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Abstract

Gels, soft polymeric or composite materials that have a high fraction of water, are often found as structural materials and actuators in nature but have so far not found many uses when fabricated synthetically. We first examine some natural systems such as jellyfish, sea anemones, starfish, legumes, and human tissue, all having interesting ways of moving or otherwise reacting to the surrounding environment. Then we discuss swelling and cross-linking of hydrogels, followed by a look at actuation by electrically, thermally, and chemically stimulated gels, noting that electrical stimulation needs a chemical intermediary to show substantial actuation (comparable to human muscle, for instance). Electroactive gels have great potential as sensors and actuators but their actual uses are mainly restricted to passive drug delivery and matrices for sensors. For most applications as artificial muscles, electrically driven actuators are too weak, but chemically driven actuators look very promising. Better ways of coupling electrical energy to chemically driven gels are needed.

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Gel Sensors and Actuators

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Abstract

Gels, soft polymeric or composite materials that have a high fraction of water, are often found as structural materials and actuators in nature but have so far not found many uses when fabricated synthetically. We first examine some natural systems such as jellyfish, sea anemones, starfish, legumes, and human tissue, all having interesting ways of moving or otherwise reacting to the surrounding environment. Then we discuss swelling and cross-linking of hydrogels, followed by a look at actuation by electrically, thermally, and chemically stimulated gels, noting that electrical stimulation needs a chemical intermediary to show substantial actuation (comparable to human muscle, for instance). Electroactive gels have great potential as sensors and actuators but their actual uses are mainly restricted to passive drug delivery and matrices for sensors. For most applications as artificial muscles, electrically driven actuators are too weak, but chemically driven actuators look very promising. Better ways of coupling electrical energy to chemically driven gels are needed.

Introduction

The applications of gels are a striking example of the contrast between the worlds of synthetic materials and biological materials. In the synthetic world, gels find many applications in manufactured foods, cosmetics, and pharmaceuticals, but there are very few applications that one could really regard as engineering. Soft contact lenses are one exception.

In the biological world, most soft tissues are gels if we accept a definition of a gel as a soft polymeric or composite material with a high volume fraction of water. These tissues, such as cartilage, have substantial tensile strength and perform serious mechanical functions, whereas gels in food are largely only required to support their own weight.

In terms of microstructure, most synthetic gels are single-phase molecular dispersions of cross-linked polymers in water. In other terms, these are cross-linked polymer solutions. Many biological gels have ordered structures due to the formation of regions of intermolecular multiple helices. They may also be reinforced with collagen fibers or minerals with linking molecules forming chemical bonds between the components.

Thus, based on analogy with the biological world, we can argue that the full range of synthetic gel structures and gel properties has not really been explored

yet, and there is considerable potential for the exploitation of strong gels in soft machines. This argument can be made for passive components. More speculative is whether we can also envisage using these materials as active materials—to carry chemical, electrical, or optical signals, or to act as sensors or actuators.

Since the early work of Katchalsky,^{1,2} many demonstration devices have been built that are driven by volume changes or shape changes in gels. In most cases, these changes result from the displacement of water out of the gel or the redistribution of water within the gel. More so than other electroactive polymers, these systems have been associated with the concept of “artificial muscles.” As will be addressed in this article, a comparison with biological muscles implies target values at least for developed stress and strain and response time. Several systems have shown that these values are reachable with chemical stimulation but not with electrical stimulation. Biological muscle is itself chemically driven through the hydrolysis of phosphate bonds. The process is simply triggered by impulses from the nerves.

These observations suggest that there is no immediate prospect of high-performance, electrically driven gel muscles and that we should be seeking

methods to trigger a chemical actuation mechanism by electrical or other inputs. Alternatively, we need new ways to couple the electrical energy to the mechanical response, possibly through heat or light.

Biological Gels

Marine Animal Tissues

The jelly bodies of jellyfish, sea anemones, and other sea animals contain proteoglycan hydrogel, elastic fibers, collagen, and muscle fibers in various arrangements.^{3–5} The submicron reinforcing fibers are loosely connected to the matrix and so provide strength at high extensions and slow the viscoelastic response of the tissue.⁶

In sea cucumbers, starfish, and other echinoderms, the mechanical properties of the gel can be quickly changed from soft to hard. This was thought to occur by release of calcium ions to harden the polyanionic matrix but actually seems to be due to the release of proteins that temporarily bond to and cross-link the collagen fibrillar network.⁷ The mechanical response is essentially a change in elastic modulus, as opposed to an active contraction such as is characteristic of muscle (Figure 1).

Plant Forisomes

In legumes, specialized cells open and close to control fluid flow in the vascular system of the plant. These “forisomes,” which are 30 μm in diameter, contract in response to calcium ions in about 50 ms and develop a force of 11 kPa.⁸ The available energy density has been estimated at 0.5 MJ m^{-3} , which is close to what would be needed for an artificial muscle.⁹

Figure 1. *Mesoglea* structure. The fine structure of the mesenteries of the sea anemone *Metridium senile*. The width of the image is 1.25 μm . (Reproduced from Reference 64 with permission of the Company of Biologists.)

Preliminary tests have been made of forisomes in a microfluidic system.¹⁰

Biological Muscles

In biological muscle chains of myosin, units pull themselves along actin rods by alternately attaching, bending, detaching, straightening, and reattaching. The bending process of the myosin units is driven by adenosine triphosphate (ATP).¹¹ Muscle is characterized by a response time of less than a second, a maximum actuator stress of ~300 kPa, and a strain of 25%. The energy density is ~50 kJ m⁻³. Reaching or exceeding these characteristics is now the goal for an artificial muscle system.

Cartilage

Articular cartilage lubricates and distributes the load in human joints. Costal cartilage binds the ends of the ribs as a tough, strong joint. Other forms of cartilage in the nose, ears, and the meniscus of the knee have mechanical properties that are tuned to the particular function. The structure is a matrix of water-swollen highly anionic proteoglycan reinforced with collagen fibers arranged in oriented layers. The large proteoglycan "bottle-brush" aggregates bond to the collagen fibers.

Mechanically, this arrangement is like a balloon inflated inside a string bag. The collagen prevents the charged hydrogel from swelling to equilibrium and so provides an increased modulus in tension and compression.

From these biological examples, it can be seen that gel properties can be controlled over a wide range by the addition of stiff or elastic fibers and by control of the degree of cross-linking between fibers and between the fibers and the matrix. From the synthetic viewpoint, this control is a problem, because it requires chemical links to be formed at specific sites within the structure. On the other hand, we can also see from these biological examples that it should be possible to make gels that have significant mechanical strength and toughness and so could be used in mechanical equipment as flexible elements or as sensors and actuators. Recent developments of strong "double-network" gels suggest that gels will be able to reach the strengths necessary to be used as a cartilage replacement *in vivo* or in machines.^{12,13}

Hydrogels

The class of hydrogel materials contains swollen homopolymers and copolymers, which may be lightly or heavily cross-linked, and the degree of property under-

standing in the community varies among the materials. Environmental responses of homopolymers may be a progressive change in swelling or de-swelling as a result of a change in solvent or temperature. Such changes are described by theories of polymer solutions. In addition, polymer solutions may form two phases as a result of precipitation of a concentrated amorphous phase or a crystallization process. The phase behavior of copolymers is more complex. Random copolymers can generally be described in terms of a progressive change from the properties of one homopolymer to those of the other.¹⁴ However, block, graft, and alternating copolymers may show special behavior that is different from the homopolymers. This behavior is now well-studied for linear polymers.

Lightly cross-linked gels are not homopolymers, because the cross-linking sites have a different structure but do show the same behavior as a polymer of infinite molecular weight. In heavily cross-linked gels, the cross-link sites may act as a co-monomer to modify the polymer properties; the cross-link sites also constrain the flexibility of the chains between the cross-links.¹⁵ This reduces the entropy of solution and thus changes the phase behavior as well as limits the swelling. As with linear polymers, additional copolymerization can further modify the thermodynamics and structure of gels. Thus, while we do have a reasonable understanding of the behavior of lightly cross-linked gels, we have only lightly explored the properties of copolymer gels. There is the potential for a range of new properties and phase behaviors to be discovered in heavily cross-linked or slightly swollen copolymer gels. Polyelectrolytes such as polyacrylic acid introduce an additional complexity, because the gel chemistry changes in response to local pH and therefore act as nonrandom copolymers with a pH-dependent chemical structure.

Electrically Driven Gel Actuators

A recent review¹⁶ of electroactive gels focused particularly on polyelectrolyte gels based on cross-linked polymers of acrylic acid. Such acidic hydrogels can show a large volume change in water as the pH changes from acid to basic and the gel becomes ionized.

A pair of electrodes in a suitable salt solution will produce hydrogen at the cathode and oxygen at the anode. This results in decreasing the pH at the cathode and increasing pH at the anode. A pH-sensitive acid gel will thus shrink if it is attached to the cathode in a solution at

neutral pH. Depending on whether one or both electrodes are embedded in the gel or are in the surrounding solution, a gel may shrink, expand, or bend.^{17,18} Several groups have demonstrated actuation by these gels, while immersed in solution, with embedded or external electrodes. Osada and co-workers^{19,20} made gel "loopers" that would crawl along a bar between electrodes. Shiga and co-workers²¹ made a "fish" that would swim along a channel between external electrodes. We have demonstrated a combination of layers of two different gels that expands and contracts in a linear fashion when the cathode is embedded in one layer and the anode in the other.²²

Almost all gel actuators work in solution, but gels have been demonstrated which respond to large electrical fields in air.²³ The gels showed a crawling motion driven by electrostatic response of the gels to the applied field (Figure 2).

Figure 2. Calvert gel. Four-layer gel: A(o)/Wire/A/M(o)/Wire/M. A: polyacrylic acid gel. M: polyacrylamide gel. (o): open mesh layer. Other layers are solid. (a) After buffering at pH 5, washing, and brief acid treatment. (b) After 6 V applied for 12 min, showing expansion. A side positive. The gel can be seen to expand along the wires.

These actuators based on polyacrylate gels have been thoroughly studied in the past but have not proven forceful or fast enough to be really applicable. These issues were discussed in more detail in the recent review.¹⁶ As seen in Table I, electrically driven gels have not succeeded in providing a fast, forceful response comparable to muscle. The response depends on setting up a pH gradient in the system, which must therefore be fairly large to counteract the rapid diffusion of hydrogen ions. However, gas release at the two electrodes may swell the gel or interfere with current flow if the current is high. Thus, for a dense, strong gel, the total charge needed to neutralize the gel cannot be delivered rapidly in normal electrolyte solutions. Although it is possible to envisage combinations of special gels, electrode geometries, and electrolytes that overcome all these problems, there has been little recent progress toward fast, forceful, electrically driven gel muscles. The final column in Table I, power density, describes the bulk and weight of actuator needed to deliver a given amount of power. A power density at least comparable to muscle would be necessary for any practical, muscle-driven robotic system.

Thermally Driven Gel Actuators

The *N*-isopropylacrylamide (NiPAAM) gel system shows a sharp transition from a swollen state to contracted state on heating above 40°C. This system has been widely studied for use as actuators or in drug delivery. While the volume change is large, the response is slow as water is extruded from the gel, and the mechanical work done is small because most of these gels have a low elastic modulus and low

strength. Recent papers have explored producing faster responses by inducing porosity into the gels.⁶⁶ While the application as artificial muscles is not practical, low-stress applications such as sensors and valves are possible.²⁴ Lenses and light modulators have been demonstrated recently.^{25,26}

A familiar thermal actuator is the thermostat on an automobile cooling system. This simple and reliable device has wax in a spring-loaded metal tube. As the engine warms, the wax melts and expands, pushing open a valve and allowing cooling water to circulate. As the engine cools, the spring forces the piston back down. This system can deliver a large strain and considerable stress. The problem is that heating, cooling, and the phase transformation kinetics make the response slow and hard to control. In any thermally driven artificial muscle, similar issues must be addressed.

Chemically Driven Gel Actuators

The favored system for chemically driven gels is polyacrylonitrile (PAN) fibers that are partially converted by alkali to cross-linked polyacrylic acid fibers. Previous work has shown that these can be made into muscle-like actuators that respond to pH changes with good force and strain characteristics.^{27,28}

Recent work has found that commercial PAN yarns treated in this way could give actuator strains of about 80% and stresses of 100 kPa to 1 MPa with a response time of about 10 s.²⁹ The same fibers can be driven electrically at 5 V, using an electrode embedded in or adjacent to the fiber, to produce similar stress; however, the response time is much longer, about

10 min.^{27,29} Electrospinning has been used to make fibers of <1 μm diameter that could then be twisted into yarns. Actuator strains of 38% were obtained in cycling from pH 1 to pH 12 with a response time of about 5 s. The research team reported an actuator stress of about 10 kPa, but the data presented for actuation force and swollen fiber diameter suggests this value is actually closer to 1 MPa.³⁰ See Figure 3, which shows cycling force for a chemically driven gel actuator.

Gels Driven by Oscillating Reactions

Several groups have recently studied self-oscillating gels that swell and contract cyclically in response to an oscillating chemical reaction in the medium. Yoshida and co-workers³¹ have demonstrated patterns of contraction in NiPAAM gels that cycle through the lower critical solution temperature (LCST) as the gel cycles between two oxidation states. Yashin and Balasz³² have modeled the moving patterns of expansion and contraction set up in these gels. Jones and co-workers^{33,34} have used oscillating pH reactions in the liquid surrounding an acidic block copolymer to drive a gel motor. However, their calculations of power density for these gels do give very low values. This approach suggests that it might be possible to use an electrical signal to set off such a chemically driven oscillation, more in mimicry of muscle.

Electro- and Magnetorheological Composites

Recently, there has been much interest in composites of magnetic particles in a soft matrix that respond to a magnetic field by a change of shape and properties.³⁵ These materials are the elastomeric equivalents of magnetorheological fluids and electrorheological fluids.

The prototypical system is a dispersion of 25% of micron-sized iron particles in a gel of silicone rubber and silicone oil, but polyurethanes and other rubbers have been investigated. When cured in a magnetic field, the particles form chains.³⁶ After curing, the stiffness of the material increases in a magnetic field. At a strain of 5%, an increase in stress of ~50% is seen in a field of 123 kA/m (1500 Oe). In a study of magnetostriction on a sample preloaded in compression to 100 kPa, a field of 800 kA/m (1 T) produced a strain of 0.3%, a strain comparable to magnetostrictive alloys (Terfenol-D).³⁷ These materials also show an electric-field response if they can be formed to be nonconductive.³⁸ A shear stress of a few kilopascals can be produced at a field of 1 MV/m.

Table I: Properties of Various Gel Muscle Systems.^a

Stimulus	Deformation Material	Force Mode	Response (MPa)	Power Density	
				Time (s)	(W/cm ³)
Electric field	PVA gel film (thickness, 120 μm)	Shrinking	0.01–0.1	1000	Not available
Electric field	PVA-PAA gel (diameter, 7 mm; length, 70 mm)	Bending	0.001	1–10	<0.001
Acetone/water change	PVA-PAA gel film (thickness, 10 μm)	Shrinking	0.2	0.4–1	0.01–0.1
	Muscle	Sliding	0.2–1	<0.1	0.1–0.5

^a T. Shiga, personal communication.

PAA: Polyacrylic acid.

PVA: Polyvinylalcohol.

Figure 3. Cycling force for a chemically driven gel actuator.⁶⁵ Force-generation curve for different concentrations of acid solution acting on a polyacrylonitrile gel actuator, 100 mm in length; 2000 filaments of $\sim 30\ \mu\text{m}$ diameter. (Reprinted from Reference 65, p. 8, with permission from Elsevier.)

A study on barium ferrite particles in carrageenan hydrogels showed a modulus decrease of about 75% (from 20 MPa) on magnetization at 800 kA/m.³⁹ Strains of about 0.2% could be produced in these gels. This effect is clearly not the same as that seen in the iron-filled elastomers, since it is larger, in the opposite direction, and irreversible.

Applications of Electroactive Gels Gel Valves and Pumps

Active gels have been developed as valves for microfluidic systems, where swelling of the gel in response to light, pH, thermal, or electrical stimulation can be used to close a fluid channel or act as a pump.^{24,40–44} Electrically driven gels essentially respond to local changes in pH or ionic strength caused by electrolysis of the water.^{40,45} Bassetti and co-workers⁴⁰ showed that at high electrical fields, there is a fast response due to ion migration within the gel and then a slower response due to pH changes in the solution as hydrogen ions are released from the anode. These effects were also seen by Shiga and co-workers.¹⁸ In a flowing system, the hydrogen ions are swept away and thus only the fast response to ion migration in the gel remains (Figure 4).

Light Modulators

A series of articles have described light modulators based on the temperature-driven shrinkage of colored NiPAAM in a second gel matrix.^{26,46,47} As the colored particles expand at lower temperature, they extract water from the surrounding matrix and occupy most of the volume, thus cutting off light transmission. Clearly, an electrically driven system based on temperature or pH change could be constructed.

Gel Drug Delivery

Controlled drug release has been a subject of intense academic interest for many years. The bulk of commercial controlled-release systems depend on pH-sensitive release in the stomach or intestine. The matrix polymer may also dissolve or swell slowly to allow sustained release. Similar controlled-release systems may also be used to release flavors in food and drinks or to release fragrances during laundry or cleaning processes. Most such systems are essentially solid polymers, because hydrogels would normally release the active small molecule too rapidly for most applications. This has changed in recent years as methods have been developed to deliver much larger and more sensitive protein and peptide

drugs. These drugs also tend to be active at very low concentrations, so there is less need to produce high loadings of the drug in the matrix.

Among the niche applications now being considered for hydrogels are the protection of protein drugs in the digestive system, adhesion of drug delivery patches to the mucus membranes, and the protection of nanoparticulate drugs from the immune system.⁴⁸ The fact that the swelling and contraction of these systems can be driven electrically, as discussed earlier, means that there are potential applications for adhesive patches⁴⁹ or implanted gels with electrically triggered release. The thermoresponsive gels have been suggested for drug-eluting stents, and here too an electrical mechanism might be used to trigger the temperature change and release.⁵⁰

Gel Sensors

Many biochemical sensors depend on enzymes immobilized at an electrode surface, such that an analyte reacts to produce a compound that is readily oxidized or reduced at the electrode. Hydrogels bound to the electrode surface can be used to immobilize the enzyme without deactivating it.⁵¹ In this case, the gel is acting as a passive matrix, but there are also systems where the responsiveness of the gel drives the sensing process.

Gel-coated silicon microcantilevers can be used to detect swelling in response to changes in pH or other species.^{52–54} Changes in swelling can also be sensed by attaching the gel to a pressure sensor. Sensors can also be based on fluorescence changes in sensitive molecules in the gel,⁵⁵ on volume changes of gel particles changing the diffraction angle from a colloidal crystal,^{56,57} by color changes in a gel hologram,^{58,59} and other responses.⁶⁰

Sensors have been made which depend directly on the electroactivity of the gel. Glucose sensors have been made based on the binding of glucose by phenylboronic acid attached to a gel, where the change in ionic conductivity of the gel is measured as the binding reaction changes the swelling of the gel.^{61,62}

Although the biocompatibility of gels is not an electroactive property, it should be noted that they may be important for implantable sensors. Many sensors have been developed that work well on implantation, but almost all surfaces become coated with proteins and lose sensitivity on a time scale of several days after implantation.⁶³ Many gels are known to resist protein binding and may be able to protect implanted sensors and electrodes.

Figure 4. Typical hydrogel response to applied dc voltage⁴⁰: (a) 23 V is applied to the electrodes, and the hydrogel jacket swells (upper image). After 1 s, the voltage is turned off, and after a time the hydrogel returns to its equilibrium jacket thickness (lower image). (b) Plot of wall thickness in response to an applied field for 1 s. (Reproduced with permission from Reference 40. © 2005 IEEE.)

Conclusion

Electroactive gels have great potential as sensors and actuators, but the actual uses of gels are mainly restricted to passive drug delivery and matrices for sensors. For most applications as artificial muscles, electrically driven actuators are too weak, but chemically driven actuators look very promising. On the other hand, delivery of chemical energy requires pumping systems that greatly add to the weight and complexity. What is needed

are better ways of coupling electrical energy to chemically driven gels. If we look for inspiration to biological muscles, we can expect to develop complex systems with several interacting components structured on a very fine scale.

With sensors, the problem is similar but reversed. There are many chemical species such as pesticides, drugs, proteins, and peptides for which electrochemical sensors are not available. Responsive gels have potential as sensors of such specific

molecules through binding interactions that result in volume changes. The problem is to find a simple electrical method to convert these changes into an electrical signal.

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