Polymer Journal, Vol. 38, No. 12, pp. 1211–1219 (2006)

AWARD ACCOUNTS: SPSJ AWARD (2005)

Polyelectrolyte Gels-Fundamentals and Applications

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(Received October 3, 2006; Accepted October 12, 2006; Published November 10, 2006)

ABSTRACT: Polyelectrolyte gels are charged polymer networks with macro-ions fixed on polymer chains. This present paper introduces fundamental aspects, properties and application of negatively charged polyelectrolyte gels, focusing on the electrical properties of polyelectrolyte gels, diffusion of proteins in polyelectrolyte gels, interactions between polyelectrolyte gels and oppositely charged molecules, and mechanical strength of polyelectrolyte based gels. These characteristic properties of polyelectrolyte gels have considerable potential for applications, such as soft and wet scaffolds of cells, soft actuators and replacement of biological tissues. [doi:10.1295/polymj.PJ2006125]

KEY WORDS Polyelectrolyte Gel / Electrostatic Potential / Protein Diffusion / Oppositely Charged Surfactant / Double Network (DN) Gel / Cell Scaffold /

A polymer gel consists of an elastic cross-linked network and a fluid filling the interstitial spaces of the network. The network of long polymer molecules holds the liquid in place to give the gel solidity. Gels are wet and soft and look like solid material but are capable of undergoing large deformation in response to environmental change, in contrast with most industrial materials such as metal, ceramics, and plastics, which are dry and hard.

A polyelectrolyte gel is a charged polymer network with macro-ions fixed on the polymer chains and micro-counter ions are localized in the network frame. Polyelectrolyte gels exhibit the ability to absorb a significant amount (up to ~2000 times the polymer weight) of water within its network structure, but do not dissolve in water.[1] When a polyelectrolyte gel is interposed between a pair of plate electrodes and a DC current is applied, it undergoes electrically-induced chemomechanical contraction and concomitant water exudation in the air.[2] Polyelectrolyte gels exhibit various unique electrical responses different from those of linear polyelectrolyte solutions. For example, a repetitive current oscillation occurs when a DC voltage is applied to a water-swollen polyelectrolyte gel through a pair of needle electrodes.[3] Shape change and motion of polyelectrolyte gels are similar to biological motion such as muscle, flagellar, and ciliary movement in terms of the molecular level deformation, and this gel actuator has been studied for construction of biomimetic system and further application to creation of artificial organs.[1]

Living organisms such as mammalian tissues are largely made of gels, in which water content ranges up to 90%, except for bones, teeth, nails, and the outer layers of skin, and this enables the organism to transport ions and molecules more easily and effectively while keeping its solidity. Especially, biological tissues consist of polyelectrolytes such as polysaccharide and charged filamentous proteins and their properties originate from the polyelectroyte nature. Articular cartilage, containing anionic proteoglycan-rich extracellular matrix (ECM), has remarkable elasticity, low surface friction, and ability to withstand enormous physical forces.[4] These features are directly related to the high water content of cartilage, which is tightly held within the matrix of negatively charged macromolecular aggrecan/hyaluronic acid complexes stabilized by link proteins. This negatively charged polyelectrolyte gels draw great attention despite considerable theoretical difficulties in analysis.

This article describes fundamental aspects, properties, and application of negatively charged polyelectrolyte gels.

ELECTRICAL PROPERTIES OF POLYELECTROLYTE GELS

Electrostatic Potential Distribution

The properties and behavior of polyelectrolyte gels, such as high swelling, phase transition, and elasticity have been mostly investigated. However, little study has been made of local electric potential distribution in the charged network because of difficulty of the analysis. Numerical calculation of electrostatic poten-

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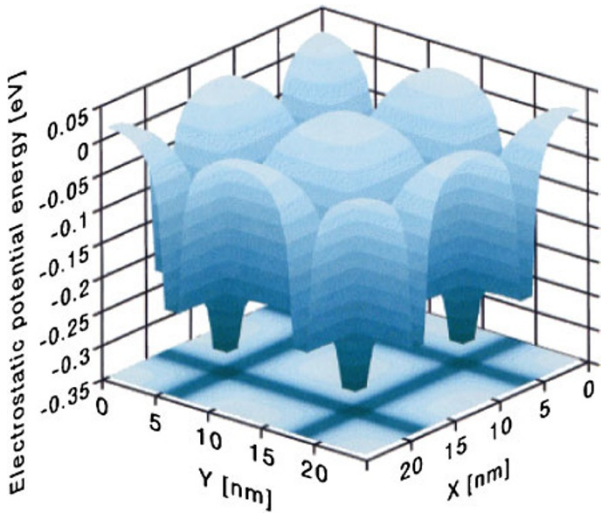


Figure 1. Spatial profile of electrostatic potential energy for the plane within the mesh-like network. X and Y axes are in unit of r_i (= 0.6 nm). Reproduced with permission from (5), Gong, J. P., *et al.*, *Chem. Lett.* 449 (1995) ©1995, The Chemical Society of Japan.

tial distribution in the charged polymer network has been made basing the Poisson-Boltzmann equation.[5]

Figure 1 shows a spatial profile of electrostatic potential energy in the unit of kT on the planes of mesh-like networks. The figure shows potential energy wells at every cross-linking points and valleys along the polymer chains.

Counter-ion distribution in the gel is determined by the Boltzmann distribution. Counter ions are mostly localized around the network knots as well as polymer chains due to the deep potential wells and valleys. Charge density of counter ions decreased very sharply with an increase in the distance from the polymer chain. Counter ions located in the deep potential valley ($\gg k_BT$) should strongly be bound to macro-ions. The number of bound counter ions would increase with cross-linking density.

The deep potential wells and high counter ion densities at cross-linking points may bring about an instability to result in counter ion condensation as predicted by Oosawa and Manning for the linear polyelectrolyte solution.[6,7]

The presence of deep electrostatic potential valleys should strongly confine the motion of water molecules which fill interfacial spaces of the network and restrict the configuration favorable to form crystal structure. This may decrease entropy and enthalpy changes of solvent molecules at crystallization, due to enhanced polarization and should decrease the melting temperature of water.

Electrical Conductance

Some effects of cross-linkage on the conductive behavior of the polyelectrolyte gel can be expected such as enhanced counter-ion "binding" which should increase with the increase in the cross-linking density.[6,7] Previous calculations revealed the presence of the deep electrostatic potential wells at cross-linking point. These potential wells should strongly localize or "condense" counter-ions through strong electrostatic interactions and should affect the conductive behaviors of the gel. Another effect is decreased contribution of ion transportation from the "giga" macromolecular network. The macroions also make a contribution to the electrical conduction of polymer solution.[8,9] This contribution is expected to be depressed in the case of the networked gel.

The equivalent (molar) conductance of the strong polyelectrolyte gel, poly(2-acrylamido-2-methylpropanesulfonic acid) (PAMPS) gel was investigated at various monomeric concentrations.[10] Figure 2c shows the concentration dependency of the equivalent conductance of the PAMPS gels with various counterions. The equivalent conductance of solutions of corresponding monomers (AMPS) and linear polymers (PAMPS) are shown in Figure 2a, 2b. This shows that a polyelectrolyte gel has equivalent conductance approximately equal to that of the corresponding linear polymer solution which showed slightly increase in the equivalent conductance with concentration. Considerable coiling of the polymer chain at such high concentrations may be responsible for the decreasing in the fraction of counter-ions condensed on the polyions, leading to higher counter-ion mobility and equivalent conductance.

However, the gels showed almost no distinct concentration dependency of equivalent conductance, which was somewhat smaller than that of linear polymer solutions at concentrations higher than 0.25 M. The polymer chain coiling effect at higher concentrations for polymer solutions may be canceled out by the increasing cross-linking points which condense counter-ions to decreases counter-ion mobility and equivalent conductance of gels.

Low Frequency Dielectric Relaxation

When the complex dielectric constant of the anionic poly(sodium 2-acrylamido-2-methylpropanesulfonate) (PNaAMPS) gels and their corresponding anionic polymer solutions were measured, the gels showed low-frequency dielectric relaxation in a frequency region lower than that of linear polymer solutions.[11] Mean relaxation time of the gel decreased with cross-linking density or the concentration, which is different from the behavior of the linear polymer solution that showed constant of relaxation time on changing polymer concentration (Figure 3). The low-frequency relaxation observed on the gels has been explained as

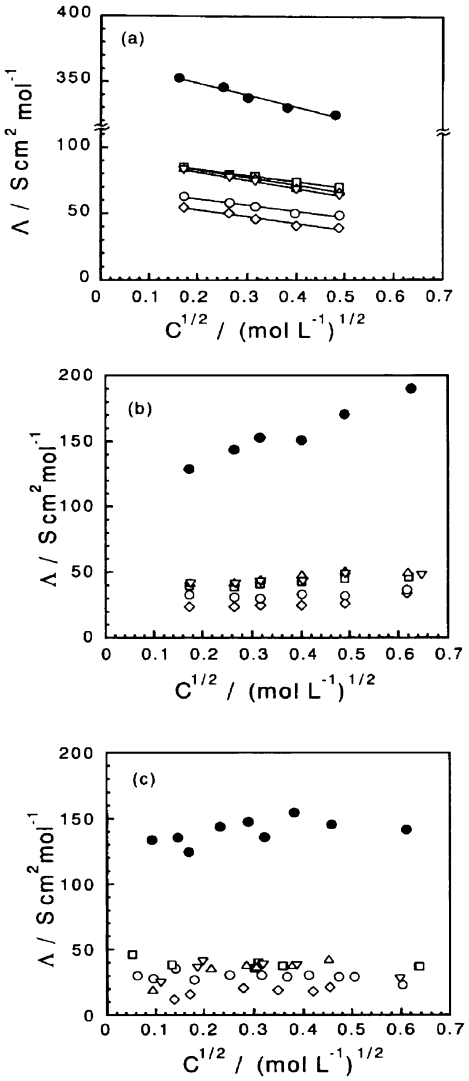


Figure 2. Equivalent conductance Λ of monomer AMPS solution (a), linear polymer solutions (b), and polymer gel (c) of PAMPS with various counterions at various monomeric concentration. T = 25 °C. (●): H; (◇): Li; (○): Na; (□): K; (△): Rb; (▽): Cs. Reproduced with permission from (10), Gong, J. P., *et al.*, *J. Phys. Chem. B*, **101**, 740 (1997) ©1997, American Chemical Society.

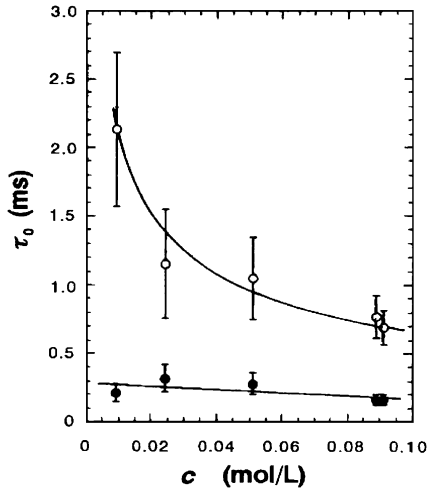


Figure 3. Concentration dependence of the mean relaxation time τ_o of PNaAMPS gels their corresponding linear polymer solutions: (○): polyelectrolyte gels; (●): linear polymer solutions. Reproduced with permission from (11), Mitsumata, T., *et al.*, *J. Phys. Chem. B*, **102**, 5246 (1998) ©1998, American Chemical Society.

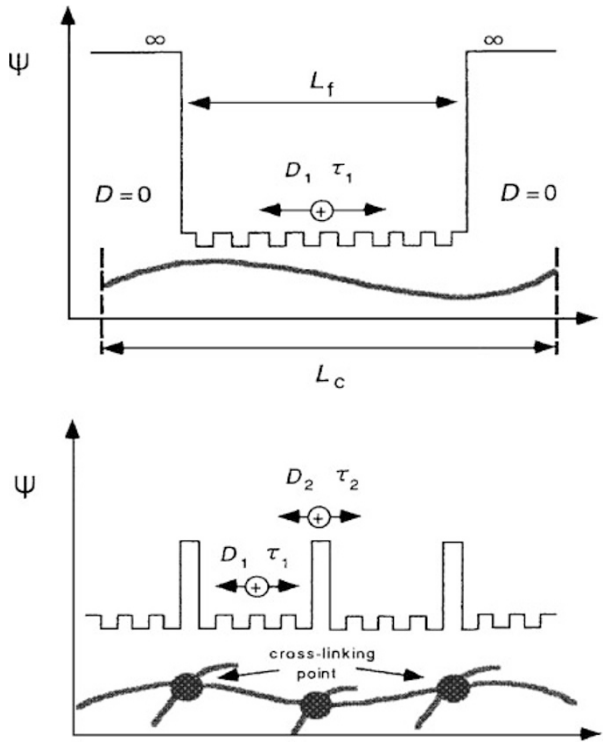


Figure 4. Schematic illustration of possible explanation for low-frequency relaxation of the polyelecrolyte gel. Reproduced with permission from (11), Mitsumata, T., *et al.*, *J. Phys. Chem. B*, **102**, 5246 (1998) ©1998, American Chemical Society.

due to the fluctuation of the bound counter ions along the polymer network by crossing through the cross-linking points (Figure 4).

PROTEIN DIFFUSION IN POLYELECTROLYTE MATRIX

Cell cytoplasm and extracellular cellular matrix (ECM) are composite aqueous solutions of multiple solutes, proteins, and electrolytes intersected by multifaceted three-dimensional structures of negatively charged polyelectrolytes such as actin filamens/microtubule and aggrecan/hyaluronic acid. Protein mobility within this complex environment should thus deviate notably from mobility in dilute solutions due to complicated intrinsic viscosity of the liquid cell medium, nonspecific binding of probe molecule to macromolecules, and the mechanical barriers as imposed by the network.[12] Study on protein diffusion in polyelectrolyte gel is thus crucial for the understanding the biochemical kinetics of cell.

The effects of charge density of an anionic gel,

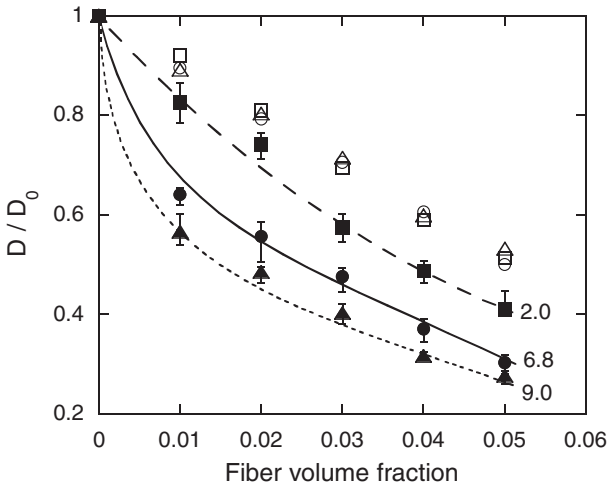


Figure 5. Dependence of diffusion coefficient of myoglobin in polysaccharide gel on fiber volume fraction of the gel with various pH and 0.01 M KCl at 20 °C. In agarose gel: (□) pH = 2; (○): pH = 6.8; (△): pH = 9. In λ-carrageenan gel: (■) pH = 2; (●): pH = 6.8; (▲): pH = 9. Numbers in the figure are pH values. Reproduced with permission from (13), Hirota, N., *et al.*, *J. Phys. Chem. B*, **104**, 9898 (2000) ©2000, American Chemical Society.

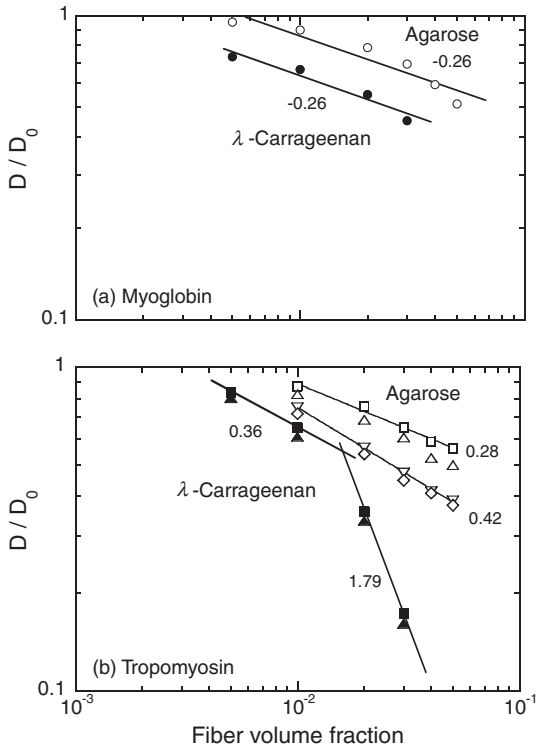


Figure 6. Logarithmic plots for fiber volume fraction of gels and diffusion coefficients of myoglobin(a) and tropomyosin(b) in polysaccharide gels. The diffusion of myoglobin in agarose gel (○) and in λ-carrageenan gel (●) was carried out at pH = 6.8 and 0.5 M KCl. Diffusion of tropomyosin in agarose gel (open marks): (□) pH = 6.8, 0.5 M KCl; (△) pH = 9, 0.5 M KCl; (□) pH = 6.8, 0.01 M KCl; (◇) pH = 9, 0.01 M KCl; Diffusion of tropomyosin in λ-carrageenan gel (closed marks): (■) pH = 6.8, 0.5 M KCl; (▲) pH = 9, 0.5 M KCl. Straight lines with slopes shown by the numbers in the figure fit the experimental data at pH 6.8. Reproduced with permission from (14), Gong, J. P., *et al.*, *J. Phys. Chem. B*, **104**, 9904 (2000) ©2000, American Chemical Society.

shape, aspect ratio of the protein, and surface charge density of proteins on protein diffusion in the polyelectrolyte gel have been investigated.[13,14] In uncharged agarose gels, diffusion of myoglobin is not effected by the change in pH and ionic strength, indicating no electrostatic interaction between the gel and myoglobin (Figure 5). In contrast, in the negatively charged λ-carrageenan gel, the diffusion of myoglobin is accelerated by electrostatic attraction at pH < pI (myoglobin) but is extensively hindered by electrostatic repulsion when pH > pI (Figure 5). The diffusion of myoglobin in λ-carrageenan gel fits the Tsai and Strieder model to give an apparent radius of the protein at various pHs.

Electronic speckle pattern interferometry (ESPI) study showed that in the λ-carrageenan gel, the diffusion of tropomyosin (aspect ratio R = 26) is different from that of globular myoglobin (aspect ratio R = 1.6) and consists a region at a low fiber volume fraction where the diffusion coefficient D of tropomyosin is similar to that of the myoglobin, decreasing with the fiber volume fraction ϕ with a scaling exponent $\alpha = -0.3$, which is close to the Rouse model and a region at a high fiber volume fraction $D \propto \phi^{-1.79}$, close to the reptation model of $\alpha = -1.75$ (Figure 6).

BINDING OF OPPOSITELY CHARGED MOLECULES TO POLYELECTROLYTE MATRIX

Thermodynamics of Binding

Oppositely charged surfactant molecules bind to a polyelectrolyte network stoichiometrically. The binding is characterized by electrostatic interaction between the surfactant ions and the oppositely charged network to form salt-like bridging, thus initiating the binding and hydrophobic interaction between bound surfactants, which stabilizes the aggregation in such a way as to settle the adjacent to the already occupied site along the polymer chain. The former is called initiation and the latter is cooperation.

Complex formation of PAMPS gels and cationic surfactant *N*-alkylpyridinium chloride (C_nPyCl) shows that hydrophobic interactions of alkyl chains are important in effective complexation.[15] That is, the positively charged surfactant molecules complex with the sulfonate moiety through electrostatic salt formation, but alkyl chains lead to additional, side-by-side, hydrophobic binding to give micellar-like structure of surfactant within the gel.

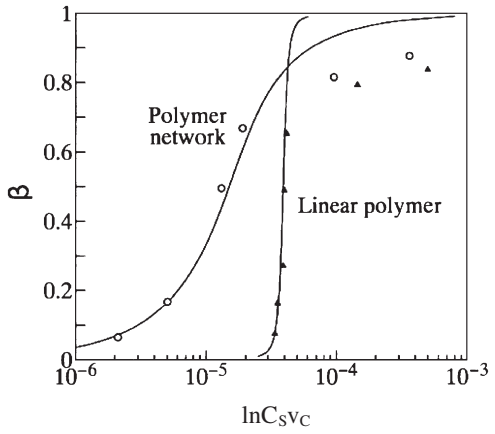


Figure 7. Binding isotherms of dodecylpyridium chloride (C_{12}PyCl) with linear and cross-linked poly(2-acrylamido-2-methylpropanesulfonic acid) (PAMPS). Solid lines are curves obtained using the following parameters: $N = 3 \times 10^{-5}$ M, $V_0 = 0.01$ L, $v_c = 0.018$ L/M, $\Delta F_h/kT = -6.2$, $q = 50$. Reproduced with permission from (23), Gong, J. P., *et al.*, *J. Phys. Chem.*, **99**, 10971 (1995) ©1995, American Chemical Society.

Complexes of polyelectrolytes with oppositely charged surfactant form well-organized structures.[16,17] Kabanov *et al.* reported the structure of the stoichiometric complexes formed between cross-linked poly(acrylic acid) (PAA) and cationic surfactants, *e.g.*, cetyltrimethylammonium bromide, to have a layered structure.[18] This structure and its functions have been investigated for many other complexes.[19–21] The complexes of polyanion gels with anionic phthalocyanine form hexagonal columnar structures.[22]

When hydrophobic interaction is treated with the nearest neighbor interaction model and electrostatic interaction is calculated using the rod-like model, the general formulas derived on the basis of the free energy minimum principle predict that the cross-linkage enhances initiation but strongly suppresses cooperation due to the osmotic pressure in the network domain. This shows good agreement with experimental data (Figure 7).[23]

Kinetics of Complex Formation

Kinetic studies of the cationic surfactant binding into an anionic polymer network have been made using surfactants with various alkyl chain lengths. In the kinetic study of C_nPyCl (n = 4, 8, 10, 12, or 16) uptake into weakly cross-linked poly (sodium 2-acrylamido-2-methylpropane sulfonate) (PNaAMPS), despite the substantial increase in network density due to the collapse of the gel, the flux of the surfactant uptake remarkably enhanced, due to increased electrostatic interaction.[24] This means that increase in the network density does not cause steric interference for the surfactant permeation, but enhance electrostatic attraction between the surfactant and oppositely charged network and accelerates surfactant uptake. This was experimentally confirmed by investigating the effects of charge density using PNaAMPS gel with various network densities. The flux increased with volume fraction of the gel, *i.e.*, the larger the swelling of the gel, the smaller the flux.

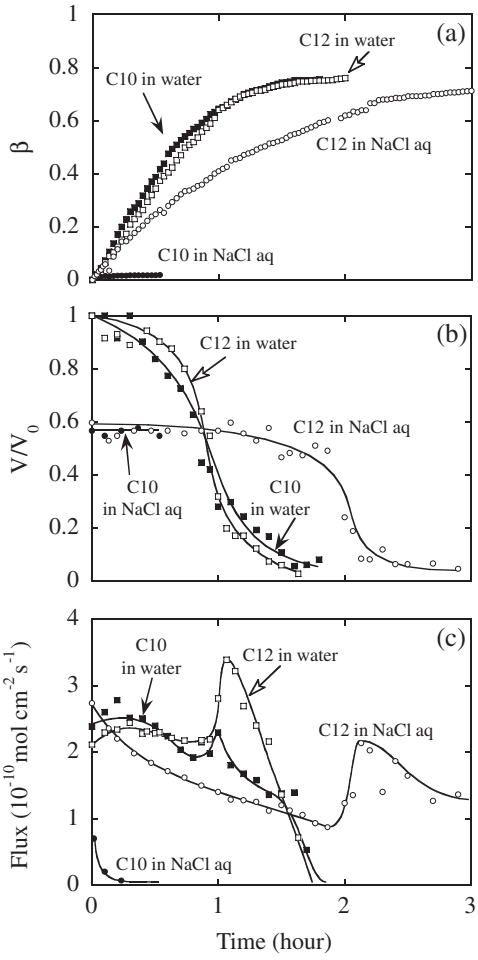


Figure 8. Time profiles of (a) binding degree of surfactants (C_{10}PyCl and C_{12}PyCl) with PNaAMPS gel, (b) relative volume change of the PNaAMPS gel by uptake of surfactants and (c) surfactant flux. (■) C_{10}PyCl in water; (□) C_{12}PyCl in water; (●) C_{10}PyCl in 0.01 M NaCl; (○) C_{12}PyCl in 0.01 M NaCl. Initial gel size V_0: $1 \times 1 \times 0.08$ cm^3; initial surfactant concentration: 4×10^{-4} mol/L; total volume of the system: 40 mL; temperature: 25 °C. Reproduced with permission from (25), Narita, T., *et al.*, *J. Phys. Chem. B*, **102**, 4566 (1998) ©1998, American Chemical Society.

The rate of surfactant uptake is strongly dependent on surfactant alkyl size and ionic strength, owing to surfactant diffusion and binding (Figure 8).[25] The driving force of surfactant diffusion into the gel is concentration gradient of the surfactant. The binding of surfactant with the polymer network sustains a high concentration gradient to facilitate subsequent surfactant diffusion.

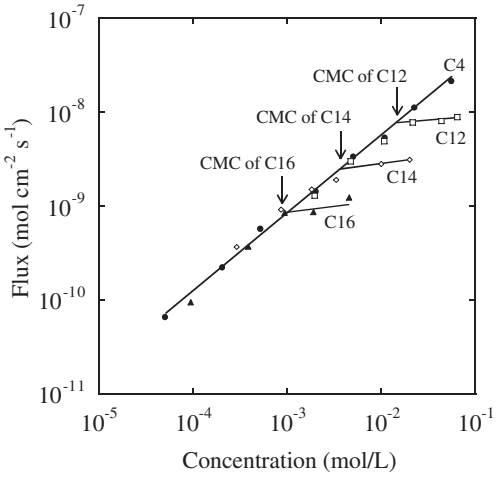


Figure 9. Dependence of the flux on the surfactant concentration. (●) C_4PyCl; (□) C_{12}PyCl (CMC: 1.5×10^{-2} mol/L); (◇) C_{14}PyCl (CMC: 3.6×10^{-3} mol/L); (▲) C_{16}PyCl (CMC: 9.0×10^{-4} mol/L). Initial gel size V_0: $10 \times 10 \times 4$ mm^3; temperature: 25 °C. Reproduced with permission from (25), Narita, T., *et al.*, *J. Phys. Chem. B*, **102**, 4566 (1998) ©1998, American Chemical Society.

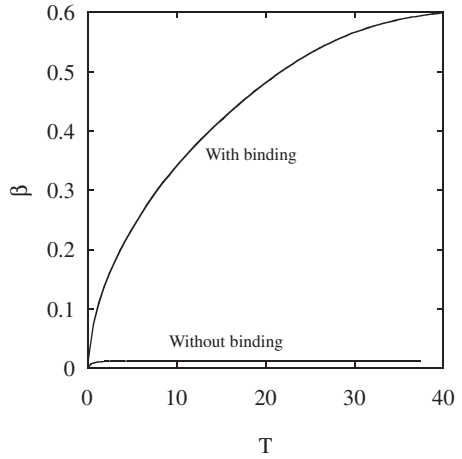


Figure 10. Time profiles of surfactant uptake with and without binding. Reproduced with permission from (25), Narita, T., *et al.*, *J. Phys. Chem. B*, **102**, 4566 (1998) ©1998, American Chemical Society.

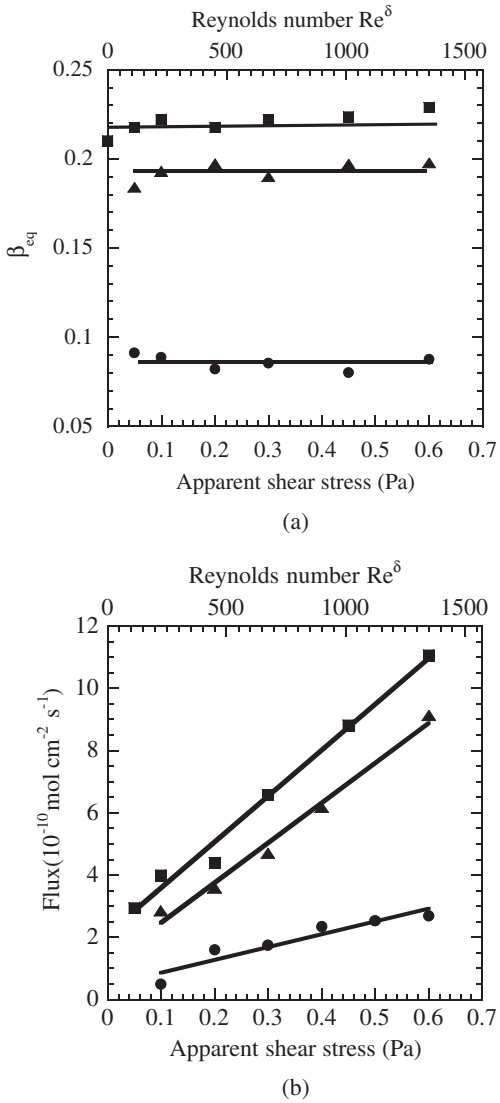


Figure 11. (a) Dependence of equilibrium degree of binding (β_{eq}) of $C_{12}Py^+$ on Reynolds number and apparent shear stress of flow at various ionic strength. (■) 0 M NaCl; (▲) 0.01 M NaCl; (●) 0.1 M NaCl. (b) Dependence of the initial flux of $C_{12}Py^+$ on the Reynolds number and apparent shear stress of the flow at various ionic strength. (■) 0 M NaCl; (▲) 0.01 M NaCl; (●) 0.1 M NaCl. Reproduced with permission from (26), Chen, Y. M., *et al.*, *J. Phys. Chem. B*, **107**, 13601 (2003) ©2003, American Chemical Society.

With the effect of micelle formation on the rate of surfactant uptake, the logarithmic plot of concentration *vs.* flux gives straight lines with slopes of 0.8 regardless of the surfactant size, while, only negligible small increase in the flux (C_{12}PyCl, C_{14}PyCl, C_{16}PyCl) occurs above each cmc and the flux of C_4PyCl, which does not form micelles increases monotonously (Figure 9).[25]

Mathematical modeling of surfactant diffusion accompanied by binding onto the polymer network well explains fact that binding enhances the velocity of surfactant uptake (Figure 10).[25]

Influence of Shear Flow on Thermodynamics and Kinetics of Binding

Studies of cationic surfactant binding to anionic polymer gel membrane at various shear flows have been performed, varying the alkyl chain length of surfactant, ionic strength of surfactant solution and the charge density of gel.[26] By exposing the gel surface to a shear flow of *c.a.* 1 Pa, the rate of surfactant uptake is distinctly enhanced and a linear relationship between surfactant initial flux and shear stress has been established, while the maximum binding ratio to the gel is not influenced (Figure 11a, 11b). This in-

dicates that the kinetics to be enhanced but the equilibrium not to be influenced by the shear flow. At high ionic strength, the effect of shear stress is suppressed, suggesting that the enhancement of surfactant uptake kinetics under shear flow is caused by decrease in the surface electrostatic potential of the negatively-charged polyelectrolyte gel, which favors uptake of the positively-charged surfactant.

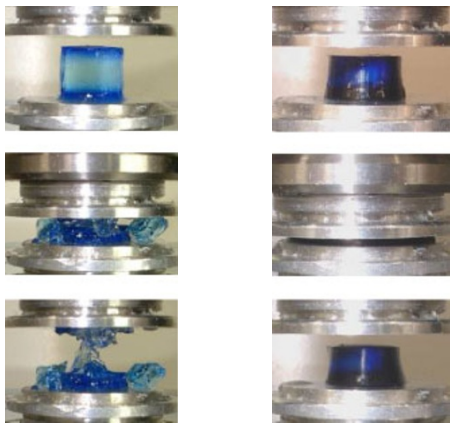
Blood flow through a vessel generates frictional drag due to movement of the fluid phase, creating a shear stress on the vascular wall surface. The endothelial cell lining on the surface of the blood vessel is exposed to a wide range of haemodynamically generated shear stress through the vascular system. This suggests that the response of the blood vessel to haemodynamicity may be also through the change of the membrane potential in the same mechanism as a polyelectrolyte gel under shear stress. This work is primarily useful in understanding the ion transmit and interaction in the blood vessel system under shear stress.

TOUGH POLYELECTROLYTE GEL WITH DOUBLE NETWORK STRUCTURE

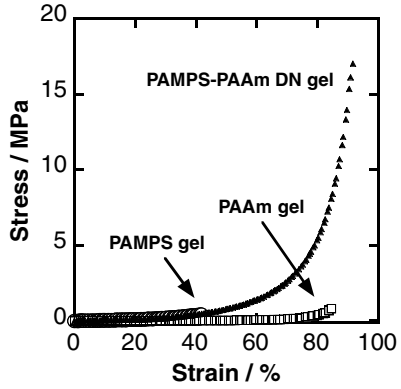
Gel scientists are providing polyelectrolyte gels as artificial connective tissues that serve predominantly biomechanical role in the body, such as articular cartilage, semi lunar cartilage, tendon, ligament and others. However, polyelectrolyte gels are very brittle and breakdown under a stress of 0.1 MPa.

However, polyelectrolyte-based gels with double network structure (DN gel) composed of two independently cross-linked networks exhibit fracture strength as high as a few to several tens of megapascals.[27] Figure 12a demonstrates how a DN gel composed of anionic PAMPS and neutral PAAm (polyacrylamide) sustains high compression while single PAMPS gel breaks down easily. Figure 12b shows a typical stress-stain curve of DN gel under compression. PAMPS gels are broken at stress of 0.4 MPa and 0.8 MPa, respectively, while PAMPS-1-4/PAAm-2-0.1 DN gel sustains even a stress of 17.2 MPa several ten times that of component SN gels. The fracture strain of the DN gel is $\lambda = 92\%$, which is much higher than that of both PAMPS gel ($\lambda = 41\%$) and PAAm gel ($\lambda = 84\%$). The stress-strain curve of the DN gel overlaps with that of PAMPS gel at small strains, suggesting that the 1st network contributes to increase in elastic stress and the 2nd one to the strain.

The high strength of the DN gels is not due to a linear combination of two component networks as with the common IPN or a fiber-reinforced hydrogel, but due to a non-linear effect of the binary structure. Although both two individual networks are mechanically weak, that is, the 1st one is stiff and brittle, and the



(a)



(b)

Figure 12. (a) Photos demonstrating how a DN gel sustains high compression. DN gels are referred as P_1-x_1-y_1/P_2-x_2-y_2, where P_i, x_i and y_i ($i = 1, 2$) are abbreviated polymer name, monomer concentration in M, and cross-linker concentration in mol % with respect to the monomer for the i-th network, respectively. left: PAMPS-1-4 gel, right: PAMPS-1-4/PAAm-2-0.1 DN gel. (b) Fracture stress: PAMPS gel, 0.4 MPa, PAMPS/PAAm DN gel, 17.2 MPa. B, Stress-strain curves for hydrogels at uniaxial compression. Circles: PAMPS-1-4 SN gel; (water content: 90 wt %), rectangular: PAAm-2-0.1 gel (water content: 90 wt %), triangles: PAMPS-1-4/PAAm-2-0.1 DN gel (water content: 90 wt %). Reproduced with permission from (27), Gong, J. P., *Adv. Mater.*, **15**, 1155 (2003) ©2003, John Wiley & Sons, Inc.

2nd network is soft and ductile, their combined DN gels are stiff but not brittle, ductile but not soft.

DN gel with non-cross-linked 3rd linear chains (DN-L gel) has high fracture strength (9.2 *MPa*) and ultra low frictional surface ($\mu \sim 10^{-5}$) at extremely high pressure of sub-MPa, as shown in Figure 13.[29] Linear polymer chains on the gel surfaces can play a role cause frictional reduction even under such an extremely high normal pressure. Soft and wet gel materials with high strength and extremely low surface friction should find wide applications not only in industry but biomedical fields, substitutes for articular cartilage or other bio-tissues.

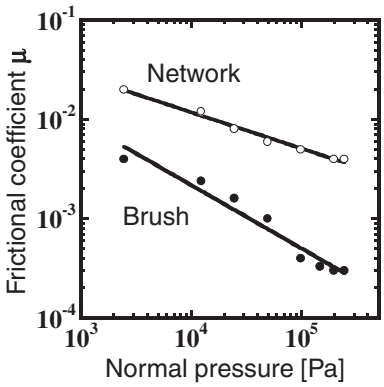


Figure 13. Normal pressure dependence of friction coefficient of hydrogels against a glass plate in pure water. Sliding velocity: 1.7×10^{-3} [m/s]. (○): Triple network (TN) gel composed of PAMPS-1-8/PAAm-2-0.1/PAMPS-1-0.1, (●): Linear PAMPS containing DN gel (DN-L) composed of PAMPS-1-8/PAAm-2-0.1/PAMPS-1-0. Solid lines are guide for eyes. Reproduced with permission from (29), Kaneko, D., *Adv. Mater.*, **17**, 535 (2005) ©2005, John Wiley & Sons, Inc.

POLYELECTROLYTE GELS AS CELL SCAFFOLDS

The soft and wet nature of hydrogels has high potential to mimic mechanical roles of tissues. Therefore, gel scientists seek to provide synthetic tissues that serve biomechanical roles in the body, such as blood vessels, articular cartilages, muscles, and others. Significant questions should be addressed such as a good adhesion to cells, proper elastic modulus, and high mechanical strength, *in vivo* and *in vitro*.

The major problem of the artificial cardiovascular is that blood clots after a certain period of implantation, due to the absence of a functional endothelial cell (EC) monolayer in the inner surface of the artificial blood vascular, to procoagulant activity. To use hydrogels as blood vessels, suitable high strength hydrogels on which endothelial cells can adhere, spread, and proliferate to form a continuous monolayer are required.

Negatively charged polyelectrolyte gels serve as good cell scaffolds, without modification of cell adhesive proteins. Bovine fetal aorta endothelial cells adhere, spread, proliferate, and reach confluence directly on poly(acrylic acid), poly(sodium *p*-styrene sulfonate), and PAMPS gels, whereas cells reach subconfluence on poly(vinyl alcohol) and poly(methacrylic acid) gels (Figure 14).[30] Negatively charged polyelectrolyte gels with a zeta potential higher than about 20 mV facilitate cell proliferation. However, nonspecific interactions of polylelectrolyte gels with cells such as the effect of charge density on cell-gel interactions are poorly understood.

OTHER NOVEL POLYELECTROLYTE GEL SYSTEMS

Anisotropic polyion-complex gels have been synthesized *via* template polymerization of cationic monomer DMAPAA-Q with an anionic mesogen PBDT template.[31] These polyion-complex gels show birefringence even at 1/40 of the lower critical concentration of the nematic phase in PBDT aqueous solution. This template polymerization route is seen as a novel method for obtaining optically aniosotropic hydrogels using very few rigid rod-like molecules.

Polyelectrolyte gels have potential for not only constructing new material such as artificial muscle and cartilage but also understanding the principles of biological system. Polyelectrolyte gels should become much more widespread as key scientific and technological materials.

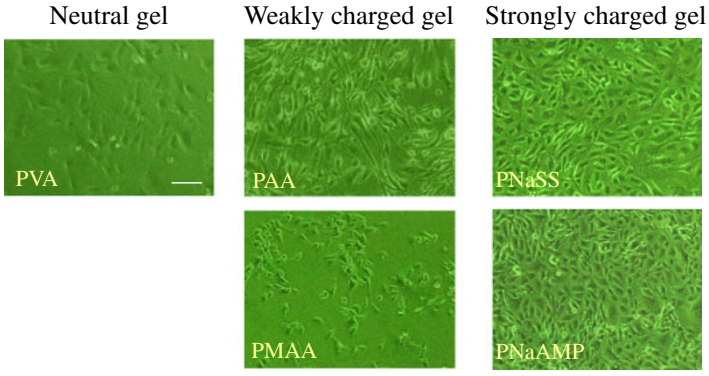


Figure 14. Phase-contrast micrographs of bovine fetal aorta endothelial cells (BFAECs) on various gels at a prolonged culture time (120 h). Crosslinker concentration of gels: PVA (6 mol %), PAA (2 mol %), PMAA (1 mol %), PNaSS (10 mol %), PNaAMPS (6 mol %). Original magnification: ×10. Scale bar: 100 μm. Reproduced with permission from (30), Chen Y. M., *Biomaterials*, **26**, 4588 (2005) ©2005, Elsevier B. V.

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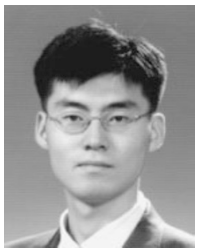

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