Chapter 12
Dielectric Gels

Toshihiro Hirai

Abstract Dielectric gels of various types are recently found electrically active, and can be used for actuators. Polymer gels swollen with large amount of dielectric solvent deformes by applying dc voltage. The deformation is based on the solvent flow (or ion drag) through the polymer network. They shows contractile, bending, and crawling deformation. Advantages are very swift deformation in air, small electric current, and large strain up to over 10 % depending on the degree of crosslinks among the polymer chain. Disadvantages are low durability because of the solvent bleed-out, and relatively high voltage.

Dielectric elastomers (sometime gel-like) can be good candidate when the polymer chains are flexible enough and sensitive enough to the electric field, although flexible polymer chains can not take the rolle of solvents. Similarity to the gel is that the electrically induced asymmetric charge distribution causes the bending deformation. Advantages of this system are low electric current, relatively swift deformation at high voltage, and good durability. Disadvantages of this system are requirement of high voltage, small strain, and basically very limited stress. For attaining large strain, very high voltages are necessary for the actuation such as over 10 kV/mm. We show the cases of polyurethane and poly(methyl methacrylate-b-n-butyl acrylate-b-methyl methacrylate) triblock copolymer.

Plasticized polymer system provides another possibility, and we think at this moment the best candidate from the viewpoint of easy processing and variable possibilities. Polymers with large content of plasticizer (we call this category as "polymer gel" in stead of plasticized polymer) shows peculiar deformation such as amoeba-like creep deformation. In some cases, we investigated the gels show high power, high toughness, very low current, and variable application possibilities. The characteristics comes out from the very large dielectric constant from the cooperative interaction between the polymer and plasticizer both of which have very low dielectric constant. By applying the characteristic properties, not only the electro-mechanical function but also the electro-optical functions and mechano-electric functions are found.

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K. Asaka, H. Okuzaki (eds.), *Soft Actuators*, DOI 10.1007/978-4-431-54767-9_12

Through these investigations, we could conclude the dielectric gels have great possibilities as novel type of electro-active materials.

Keywords Actuator • Artificial muscle • Dielectric gel • Electro-mechanical deformation • Electro-optical function • Impact sensor • Light deflection

12.1 General Background

Polymer gel actuator was proposed long time ago using hydrogel [1]. After the discovery of discrete volume change as a phase transition, the researches on the gel heated-up since the variable phenomena have been found and their potential has been recognized [2, 3]. On the electrical actuation of polymer gels have also been investigated from the viewpoint of highly efficient controllable actuator [4, 5]. Most of the researches on these gel actuators were carried out on hydrogels. In the hydrogel actuator, bleed-out of water is usually inevitable, and the practical actuation in air is difficult as it is. In the case of polyelectrolyte gel actuators, they can be actuated at low voltage such as below 10 V in ordre to avoid the electrochemical reaction on the electrodes.

On the other hand, dielectric gel had been considered difficult to be actuated by controlling electric field. However poly(vinyl alcohol) (PVA) gel swollen with dielectric solvent was actuated very efficiently in air without the presence of water [6]. The mechanism was revealed as solvent drag (or "ion drag") induced deformation [7–14]. The motion of the gel was far much faster than that of conventional methods for the gels. Disadvantage of this method is durability in terms of physical strength and bleeding of the solvent.

Elastomer with flexible polymer structure seems to be the best choice for electrically active actuator. We found amorphous polyurethane elastomer (PU) deformes easily by applying dc electric field [15, 16]. The PU as electrically active materials was investigated by controlling soft segment chain length and chemical structure [17]. It turned out that the asymmetrically accumulated charge caused bending strain of the polyurethane films, and the direction can be controlled by modifying the chemical structure of the soft segment [18]. However, the strain of the dielectric elastomers can not be increased without applying very high voltage like 50 kV/mm. There are intensive works on the dielectric elastomer actuators and various successful applications have been proposed [19, 20].

Slightly different from solvent swollen polymer gels, we mentioned plasticized polymers, typically that of poly(vinyl chloride) (PVC). In the case of plasticized PVC (PVC gel), breed-out of plasticizer is very small and almost negligible compared to the highly swollen polymer gels. Durability and stability is far much better from the viewpoint of practical application. The PVC gel showed huge electrically induced elastic-deformation, amoeba-like pseudopodial creep deformation [21–23]. The application has also be investigated as an optical device [24].

The mechanism of the amoeba-like deformation has been explained as an asymmetric charge distribution but not accompanied the solvent drag [23, 25, 26]. Even if the case, the effect is negligibly small. It has been suggested recently that the dielectric properties of the PVC gels are peculiar and depend on the chemical structure of plasticizer [27–30]. They implied the so-called "colossal" dielectric constant.

Plasticizers does not bleed out of the gel body, but the deformation accompanies polymer chain rearrangement together with that of plasticizer, and realized huge strain in the vicinity of near the anode surface [31–33].

New function expected for future possibility of dielectric gels will also be mentioned in this chapter. The colossal dielectric constant of the plasticized gels expands the application [27]. For instance, electro-optical function or the electrical control of refractive index can be possible. We also revealed the proportionality can be varied by changing the gels [34]. The effect observed on PVC gel was much larger than that of nitrobenzene as a control. PVA gel could also show very efficient electro-optical function.

Electrically induced colossal dielectric nature of these materials suggests that the gels can be used as mechano-electric sensor with high sensitivity. The experiment revealed the validity of the idea, and the gels turned out to be an efficient touch sensor or mechano-electrical energy conversion device [35].

Electro-mechanical oscillation was demonstrated on polyethylene terephthalate (PET) film. In the case of PVC gel, PVC and plasticizer, lead to a interesting oscillatory deformation [36]. They are similar phenomena but the mechanism is different each other.

Through these researches, it turned out that not only the dielectric gels but also almost every dielectric polymers have great possibilities as excellent electro-active materials.

12.2 Electroactive Dielectric Actuators

12.2.1 Gels Swollen with Dielectric Solvent

12.2.1.1 Behavior of Dielectric Solvent Under dc Electric Field

When we look back the history of electrical activation of dielectric materials, even an argon gas could be activated by applying an dc electric field [37]. Dielectric solvent has been considered inactive to the electric field in common sense. However, they are revealed to be active in 1960s. First recognized as electrohydrodynamic insability and pressure generation by ion drag [14, 38]. The idea was, then, applied to ion drag pump [39, 40]. Some challenges had been carried out for practical applications [41–44]. (Fig. 12.1 Ion drag)

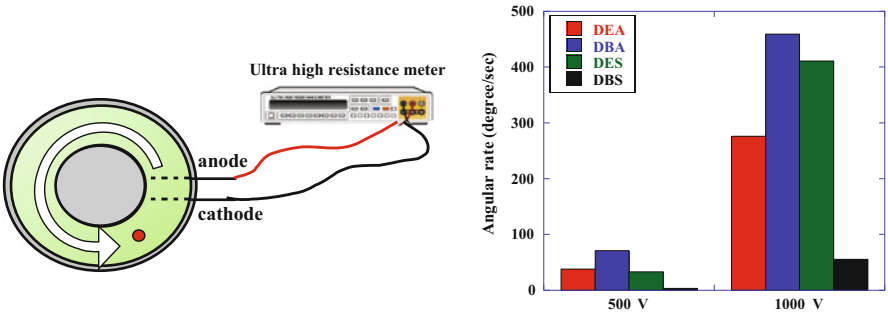


Fig. 12.1 Electrically induced flow of a dielectric organic solvent. Solvent (DMSO) flow was induced from cathode to anode. Electrode (comb) distance is 2 mm. Applied voltage over 100 V is enough for inducing an "ion drag" flow. Flow rate increased with the voltage

12.2.1.2 Highly Swollen Chemically Crosslinked Dielectric Gel

So it is not difficult to expect electrical actuation of the dielectric gels from the fore mentioned researches. However we have no idea of those phenomena when we came across the electrical actuation of an dielectric gel at the first time.

In the research of highly swollen PVA hydrogel, however, we had vast amount experience and knew already the solvent flow or water diffusion in the gels. But if the diffusion causes the bleed-out of solvent, it is not desirable as an actuator. Fortunately, the solvent leak is depressed and some asymmetric pressure distribution was induced in the dielectric PVA-dimethyl sulfoxide (DMSO) gel, resulting in the very efficient electrical actuation in air [6, 45]. Contractile deformation depended on the degree of chemical-crosslinks induced in the gel network. We demonstrated over 10 % of contractile strain could be induced. The voltage required for the contractile deformation was 1 kV for the gel of thickness of 4 mm [9]. Careful observation, however, revealed the gel deformed asymmetrically on the electrodes. The gel tend to swell on cathode and shrink on anode. When the gold sheet was employed as flexible electrodes, although they are fragile, we could have observed the gel bent very swiftly in air [13]. It took 40 ms to attain 90° of bending on the gel of 2 mm thickness and 2 × 1 cm (length × width) [46]. By applying the ion drag theory, the bending deformation could be estimated in the low bending deformation range. At higher bending deformation or at high voltage region the fitting became poor. The phenomena suggested the role of electro-rheological process became important. The other characteristic deformation is "crawling" on the electrode array. The gel could be expanded on the anode by accumulating the charge-tapped solvent ejected from cathode side, on which the gel became thinner, thus, resulting in the worm-like crawling deformation. This type deformation can only be realized at very low degree of chemical-crosslink. For instance in the case of PVA-DMSO gel, the solvent content is over 98 % by weight. Therefore, this type of the gel is difficult for practical application as it is in spite of some advantages (Fig. 12.2 Gel bending, crawling, contractile).

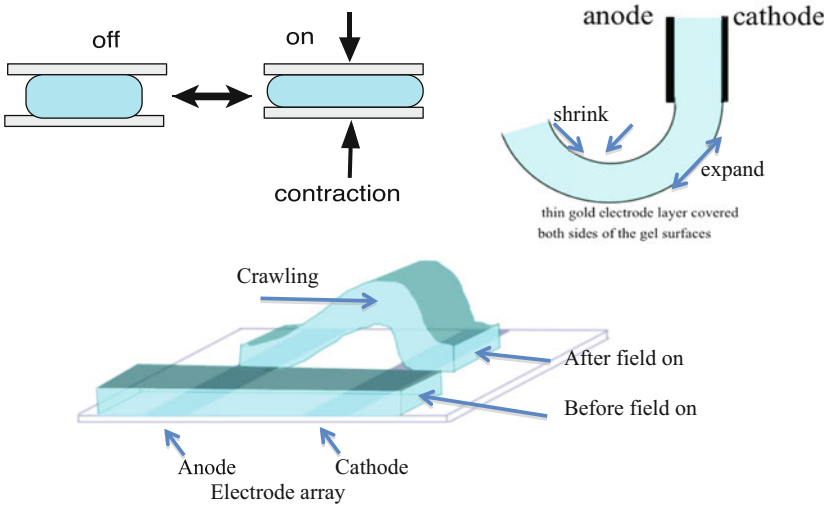


Fig. 12.2 Electrically induced PVA-DMSO gel deformation. (1) Contractile deformation was applied for the fluttering motion of the plastic wing. Strain was over 10 % at the voltage of 250 V/mm, and the rate is ca. 10 Hz. (2) Bending deformation of PVA-DMSO gel. Thickness of the gel was 2 mm. Bending angle increased with voltage. Curves were estimated theoretically. (3) Crawling deformation on a electrode array. Gel rose up on cathode and the gel on anode slide to the cathode

12.2.2 Possibility of Elastomers as Electroactive Dielectric Actuator

From the studies on highly swollen polymer gels, it is clear that solvent leak or bleed out have to be overcome for the practical use in air. To avoid the presence of solvent, the best way is to employ the soft materials without solvent. From this viewpoint, polyurethane (PU) was investigated, since many controlling factors contribute to the physical properties of the elastomer [15]. Detailed analysis on the relationship between the chemical structure and electro-mechanical function revealed several interesting feature of PU [16, 17, 47–49]. However under the voltage range of 10 kV/mm, PU coated with gold nano-layer showed swift bending motion, but could not attain large strain and high durability, neither. Further detailed and systematic development is necessary for the practical application. We could anyway have shown the bending deformation can be controlled by varying chemical structures, such as crosslinking density, functional groups (hard segment and soft segment), etc. The bending direction was turned to be controllable by the space charge distribution. It is well known that the sheets or films of the elastomers have not necessarily homogeneous structures but often have inhomogeneous structure [50], and this makes the controlling factor complicated.

We investigated another type of elastomer from tri-block copolymer. Tri-block copolymer of poly(methyl methacrylate (MMA)-b-n-butylacrylate (nBA)-b-MMA)

was investigated [51]. The concept of the actuator of this material is nano-scale phase separation of each component in the polymer film. If the stiff segment of MMA forms continuous phase the sheet will be electrically inactive because of the stiffness of the sheet. But it will be soft and can be electrically active, when the nBA forms percolated continuous phase and PMMA forms and takes the role of isolated physical-crosslinking point. For attaining the phase separation, we investigated the effect of casting solvent and successfully the continuous phase control was accomplished [52]. However to attain the large strain, the control of segment chain length or mixing small amount of diblock copolymer will be effective. This method will provide high possibility for the efficient control of motility in future.

12.2.3 Plasticized Polymer (PVC Gel)

As pointed out above, highly swollen gel and elastomers from conventional dielectric polymers have some limitations. We changed viewpoint to plasticized polymers. Poly(vinyl chloride) is most popular among the plasticized polymers, and has long experience with wide range of plasticizers, although it has some unpopularity from the viewpoint of chloride containing material.

As a candidate of dielectric material with high dielectric constant, plasticized PVC is the lowest ranking. When we started the electrostriction measurement, actually no striction was detected in d3 direction as expected. But I observed a very slight deformation on the vicinity of anode surface through optical micrograph. That was the amoeba-like pseudopodial creep deformation or electrotactic creep deformation on the anode (Fig. 12.3 Creep).

The creep deformation was first applied for folding type bending motion by Zulhash [21]. It was very efficient, but unusual way of bending, that is, the creep onto the other surface of anode tip leads the gel bend to the direction. The layer crept out on to the anode was very thin but pull the whole gel body bend in the direction finally. When the electric field is off, it swiftly restores original shape. The gel contains anomalously large amount of plasticizer. Content of plasticizer reaches up to 80 % by weight, so we call the plasticized PVC as “PVC gel” (Fig. 12.4 Bending).

The creep deformation of the PVC gel showed fairly large power such as tacking or attractive force to the anode. We have been investigated how to utilize the unique deformation for practical devices. One way is micro-soft gripper that can hold fragile tiny parts for electronic device as an example. The other way is to utilize the creep deformation of the transparent PVC gel directly for instance as a focus controllable lens [24, 53] (Fig. 12.5 Lens, gripper, contractile).

The mechanism of the PVC gel deformation has been clarified to be originated asymmetric charge distribution on anode, that is, negative charge accumulation on the anode plays important role for the deformation [27]. The charge accumulation was suggested to be attributable to the colossal dielectric constant that is induced by applying dc electric field at higher concentration of the plasticizer in the gel. The

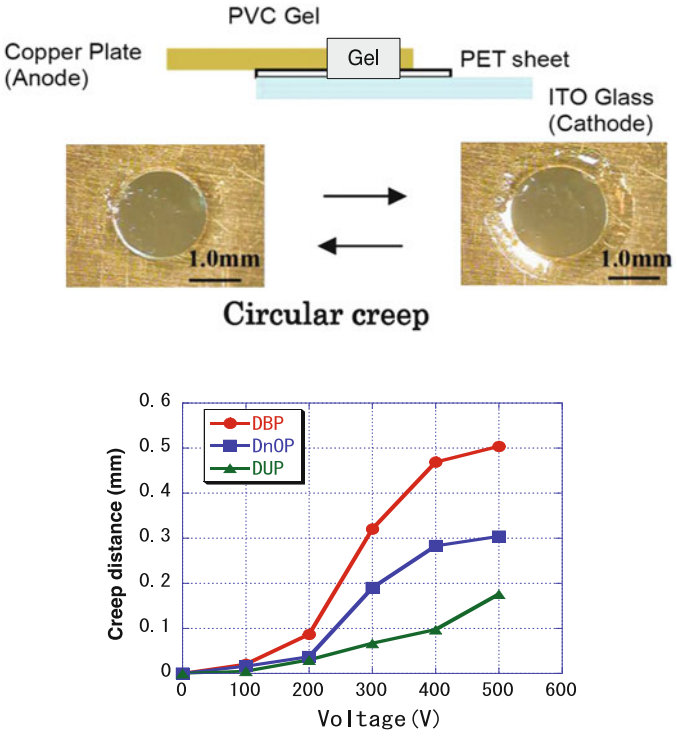
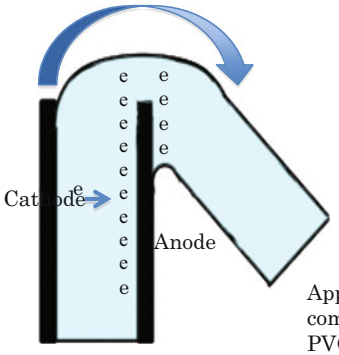


Fig. 12.3 Amoeba-like pseudopodial creep deformation of PVC gel. Creep deformation is non-linear to the applied electric field. Deformation depends very much to the plasticizer



Sample name	Angular velocity / degree s^{-1}
PVC-DEA gel	522
PVC-DBA gel	1162
PVC-DES gel	899
PVC-DBS gel	153

Applied voltage is 2.5 kV/mm, 100 degree bending completed in 100 ms on PVC-DBA gel in air. PVC:DBA=1:8 by weight, thickness=0.25 mm

Fig. 12.4 Bending deformation observed on PVC gel. The gel bends on the tip of anode and totally different from ordinal bending deformation, that is, it is a creep-induced bending. Contractile deformation of Maxwell type is very small and almost neglible. Bending speed is very high

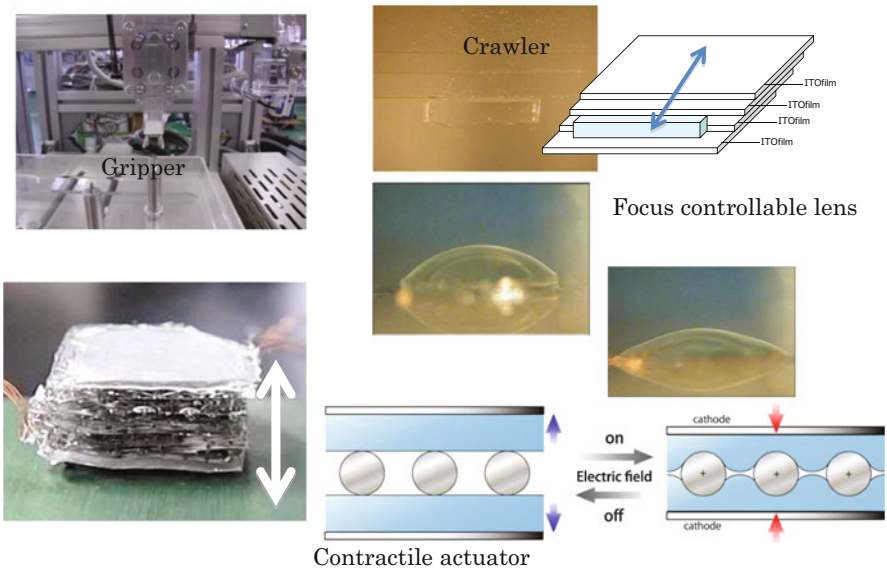
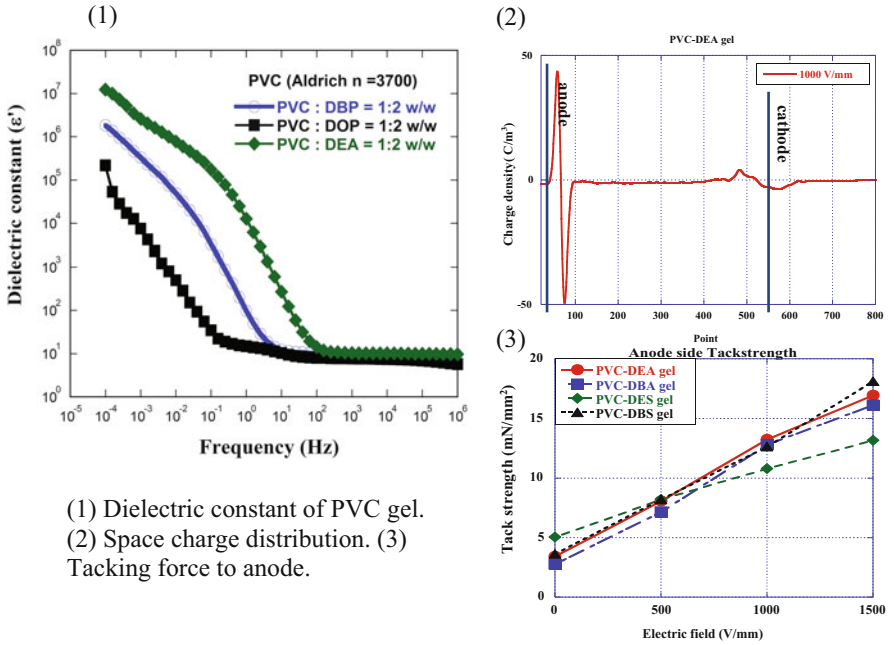


Fig. 12.5 Application of PVC gel for (1) lens, (2) gripper, and (3) contractile actuators. Gripper is for the factory machine. Crawler is for transporter. Contraction actuator has many application. Lens is for optical device

dielectric constant of the PVC gels reaches from 10^3 to 10^4 at 1 Hz around, although those of PVC and plasticizers are usually around 5–10 at most. The phenomena are often occurs for adipate plasticizers but not remarkable for phthalate plasticizers [54], suggesting that the colossal dielectric constant was induced by cooperative phenomena between the polymer and the plasticizer (Fig. 12.6 Ali).

12.2.4 Solid Crystalline Polymer Film

Poly(ethylene terephthalate) (PET) or nylons are most commonly used conventional polymers, and have been known to be inactive to the electric field, and this is true from the viewpoint of high modulus for the deformation and small dielectric properties. But electrostatic behavior of these polymers can be utilized for the actuators, for instance, as oscillators [36]. PVC gel can also be applied for oscillator, and the mechanism is almost the same as that of PET film, although the tacking to anode or creep deformation on to the anode is different from that of PET [36]. The oscillation is an enforced vibration by tacking force or attractive force to the anode only, and repulsive force from cathode does not take any role (Fig. 12.7 PET, PVC gel oscillation).



(1) Dielectric constant of PVC gel. (2) Space charge distribution. (3) Tacking force to anode.

Fig. 12.6 Dielectric charactseristics of PVC gel plasticized with dibutyl adipate (DBA). (1) Permittivity dependence on the frequency, (2) Space charge distribution, (3) Tacking force to anode

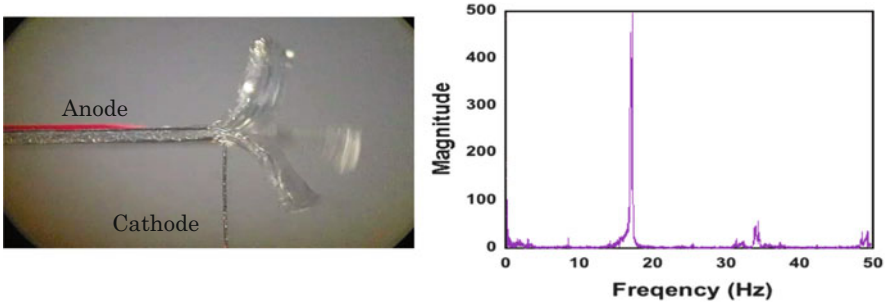


Fig. 12.7 Oscillation of PVC gel. PET can also be a good oacillator, and can be used as micro-fan. Electrode array is different for PVC gel from the case of PET film

12.3 Electro-Optical Functions

On the electro-optical function, the gel, in appearance, implied huge Kerr effect. PVA-NMP gel, on the other hand, showed very large Pockels effect. Once we accept the colossal dielectric constant emerged through the interaction between polymer and plasticizer, the application of the gel expands to various fields. We are investigating on the electro-optical function and mechano-electric function on the

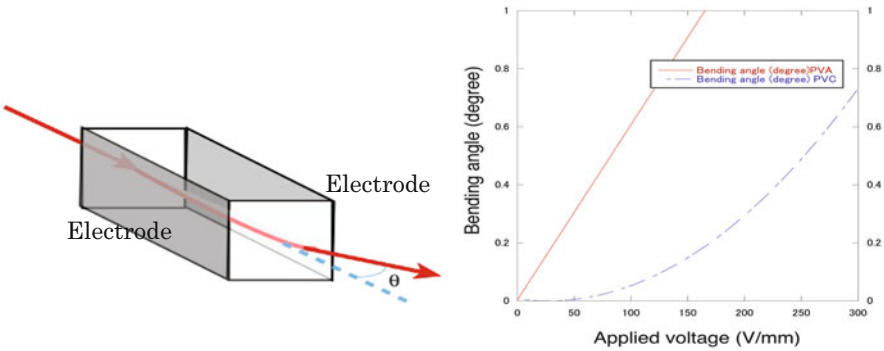


Fig. 12.8 Electro-optical function of dielectric gels (PVC gel and PVA gel)

various polymer systems. On PVC gel, light could be bent by applying a dc field with proportionality to the square of the field. The effect observed was much larger than that of nitrobenzene as a control. PVA gel with *N*-methylpyrrolidone showed proportional dependency to the applied dc electric field [34]. The electrical phenomena did not accompany any serious hysteresis, and on-and-off control was possible. The electro-optical effect is also found to depend on the plasticizer and its concentration in remarkable manner [55] (Fig. 12.8 EO effect on PVC gel and PVA).

As the control, PVC, PVA, plasticizers, solvents (DMSO, NMP) themselves did not show any electro-optical phenomena such as observed in the gels.

The mechanism of these functions are originated from the colossal dielectric properties of the gels and cooperative interaction between PVC and coexisting low molecular weight compounds (plasticizers and/or solvent), although the further details are under investigation. The cooperative phenomena on the dielectric materials have been attracting strong interests for years [56, 57].

12.4 Mechano-Electric Functions

The other function observed on the PVC gels was as an impact sensor [35]. The gels with thickness of submillimeter could generate voltage of ca.1.5 V/cm^2 with the impact of 10 g of steel ball falling from 10 cm height. These functions depend on the combination of plasticizers or solvent coexisting in the materials.

PVC gel can be considered to be homogeneous, although the homogeneity depends seriously on the preparation conditions. The gels did not show any electrostrictive deformation in d3 direction, suggesting the mechanism for the crystalline materials, which has large poling effect, can not be applied, and actually very small voltage could be attained by pressing the gel. The mechanism of the piezo-function of the "homogeneous" non-poled soft polymer material has been considered as the following, that is, PVC rich gel can negatively charge on the

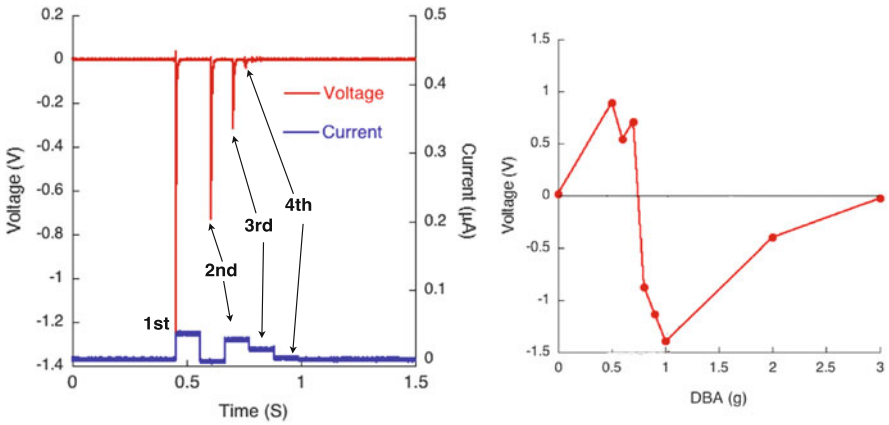


Fig. 12.9 Dielectric gel as an impact sensor. PVC gel implied voltage generation that could be controlled by the content of plasticizer

impact surface because of the friction between polymer phase and plasticizer phase, and in the plasticizer rich gel can bear positive charge on the vicinity of electrode surface. The experimental results can be explained by the triboelectric series [35]. Further detail will be clarified and the method can be applied practically for the sensor and/or energy harvesting systems (Fig. 12.9 Tanaka).

12.5 Concluding Remarks

The dielectric polymers or polymer composites of conventional use can be possible actuators and sensors as have been recognized just recently. Scientific understanding is still underway as this field is interdisciplinary area. I hope the application of conventional polymer in this way might cause very reasonable and big innovation in near future. For example, the nano- and micro-fiber weaving, which has been considered difficult to manipulate such a thin fiber because of the low tenacity, by using electrical motility of the fibers [58, 59]. These technology might be applied for totally new devices of various functions.

We can provide a guideline as described above for the electroactive dielectric gel, although the mechanism is still remains some ambiguity. The parameters that controls the phenomena became clearer during this decade [60]. I am very convincing the future of the soft actuator from conventional polymers, as the control of nano-level control or manipulation of the structure is developing very rapidly. This field will make burst into "nano-tech controlled molecularly assembled devices" for the society of "ambient technology" [61].

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