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Review of Artificial Muscle based on Contractile Polymers

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Abstract: An artificial muscle with strength and speed equal to that of a human muscle may soon be possible. Polymer gels exhibit abrupt volume changes in response to variations in their external conditions — shrinking or swelling up to 1000 times their original volume. Through the conversion of chemical or electrical energy into mechanical work, a number of devices have already been constructed which produce forces up to 100N/cm^2 and contraction rates on the order of a second. Though the promise of an artificial muscle is real, many fundamental physical and engineering questions remain before the extent or limit of these devices is known.

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1 Introduction

The electric motor is the primary actuator used in robotics. However, its high weight, limited sizes, complex transmissions and restrictive shapes have constrained the design and development of robotic systems. Alternatives such as pneumatics and hydraulics have achieve some successes, but these depend on compressors which are themselves driven by electric motors. Nitinol, a shaped memory alloy, has also been used in robotics, however the thermal to mechanical energy conversion has proven difficult in practical applications. What would be ideal for robotic actuation would be an analog of a human muscle — a contractile compliant material driven by chemical or electrical signals. Recent research in polymer gels offers the hope for an artificial muscle and a potential revolution in robot actuator design.

Gel is intermediate between liquid and solid, consisting of a polymer network and interstitial fluid. The properties of the gel, including its equilibrium and dynamic aspects, are defined by the interaction between the polymer and the liquid. Examples of gels abound including natural gels, such as Jello, the vitreous humor of the eye, the lining of the stomach, intestine and lung, and muscle; and artificial gels, such as polyacrylamide, polystyrene, and others used in the manufacture of rubber, plastics, glues and films. A property common to all gels (Li 1989) and one important for actuator design is their unique ability to undergo abrupt changes in volume.

Gels can swell or shirk as much as 1000 times in response to small changes in external conditions such as temperature, pH, electric fields (De Rossi 1986, Tanaka 1982), or solvent and ionic composition (Tanaka 1987). Not only are these changes large, they are also reversible. Restoration of the initial external conditions returns the gel to its original volume. In general, the rate of contraction is proportional to the square of the linear dimension of the gel. For example, micro sized gel fibers contract in milliseconds. In addition, some gels support substantial loads. Polyacrylonitrile-polypyrrole (PAN-PPY) and polyvinylachohol (PVA) gel fibers generate up to 100N/cm^2 (Chiarelli 1989), approximately equal to that of a human muscle.

The possibility of an artificial muscle is quite exciting. In fact a number of researchers have already constructed robotic and prosthetic prototypes based on contractile gels. However, to move beyond a mere laboratory curiosity, a number of fundamental issues must be addressed, such as efficiency, power density, energy storage and transmission, dynamics, control, heat dissipation, actuator design, and others. In addition, contractile gel actuators must compared favorable to existing actuators, at least in some applications, to be considered a viable alternative in mechanical design.

This review outlines some of the basic research in polymer gels, providing a background for the analysis and design of gel actuators. First, the physical mechanisms of gel volume change are discussed. Second, the processes and methods of gel preparation are presented. Third, the gel kinetics or rate of contraction, which are of primary important in actuator applications, are reviewed. Finally, some examples are given of actual devices constructed from these contractile polymers.

2 Volume change

There are three competing forces acting on the gel polymer network: the rubber elasticity, the polymer-polymer affinity and the hydrogen ion pressure. These forces, collectively called the *osmotic pressure*, determine the equilibrium state of the gel. The competition between these forces determines the osmotic pressure while the changing balance of these forces produces the volume change.

Rubber elasticity tends to shrink the gel under tension and expand it under compression. The elastic force is in equilibrium when the polymer ends are at their root mean square distance. Although the equilibrium volume for elasticity is independent of the external conditions, its force is proportional to the absolute temperature.

Polymer-polymer affinity depends on the electrical attraction between the polymer and the solvent. An attractive force between the polymer and the solvent causes the absorb solvent molecules, while repulsive force produces the opposite effect. This force does not depend on the temperature, but on the solvent and volume of the gel. Since polymer-polymer affinity is a short range force and depends only on polymer-polymer contact, its effect is inversely proportional to the square of the volume.

Hydrogen ion pressure is the force exerted by the motion of the hydrogen ions H^+ within the gel network. Hydrogen ions enter the gel attracted by the negative charges on the polymer chain while their random motions tend to expand the gel much as a gas exerts pressure within a contained volume. The hydrogen ion pressure depends on the ionization of the polymer, as well as, both temperature and volume. The force is linearly proportional to the absolute temperature and inversely proportional to the square of the volume.

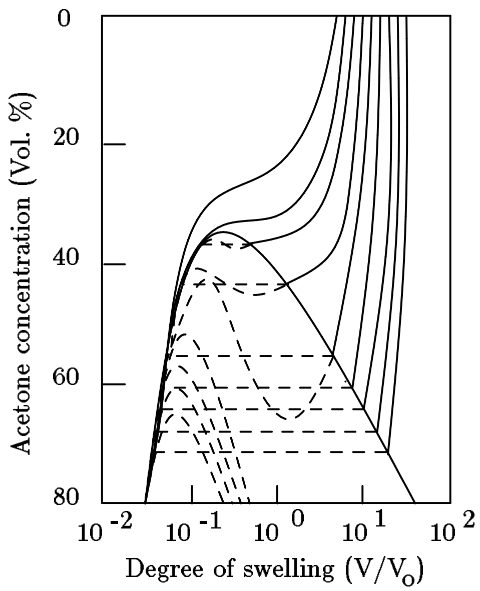

Figure 1 Volume changes in acrylamide gels are shown as a function of acetone concentration and degree of hydrolysis.

3 Gel Preparation

Gels can be formed by condensation polymerization, where bifunctional and polyfunctional units are combined, or by cross-linking polymers from bifunctional monomers. Cross linkages can form from covalent bonds or through weak forces such as hydrogen bonds, Van Der Waals forces or hydrophobic or ionic interactions. As an example, consider the preparation of standard polyacrylamide gels (Tanaka 1981). Acrylamide and bisacrylamide are dissolved in water together with polymerization initiators, ammonium persulfate (AP) and tetramethylethylene diamine (TEMED). TEMED reacts with AP leaving an unpaired valence electron which combines with acrylamide or bisacrylamide transferring the unpaired electron to the acrylamide molecule, figure 2. This procedure continues, forming an indefinitely large polymer network. After washing, the gel is hydrolyized in a basic solution, converting the aminocarbonyl side chains into carboxyl groups. The degree of hydrolyization determines the percentage of carboxyl groups which greatly affects the volume phase transitions of the gel (Hirokawa 1985, Nicoli 1983, Tanaka 1987a)

There are many other gels, all of which are characterized by their unique side chains. The macroscopic structure of these gels and their preparation are basically the same as the polyacrylamide. The preparation procedure for some of these gels are outlined in appendix 1.

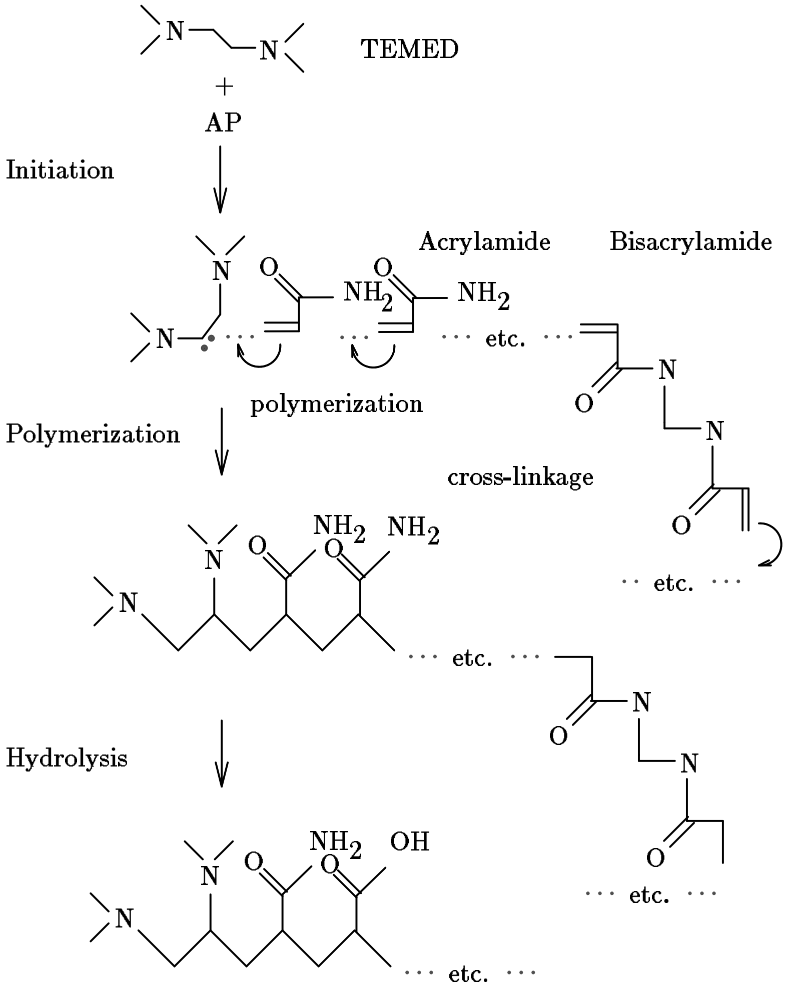

Figure 2 Activation of TEMED by AP yields an unpaired valence electron which combines with acrylamide or bisacrylamide forming an indefinitely large polymer network. Hydrolysis in a basic solution converts aminocarbonyl groups into carboxyl groups, which affects the volume phase transitions. (Tanaka 1981)

4 Kinetics

The kinetics or rate of swelling (or contraction) is an extremely important characteristic in the application of gels to mechanical actuation. The gel kinetics is a diffusion limited process and is therefore proportional to the square of the dominant linear dimension of the gel. Thus for a gel fiber, the contraction rate t_c is equal to a contraction rate constant c times the square of the diameter d^2, $t_c = cd^2$. For polyacrylamide, c is approximately 2×10^9 s/m^2. Therefore polyacrylamide gels with a diameter of 1 cm take about 2.5 days to contract, while micron diameter fibers take milliseconds. An artificial muscle formed by bundling 25μm diameter PAN-PPY fibers was constructed and demonstrated contraction rates on the order of a second (Chiarelli 1989).

In spite of its important for actuator design, gel kinetics has only recently been investigated (Matsuo 1988a, 1988b, Tanaka 1987a). Initially it was thought that volume change was governed by the diffusion of individual water molecules through the network, but it was latter shown to be a collective diffusion process of the entire polymer network. The local motion of the polymer network is given by a diffusion equation $D = K/f$, where D is the diffusion coefficient, K the elastic modulus of the network and f the frictional coefficient between the network and the liquid (Chiarelli 1988). The collective diffusion explains both the macroscopic swelling rates and the dynamics of local density fluctuations.

5 Application

Though the phenomena of volume change within polymer network and the chemical driven contractile forces they produce has been known for years (Kuhn 1950, Flory 1956, Hamlem 1965), only recently have there been any attempts to apply these materials to robotics. A parallel jaw gripper driven by antagonistic polyvinylachohol contractile elements powered by changes in acetone concentration has been constructed (Caldwell 1989), along with an artificial urethral sphincter, (Chiarelli 1989). For the most part, however, these devices are still simple prototypes stages.

The first gels were very slow, reaching an equilibrium volume anywhere from one half an hour to a number of days. Most gels were also weak, unable to support significant loads. However, recent research has focused on thin films and bundles of small fibers whose polymer networks are aligned with the strain axis, allowing faster contraction rates and higher strength. Other attempts include superimposing a cross-linked a network onto a fiber by projecting UV light through a mask. This should yield a strong polymer and an effectively smaller dominant linear dimension, producing a gel with both high strength and rapid contraction (Zhang 1990).

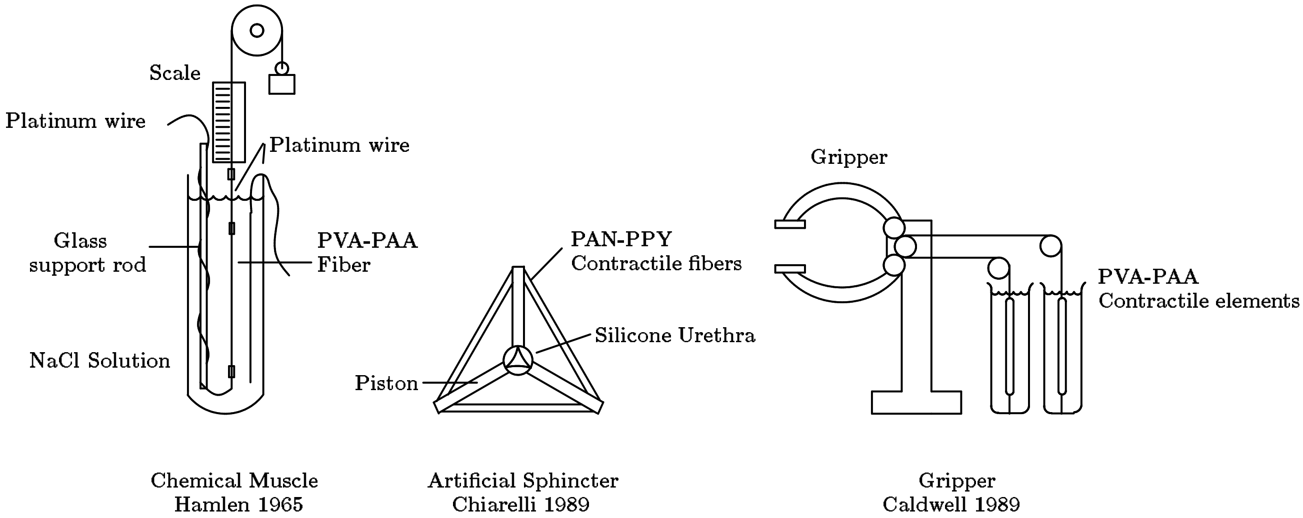

Figure 3 A force generating element, a robot gripper and artificial urethral sphincter are some of the mechanical devices constructed from contractile polymer gel.

6 Conclusion

Despite some success in contractile gel actuator development, there are a number of fundamental questions which must be answered before consideration gels in robotic devices. First, there are material questions such as the elastic modulus, tensile strength, stress-strain relations (Chiarelli 1990), fatigue life, and thermal and electrical conductivity. Second, there are thermodynamic issues such as efficiency, power and force densities, and power limits. Third, there are basic engineering concerns such as power supply and delivery, device construction (De Rossi 1988), manufacturing, power transmission, dynamic modelling (Genuini 1990, Morasso 1990), control, integration and packaging. Finally, there are other concerns such as the coordination and integration of multiple actuators, material and manufacturing costs and the toxicity of both the gels and their precursors. Many promising technical advances (e.g. ceramic superconductors) have been limited by basic physical or engineering deficiencies. However, given limitations, there may be technical niches were these devices may provide practical results. In any case without a thorough investigation of the application of contractile gels to actuator design, the extent or limit of this technology is still unknown.

Appendix
Gel Preparation

N-Isopropylacrylamide (N-IPA) (Matsuo 1988)

- **Preparation of paraffin oil**
  Wash paraffin oil with alkaline (1N NaOH) and acid (1N HCl) to remove contaminants. Flush with large amounts of water. Pass paraffin oil through a column packed with molecular sieves to remove water.
- **Purify N-IPA**
  Recrystallize N-IPA with toluene, filter by aspiration and dry under a vacuum. Store in a colored bottle to avoid light exposure.
- **Monomer solution**
  Combine 700mM N-isopropylacrylamide (N-IPA), 8.6mM N,N'-methylenebisacrylamide (BIS) and 0-128mM sodium acrylate (prepared from acrylic acid and sodium bicarbonate).
- **Polymerization**
  Add 48 μl tetramethylethylenediamine (TEMED) and 0.2 ml of ammonium persulfate to 10 ml monomer solution
- **Gelation**
  Inject 1 ml per-gel solution into 50 ml paraffin oil, degas, saturate with nitrogen, and disperse into submillimeter droplets. Agitate for one hour at 20°C, then pour solution in 1000 ml water. Use petroleum ether to removal paraffin oil. Wash gel beads several times.

Polyacrylonitrile-polypyrrole (PAN-PPY) (Chiarelli 1989)

- **Anneal PAN**
  Anneal PAN (Mitsubishi Rayon Co.) at 220°C for 5 hours and then boil in 1N NaOH for 30 min. to produce a partially ionized carboxylic structure
- **Combine with PPY**
  Immerse fibers for 24 hrs. in an aqueous solution of ferric chloride (40% by weight). Add HCL to pH = 0.5. Polymerize PPY in PAN by gas state technique (Ojio and Miyata). Place in flask under saturated atmosphere of water and PPY at 10°C for 5 to 20 hours.

Polyvinylalcohol-polyacrylic acid (PVA-PAA) (Chiarelli 1988)

- **Mix components**
  Dissolve 80% PVA with a degree of hydrolisis of 98% and molecular weight ~10k (AnalytiCals, Carlo Erba, Milano, Italy) with 20% PAA molecular weight ~ 250k (Aldrich Chemical Co., Milwaukee, WI, USA) in bidistilled water and stir for 20 min. at 60°C.
- **Dehydrate**
  Dehydrate solution in at 40°C under mild vacuum
- **Cross-link**
  Thermally cross-link polymer at 130°C for 45 min., then equilibrate with bidistilled water.

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