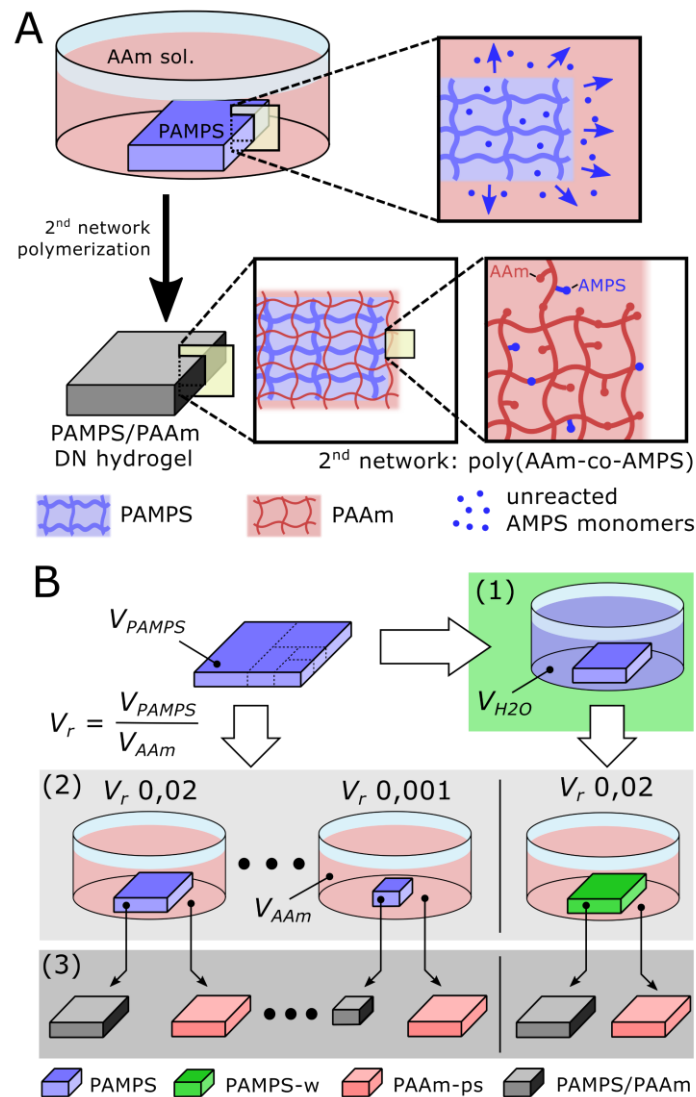
81 We synthesized DN hydrogels using PAMPS as the 1st network and polyacrylamide (PAAm) as the 2nd
82 network. Here we varied the volume ratio V_r of the as-prepared PAMPS 1st network hydrogel (V_{PAMPS})
83 and the 2nd network precursor solution (V_{AAm}) that the PAMPS hydrogel is immersed in (**Scheme 1B**).
84 By doing so, we expect to change the concentration of unreacted AMPS monomers in the 2nd network

85 precursor solution. First, we measure the conductivity of the 2nd network precursor solution with
 86 different volume ratios V_r and estimate the unreacted AMPS monomers concentration ($C_{AMPS,R}$) in the
 87 2nd network precursor solution. Second, we measure the electric potentials ($\Delta\varphi$) and estimate the
 88 charge concentration (C_G) in the surface layer of the DN hydrogels. These results (C_G) were compared
 89 to that of the SN hydrogels synthesized from the 2nd network precursor solution (PAAm-ps). A DN
 90 hydrogel where the residual 1st network AMPS monomers were removed from the PAMPS hydrogel by
 91 a washing step with water (PAMPS-w) prior to immersion in the 2nd network precursor solution was
 92 also measured for comparison. Third, the charge concentration (C_G) of DN hydrogels and SN hydrogels
 93 from the 2nd network precursor solution (PAAm-ps) was compared to that of co-polymer hydrogels
 94 (poly(AAm-co-AMPS)) synthesized from a solution containing a similar concentration of the unreacted
 95 AMPS monomers concentration ($C_{AMPS,R}$). These comparisons allow us to understand the magnitude of
 96 impact the incorporation of unreacted 1st network monomers in the 2nd network has on the DN
 97 hydrogel surface charge density. The hydrogel formulations are found in **Table 1** and **Table S1**, and
 98 details on the experimental methods are found in the **supporting information**. For the reason of
 99 simplification, the AMPS concentrations in the precursor solution of the co-polymers (poly(AAm-co-
 100 AMPS)) are also referred to as $C_{AMPS,R}$.

101 **Table 1: Hydrogel sample codes and formulations**

Sample code	PAMPS - 1 st network			PAAm - 2 nd network		
	Monomer (M)	Crosslinker* (mol%)	Initiator* (mol%)	Monomer (M)	Crosslinker (mol%)	Initiator* (mol%)
PAMPS (-w)	1	3	1	-	-	-
PAAm (-ps)	-	-	-	2	0,1	0,1
PAMPS/PAAm	1	3	1	2	0,1	0,1

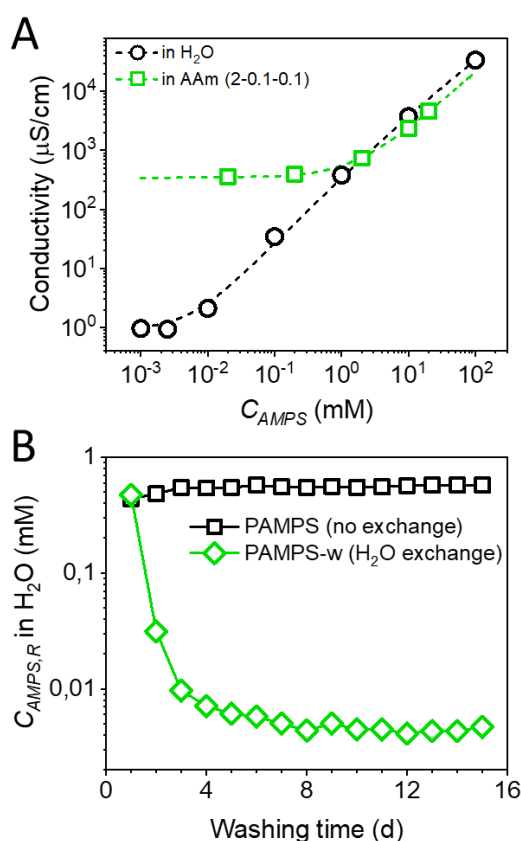
102 *The mol% of crosslinker N,N'-methylenebis(acrylamide), and initiator 2-oxoglutaric acid are relative to the concentration of monomer.



103
 104 **Scheme 1: (A) The scheme and (B) the methodology to prove the incorporation of unreacted residual**
 105 **1st network monomers (AMPS) in the 2nd polymer network (PAAm). Performed by (1) monitoring the**
 106 **removal of unreacted monomers of as-prepared PAMPS hydrogel, (2) the detection of unreacted**
 107 **AMPS monomers in the 2nd network precursor solution, and (3) the quantification of surface charge**
 108 **density of the synthesized PAMPS/PAAm DN and corresponding SN hydrogels.**
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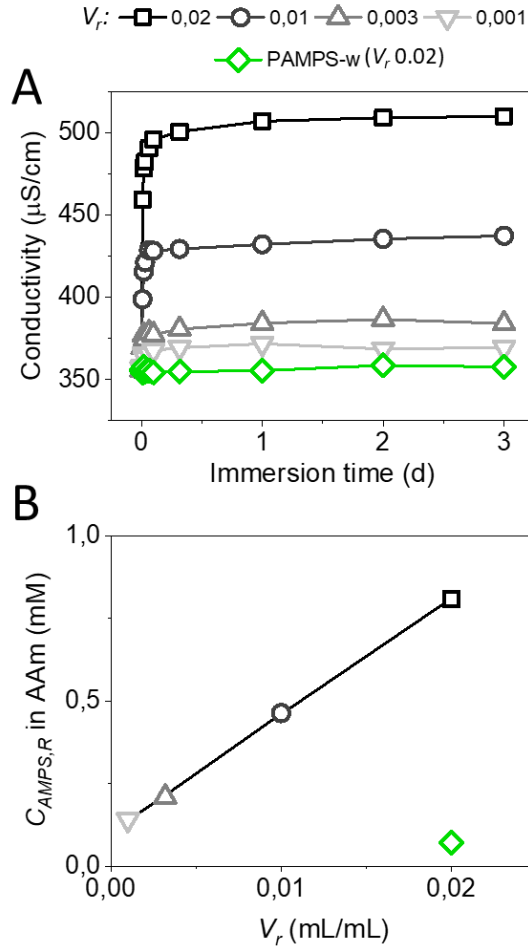
110 Monomers of polyelectrolytes such as AMPS are highly soluble in water and unreacted monomers
 111 could be extracted from the hydrogel by washing with water. The concentration of the unreacted
 112 monomer ($C_{AMPS,R}$) can be determined by measuring the ionic conductivity of the hydrogel washing
 113 water²². We can use this method to monitor the transport of AMPS from the hydrogel into the bathing
 114 solution. Initially, we established the calibration curves in **Figure 1A** for the correlation between the
 115 ionic conductivity and the AMPS concentration (C_{AMPS}) in water and in 2nd network precursor solution.
 116 These calibration curves show that the detection down to 2×10^{-3} mM of C_{AMPS} in water and down to
 117 0,1 mM of C_{AMPS} in AAm is possible.

118 PAMPS hydrogels were synthesized as described in the **supporting information**, and unreacted
 119 monomers were removed via immersing PAMPS hydrogel (V_{PAMPS}) in water (V_{H_2O}) at a volume ratio V_r
 120 of 0,02. For samples equilibrated in water without daily water exchange, conductivity constantly
 121 stayed high at $C_{AMPS,R}$ of 0,6 mM. On the sample with daily water exchange (PAMPS-w), where the ionic
 122 conductivity measurements of the washing water were performed after 24 hours of incubation, **Figure**
 123 **1B** shows that $C_{AMPS,R}$ in H_2O decreased fast and after 5 - 7 days stayed constant below 0,05 mM,
 124 corresponding to the ionic conductivity of fewer than $2 \mu S/cm$. The results indicate the necessity to
 125 wash hydrogels for at least 5 - 7 days to reduce the concentration of unreacted residual AMPS
 126 molecules from the 1st network polymerization. It needs to be mentioned that the conductivity
 127 measurements give the overall results of electric conduction from both cationic and anionic species.



128
 129 **Figure 1: Determine $C_{AMPS,R}$ in the hydrogel washing procedure for PAMPS-w.** (A) Calibration curves
 130 for the ionic conductivity of AMPS in water and in the 2nd network precursor solution with various
 131 concentration C_{AMPS} . (B) The reduction of $C_{AMPS,R}$ in the washing water of the PAMPS-w hydrogel (V_r
 132 0,02) by daily water exchange, indicating the removal of unreacted monomers.

133 To investigate if the residual AMPS amount reaches a detectable concentration in 2nd network
134 precursor solution, the washed PAMPS-w and the as-prepared PAMPS hydrogels were immersed in 2
135 M AAm containing 0,1 mol% crosslinker and 0,1 mol% photoinitiator and ionic conductivity was
136 measured for 3 days. **Figure 2A** visualizes the increase of conductivity in the AAm solution with
137 immersion time. By immersing bigger volumes of PAMPS hydrogels (V_{PAMPS}) in AAm solution (V_{AAm}) at
138 volume ratios $V_r (=V_{PAMPS}/V_{AAm})$, we achieved higher conductivities at the equilibrium time. This
139 demonstrates that residual AMPS monomers diffuse into the AAm solution and that the equilibrium
140 $C_{AMPS,R}$ depends on the volume ratio V_r between the immersed 1st network hydrogel and 2nd network
141 precursor solution. By using the calibration curve shown in **Figure 1A**, we obtain the equilibrium $C_{AMPS,R}$
142 for various V_r (**Figure 2B**). The tested volume ratios V_r of 0,01 and 0,02 are widely applied ratios in the
143 laboratory format because these ratios generate less 2nd network precursor waste solution. We see
144 that $C_{AMPS,R}$ reaches 0,46 – 0,81 mM, which could affect the charge density of the DN hydrogel. This
145 amount of $C_{AMPS,R}$ relates to a conversion rate from monomer to polymer of approximately 95 %. In
146 contrast, when the washed PAMPS-w hydrogel is immersed in AAm solution, the conductivity stays
147 stable and shows comparable results to the conductivity of pure AAm solution (**Table S2**). This shows
148 that the PAMPS-w washing procedure effectively reduces $C_{AMPS,R}$ in the 2nd network precursor solution
149 to a concentration below the conductivity measurement detection limit of 0,1 mM shown in **Figure**
150 **1A**. It should be noted that due to the small volume ratio $V_r (< 0,02)$, the concentration of AAm
151 monomers in the precursor solution of the second network hardly changes by the immersion of the
152 PAMPS network. The effect of impurities in the 2nd network precursor solution and hydrolysis of the
153 polymer on the conductivity can be neglected because its conductivity is mainly defined through the
154 initiator and residual AMPS (**Figure S1**).



155
 156 **Figure 2: Determining the concentration of the 1st network monomer in the 2nd network precursor**
 157 **solution.** (A) The time profiles of ionic conductivity after the immersion of the as-prepared PAMPS
 158 hydrogels in AAm solutions with volume ratios (V_r) ranging from 0,02 to 0,001. (B) The equilibrium
 159 residual AMPS monomer concentration in AAm solutions $C_{AMPs,R}$ as a function of the volume ratios (V_r).
 160 $C_{AMPs,R}$ was calculated from conductivity. For comparison, the results after the immersion of the pre-
 161 washed PAMPS-w at V_r 0,02 was also shown.

162 To determine if higher concentrations of $C_{AMPs,R}$ in the 2nd network precursor solution increase the DN
 163 hydrogels surface charge density C_G after synthesis, we deployed electric potential measurements by
 164 using MET. When a polyelectrolyte hydrogel is equilibrated in a salt solution, a characteristic
 165 distribution of small mobile ions inside and outside of the gels is created, which generates an
 166 electrostatic potential difference (Donnan potential) at the gel-solution interface $\Delta\varphi^{20,21}$:

167
$$\Delta\varphi = \frac{RT}{zF} \ln \left(\frac{\gamma_S \times C_S}{\gamma_G \times C_G} \right) \quad (\text{Eq. 1})$$

168 Here, R is the gas constant, T is the absolute temperature, z is the valence of the small mobile ions, F
 169 is the Faraday constant, γ and C are the activity coefficient and the concentration of the small mobile
 170 ion of interest, respectively, in bath solution (γ_S , C_S) and in the gel (γ_G , C_G). By measuring the Donnan

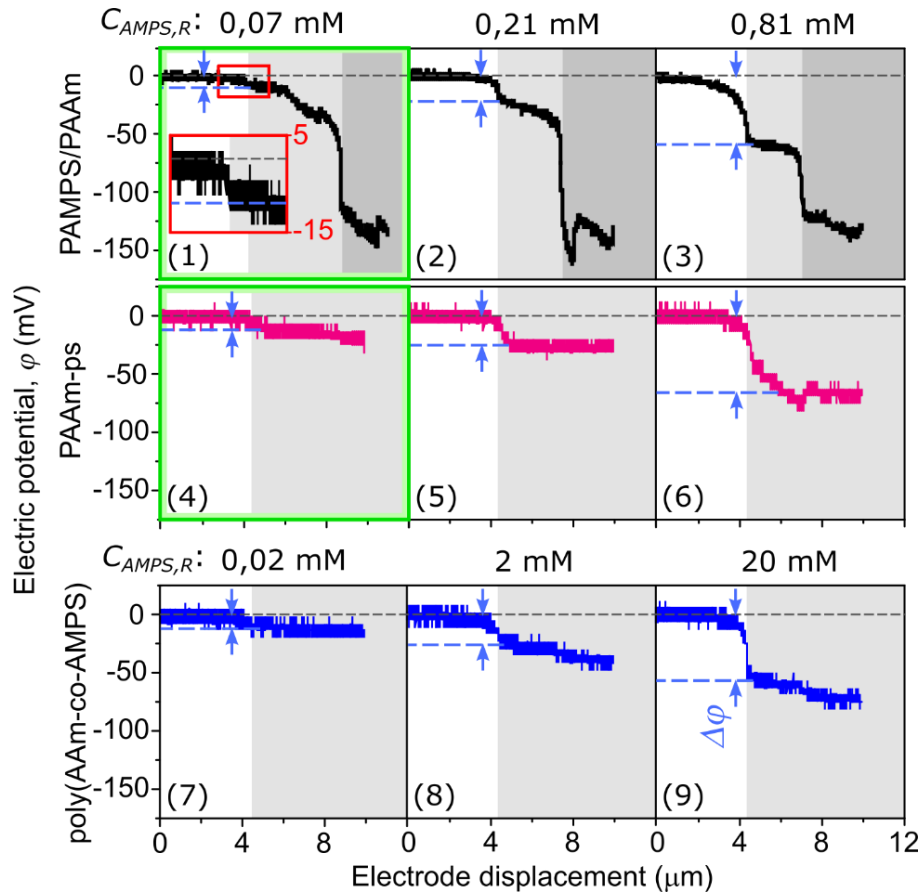
171 potential of the hydrogel in a low salt solution, we can estimate the counter ion concentration C_G in
172 the surface layer of DN hydrogels. This C_G equals the charge concentration incorporated in the surface
173 layer according to the charge neutrality condition.

174 We found (**Figure 3**, 1-3) that the electric potential difference to the bath solution at the DN hydrogel
175 surfaces $\Delta\varphi$ increased with the $C_{AMPS,R}$ (low to high) while the bulk electric potential (dark grey area)
176 was constantly low at values around -150 to -160 mV. The DN hydrogel synthesized with the washed
177 PAMPS-w hydrogel (green box) had the lowest $\Delta\varphi$ of $-16,7 \pm 3,2$ mV among the DN hydrogels, which
178 indicates that the residual AMPS monomers are incorporated in the 2nd network of PAAm. The
179 difference in electric potential between the surface layer and the bulk of DN hydrogel indicates the
180 presence of a surface layer of several μm -thick, which is consistent with the previous study¹⁴.

181 The surface electric potential could also be influenced by the limited swelling of the 2nd network
182 confined in the 1st network structure and a change of distribution of ions in the gel. To investigate the
183 influence of confinement, SN AAm hydrogels were synthesized from the residual 2nd network precursor
184 solution 3 days after the immersion of the PAMPS hydrogel (PAAm-ps). The electric surface potentials
185 of the PAAm-ps hydrogels (**Figure 3**, 4-6) are similar to the potentials found in the surface layer of the
186 corresponding PAMPS/PAAm DN hydrogels, showing that the surface layer of the DN hydrogel is
187 swelling freely and thus not affect the electric potential at the surface. It needs to be mentioned that
188 based on this measurement, we cannot account for non-uniform swelling that does not affect the
189 electric potential.

190 To strengthen the claim that $C_{AMPS,R}$ regulates $\Delta\varphi$ at the DN hydrogel surface, a control group of
191 poly(AAm-co-AMPS) hydrogels were tested (formulation in **Table S1**). The co-polymer hydrogels,
192 having similar concentrations of AMPS in feed with that of residual AMPS in the second network
193 precursor solution of DN hydrogels, show similar electric surface potentials (**Figure 3**, 7-9) to the ones
194 of the PAMPS/PAAm DN hydrogels.

195



196
197

198 **Figure 3: Electric potential profiles of hydrogels $\Delta\phi$ measured by MET.** DN hydrogels (PAMPS/PAAm, 1-3), the corresponding PAAm hydrogel synthesized from the AAm precursor solution (PAAm-ps, 4-6), and the co-polymer hydrogel (poly(AAm-co-AMPS), 7-9). The profile backgrounds in white, grey, and dark grey indicate the potential in the bathing solution, in the hydrogel surface layer, and in the hydrogel bulk, respectively. The potential differences between blue arrows indicate the electric surface potential $\Delta\phi$ of hydrogels relative to bath solution ($C_s = 10^{-4}$ M NaCl aq. solution). (1 and 4) The green frames indicate the samples prepared with pre-washed PAMPS-w as the 1st network.

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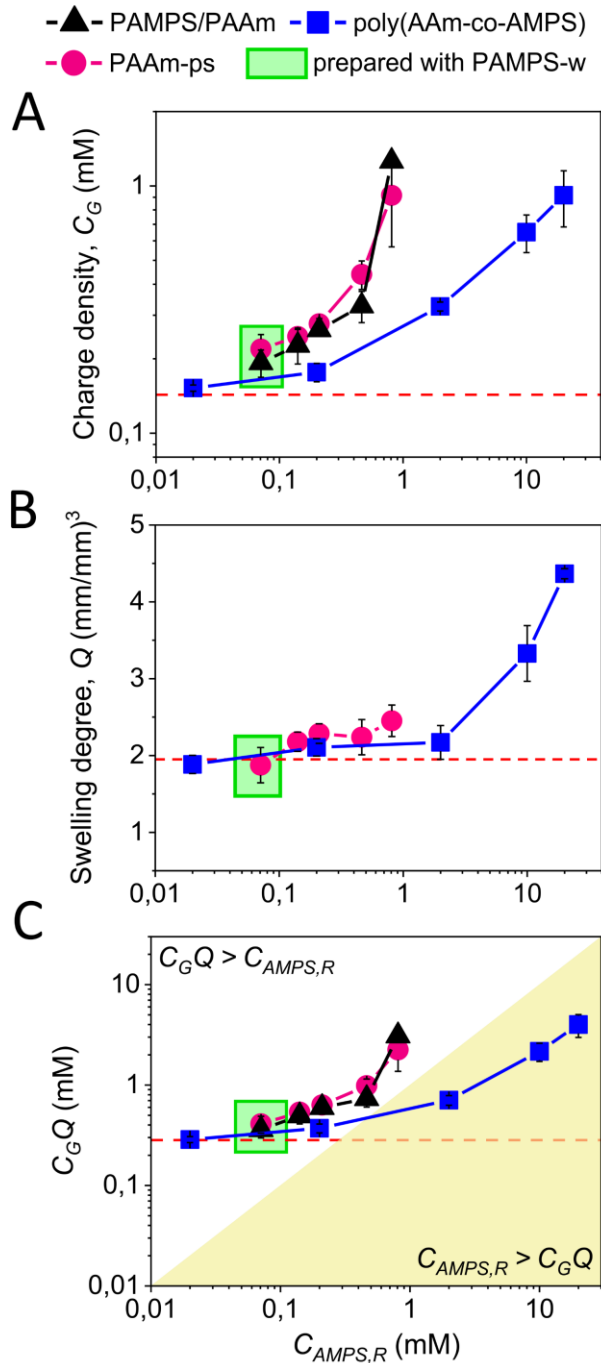
206 When we compare the charge density C_G , calculated from the electric potential $\Delta\phi$ (Eq. 1) between
 207 the sample groups, it is observable that at the same $C_{AMPS,R}$, the co-polymers show a lower charge
 208 density than that of PAMPS/PAAm DN hydrogels and PAAm-ps hydrogels (**Figure 4A**). To investigate
 209 where the discrepancy in the electric potential measurements originates from, the hydrogel swelling
 210 after 2nd network polymerization in the MET bath solution Q was correlated with the $C_{AMPS,R}$ since
 211 hydrogel swelling is linked to the fixed ions of the hydrogel. Here it was found that in the range of 0,07-
 212 0,81 mM $C_{AMPS,R}$, PAAm-ps, and Poly(AAm-co-AMPS) hydrogels have almost the same Q of about 2,
 213 independent of $C_{AMPS,R}$ (**Figure 4B**). This result indicates that a small amount of charges incorporated
 214 into the PAAm hydrogels hardly influence their swelling ability. This conclusion is also confirmed by

215 the results of macroscopic mechanical properties, which were constant in the observation range of C_G
216 **(Figure S2)**.

217 Next, we investigate the correlation between the true surface charge density incorporated in the PAAm
218 network and $C_{AMPS,R}$ in the precursor solution. Since the hydrogels swelled in the bath solution for
219 performing the MET, the values C_G measured by MET should be corrected, taking into consideration
220 the swelling. It is assumed that the surface layer of DN hydrogel should have the same swelling ratio
221 as that of PAAm-ps, although the bulk of DN hydrogel hardly swells **(Figure S3)**. So, for DN hydrogel,
222 the Q of PAAm-ps was used in the calculation of C_GQ of its surface layer. The plot of C_GQ against $C_{AMPS,R}$
223 is shown in **Figure 4C**, where C_GQ stands for the true charge density incorporated in the PAAm hydrogel
224 at the as-prepared state. With this plot, we can elucidate if all the charges in the PAAm gels are from
225 residual monomer of $C_{AMPS,R}$ or not. For the co-polymer, when $C_{AMPS,R} \leq 0,2$ mM, C_GQ is bigger than
226 $C_{AMPS,R}$, because C_GQ in this region is dominated by the weak charges of the PAAm network. When
227 $C_{AMPS,R} > 0,2$ mM, C_GQ is smaller than $C_{AMPS,R}$ which can be accounted for by a low polymerization
228 efficiency of the AMPS monomer at the tested low $C_{AMPS,R}$ and polymer ratios of $C_{AMPS,R}/C_{AAm} \leq 1/98$
229 **(Table S2)**. Surprisingly, we found that the C_GQ of PAMPS/PAAm and PAAm-ps is always higher than
230 $C_{AMPS,R}$. This deviation with the co-polymer indicates an underestimation of $C_{AMPS,R}$ in the second
231 network monomer solution.

232 This could be caused by two connected effects. First, the underestimation of $C_{AMPS,R}$. The incomplete
233 reaction of the 1st network gives not only unreacted AMPS monomers but also oligomers and sols that
234 can dissolve in the 2nd network precursor solution. These oligomers also contribute to electric
235 conductivity but less per monomeric unit in comparison with monomers. Therefore, the $C_{AMPS,R}$ from
236 conductivity using AMPS monomer solution as calibration curve could be underestimated²³. Second,
237 the oligomers and sols could also be incorporated into the second network. It is known that by free
238 radical polymerization, some vinyl groups of the 1st network crosslinker have not reacted⁸. Through
239 these unreacted crosslinkers, the oligomers or sols could also be incorporated into the PAAm-ps and
240 PAMPS/PAAm hydrogels.

241 In summary, the results in **Figure 3 and 4** clearly show that the surface charge density of the
242 PAMPS/PAAm DN gels could sensitively change with the polymerization procedures and should be
243 carefully quantified by MET. The plot of C_GQ against $C_{AMPS,R}$ could be used as a calibration curve to get
244 the required surface charge density of the hydrogels for a well-controlled experiment. Here we
245 emphasize that the amount of unreacted first network monomer copolymerized in the second network
246 is in mM concentration, and it is too low to be detected by ATR-FTIR (**Figure S4**)¹⁴. This work also
247 demonstrates that MET is a powerful method to characterize the electric properties of hydrogels.



248

249 **Figure 4: True Charge density C_G in the surface layer of hydrogels.** (A) C_G in the surface layer as a
 250 function of the residual AMPS concentration $C_{AMPS,R}$ in the precursor solution of gels. (B) The swelling
 251 ratio Q of the bulk hydrogels in the state of MET measurement against $C_{AMPS,R}$ in the precursor solution
 252 of the PAAm network. For PAMPS/PAAm, only the Q of the surface layer is relevant, so its Q of the bulk
 253 is not shown to avoid confusion. (C) True surface charge $C_G Q$ in the surface layer of hydrogels as a
 254 function of $C_{AMPS,R}$ in the precursor solution of gels. For PAMPS/PAAm, the Q of PAAm-ps was used in
 255 the calculation $C_G Q$ for its surface layer. The orange-yellow background in (C) indicates the area $C_{AMPS,R}$
 256 $> C_G Q$. The red dashed lines in (A, B, C) indicate the results of pure PAAm (Table S2). The Error ranges
 257 in the figure indicate STD for $n = 3 - 5$ measurements per hydrogel. The activity coefficients in the bath
 258 solution (γ_s) and in the gel (γ_G) are set to 1 in calculating C_G from $\Delta\varphi$ using equation 1.

259

260 We documented here, to our knowledge for the first time, that unreacted monomers from the 1st
261 network polymerization diffuse into the 2nd network precursor solution, which in consequence, leads
262 to the incorporation of a small amount of monomers of the first network into the second network by
263 the two-step sequential polymerization, resulting in the formation of poly(AAm-co-AMPS) in the 2nd
264 network. Although the concentration of AMPS monomer (< 1 mM) and the molar ratio of AMPS to
265 AAm (< 1/1000 M/M) are very low, this incorporation of unreacted 1st network monomers in the
266 second network leads to an increase of true charge density C_GQ from 0,28 of pure PAAm to 3,09 mM
267 at the surface of the DN hydrogel. Further, the increase of C_GQ was even higher than expected, which
268 could be related to the incorporation of unreacted oligomers or sols in the 2nd network. These
269 concentrations at first seem negligible. However, it was shown that such low charge densities lead to
270 the adhesion of molecules²⁴. We demonstrated that it is possible to remove the residual AMPS
271 monomer and synthesis DN hydrogels with a PAAm hydrogel layer by first, an additional washing step
272 of the PAMPS hydrogel after the 1st network polymerization as performed with the PAMPS-w hydrogel.
273 Second, the immersion of the as-prepared PAMPS hydrogel in the 2nd network precursor solution at a
274 volume ratio (V_r) of less than 0,001 has a similar effect to the washing but would waste a lot of reagent
275 solution. Further, it is possible to regulate the charge density at the surface of the DN hydrogel while
276 maintaining the swelling ratio and bulk elastic modulus constant by adding a small amount of AMPS
277 ($C_{AMPS, R} < 1$ mM or $C_{AMPS, R}/C_{AAm} < 1/1000$) in the 2nd network. Such separable modulations of surface
278 charge density and elastic modulus should greatly merit the understanding of soft interface interaction
279 between hydrogels and cells.

280 Supporting Information Available: Contains experimental details, figures on additional
281 measurements, and a summary data table.

282

283 M.F. thanks MEXT, Japan, for the provided scholarship to conduct his Ph.D. studies. This research was
284 supported by JSPS KAKENHI grants JP17H06144, JP22H04968, and JP22K21342.

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