Development of human-friendly polymeric actuators based on nano-carbon electrodes

— Toward the practical realization of artificial muscles —

Kinji Asaka

[Translation from *Synthesiology*, Vol.9, No.3, p.117–123 (2016)]

Human-friendly machines are expected to increase in demand. To meet this demand, we have developed electrically driven soft actuators based on ionic polymers. This paper describes the development process, design guidelines, current state of R&D, and future prospects for low-voltage, polymeric actuators based on nano-carbon electrodes.

Keywords : Polymeric actuator, soft actuator, nano-carbon, ionic liquid, gel

1 Background—Research of muscles and soft actuators

Japan has become an unprecedented aging society where 22.5 % of the population are 65 years old or older. The use of welfare devices and services that enhance the quality of life (QOL) of the disabled and the elderly and support their social participation are expected to increase. Requirements for such devices are safety, increased operability, downsizing, weight reduction, cost reduction, and others to enable adaptation to unfamiliar environments. The development of soft actuators has been conducted worldwide as a key technology for such devices. The development is being done for various human-friendly medical devices that can be directly worn by people for the purpose of home rehabilitation or care, as well as for communication devices for physically challenged people that allows the transfer of information by tactile or auditory senses while worn on the body.

The goal of soft actuator development is to create an actuator that functions like muscles of organisms, and this is the aim of the researchers around the world. The motive power of movement of living organisms is the muscle, and it is well known that muscles have excellent characteristics as actuators. While excellent actuators with excellent individual specs have been developed, an actuator that is light, soft, and powerful and is capable of working in groups like muscles has not been developed.

Setting the simulation of muscles as a guideline, the research of soft actuators using polymers as basic materials are being conducted around the world.[1] Particularly, the development of an electric-power-driven expanding/contracting actuator using various electroactive polymers is nearing practical use, and there are high expectations. At AIST, since the time of the Government Industrial Research Institute, Osaka, the Agency of Industrial Science and Technology, we have engaged in the research of low-voltage-driven soft actuators with ion conductive polymers. Such a polymeric actuator is light weight, excellent in flexibility, and workability, and major deformation can be obtained at low voltage of 1 V order. Therefore, it is an essential device in advancing the technical development of the aforementioned human-friendly devices.

In this paper, we describe the scenario, the current status, and the future prospect for conducting the development of electrically driven polymeric actuators using nanocarbon electrodes, as the soft actuator technology essential for developing human-friendly devices.

2 Basic soft actuator technology and the guideline for materials of new soft actuator development

At AIST Kansai, the development of ion conductive polymeric actuators was started when the organization was called the Government Industrial Research Institute, Osaka, the Agency of Industrial Science and Technology. This involved a conjugate of fluorine ion exchange resin used in polymer electrolyte fuel cells treated by electroless deposition of precious metals such as platinum and gold. When low voltage of about 1 V is applied between the electrodes, deformation occurs as the counter ions or cations move to the negative electrode (Fig. 1).[2] This technology was developed for the first time in the world for polymeric actuators in 1991, and various application research was conducted based on

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Original manuscript received June 6, 2015, Revisions received July 19, 2015, Accepted July 21, 2015

this technology for active catheters, tactile displays, aquatic micro-robots, and others.[3] Although this actuator was an excellent device, there were many problems that hampered practical use in devices. The main problems were as follows: 1) material costs as precious metals such as gold or platinum had to be used as electrodes; 2) manufacturing costs because the method of electroless deposition took time; 3) surface area was too small to achieve high electrode performance (i.e. capacitance or ability to store ion was small); and 4) since the fluorine ion exchange resin must contain water to maintain ion conductivity, it was difficult to operate in air, and practical use was limited. To solve these problems, we started reviewing the development of a low-voltage-driven soft actuator around 2000 with the following characteristics: a) use of low cost materials; b) manufacturable by a simple process; c) use of electrode materials with large capacitance; and d) use of ion conductive polymers that can be operated in air.

3 Research scenario for the polymeric actuators using nanocarbon and ionic liquids

In 1999, a famous paper on carbon nanotube (CNT) actuators was published in *Science*.[4] This paper reports that when paper-like electrodes are made from single-layered CNTs and voltage is applied to the counter electrode in the aqueous electrolyte solution such as sodium chloride, the CNT electrodes expand and contract. The paper explains that when the voltage is applied, the ions with different charge adhere to the surface of CNTs that compose the electrodes, the electric double layer is created and the CNTs become charged, the state of the graphene bond that comprises the CNTs changes due to a quantum effect, and the CNTs expand and contract. The paper also predicts computationally that an actuator with extremely high energy and power densities can be created considering the large Young's modulus and electroconductivity of CNTs. Moreover, other papers found that CNTs have high electroconductivity,[5] strong mechanical strength,[6] and large specific surface area i.e. large capacitance,[7] and may be ideal as actuator electrodes. Therefore, we decided to use nanocarbons such as CNTs for our electrode material.

For in-air operability that is another issue, we attempted coating the aqueous element surface or impregnating the ion exchange resin with a solvent with a high boiling point instead of water. However, we were unable to obtain any practical element due to various problems. On the other hand, the research of ionic liquids, which is salt that maintains a liquid form at room temperature (Fig. 2), became active around 2000, and this became available for use in device research. This is an organic substance as shown in Fig. 2, and has the characteristics of being liquid at room temperature, of refractory, high conductivity, and stability. That is, it became possible to use this substance like water at room temperature without concern about evaporation.

Based on the above findings, we started reviewing the use of CNTs as the electrodes of ion conductive polymeric actuators, and the use of the ionic liquid system as the electrolytes. At the time, single-layered CNTs available were extremely expensive, but we expected that the price would decrease if they were mass produced in the future. Therefore, we decided the following: to use nanocarbon electrodes such as CNTs to solve problems 1) and 3) described above; to form using a process of printing dispersed CNT electrodes for problem 2); and to use an ionic liquid gel for problem 4).

4 Polymeric actuator research using the bucky gel

At first, we engaged in research starting from improvement of ion conductive polymeric actuators, pasted the aforementioned CNT paper electrodes to the fluorine ion exchange membrane,

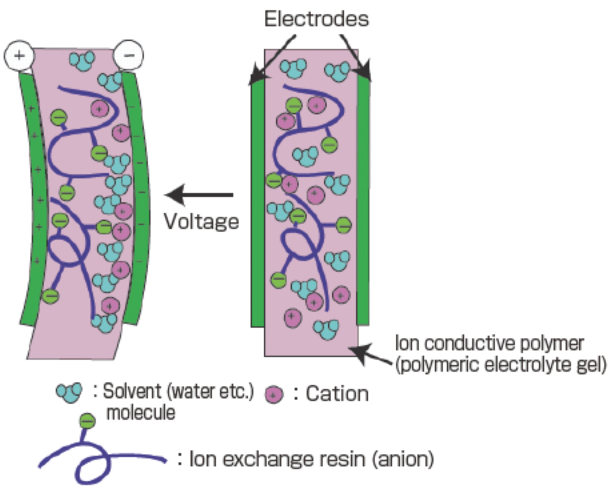
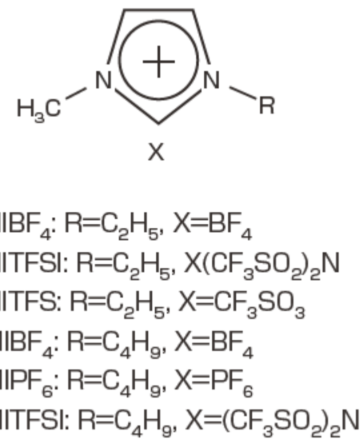


Fig. 1 Schematic diagram of the structure of an ion conductive polymeric actuator and the bending response principle



$EMIBF_4$: $R=C_2H_5$, $X=BF_4$
EMITFSI: $R=C_2H_5$, $X(CF_3SO_2)_2N$
EMITFS: $R=C_2H_5$, $X=CF_3SO_3$
$BMIBF_4$: $R=C_4H_9$, $X=BF_4$
$BMIPF_6$: $R=C_4H_9$, $X=PF_6$
BMITFSI: $R=C_4H_9$, $X=(CF_3SO_2)_2N$

Fig. 2 Structural formula of the ionic liquids used in polymeric gel actuators

soaked them with ionic liquids, and fabricated the actuator element. Yet, we could not obtain good results. During this time, we were introduced to bucky gel research by Researcher Takanori Fukushima (currently, Professor, Tokyo Institute of Technology) of the JST ERATO Aida Project in 2003, and we commenced joint research for its application to actuators. Fukushima *et al*. discovered the phenomenon in which a gel was formed when CNTs and ionic liquids are ground together in a mortar, and this gel was named bucky gel.[8] Fukushima *et al*. also found that this phenomenon occurred as the imidazolium cation of the ionic liquids bonds with the π electron of CNTs, bridges the CNTs, and forms the gel.[8] We started the joint research for using the gel as the actuator electrode.

While the CNTs have various excellent properties as actuator electrodes as mentioned earlier, the problem was how to form them into an electrode. In forming the electrode from the CNT materials, the electrode must be made by dispersing the individual molecules of CNTs so they may come into contact with the electrolytes, or the characteristic large specific surface area cannot be utilized, and the electrode will not store the ions. We thought the reason we failed with the paper electrode described earlier was because of this point.

The technology of the CNT bucky gel using ionic liquids was considered ideal as a manufacturing method of actuator electrodes because it solved the problem of CNT dispersal, and the ionic liquids themselves would become the electrolytes. Moreover, this electrode forming method would be applicable to a low-cost mass production method such as printing and casting. However, A bucky gel of CNTs and ionic liquids only was too soft as an actuator electrode. Therefore, poly(vinylidene fluoride-co-hexafluoropropene) [PVdF (HFP)] that has good compatibility with ionic liquids was added as a mechanical support polymer to form the electrode. Using the ion gel of PVdF (HFP) and ionic liquids as the ion conductive polymer, we succeeded in developing the actuator element with three layers as shown in Fig. 3.[9][10] As described in the initial scenario, this element is a low-voltage-driven soft actuator with the following characteristics: 1) uses materials whose costs can be reduced; 2) can be fabricated by a simple process; 3) uses electrode materials with large capacitance; and 4) uses ion conductive polymers that can be operated in air.

5 Characteristics of the bucky gel actuator and the scenario for its application

The characteristics of the developed bucky gel actuator will be summarized. The manufacture method is extremely simple. The solution, in which the components of the CNT electrode layer and the ion gel layer are well dispersed, is prepared, the solvent is evaporated by casting, electrode films and an ion gel film are manufactured, and the ion gel film is sandwiched with the electrode films and hot pressed together. It is possible to apply this to mass production methods such as printing.

The principle of deformation response is as shown in Fig. 4, and the deformation occurs as the ions move to the respective electrodes upon application of voltage, and the electrode layers expand or contract. A thin actuator film with large deformation at voltage of 3 V or less can be obtained, as shown in Fig. 5. The characteristics of the bucky gel actuator are as follows: 2) it is a thin film, 3) has large deformation, 3) is low-voltage driven, and 4) has good workability. Utilizing these characteristics, developments of basic materials through joint research with the JST Aida Project, and by AIST alone

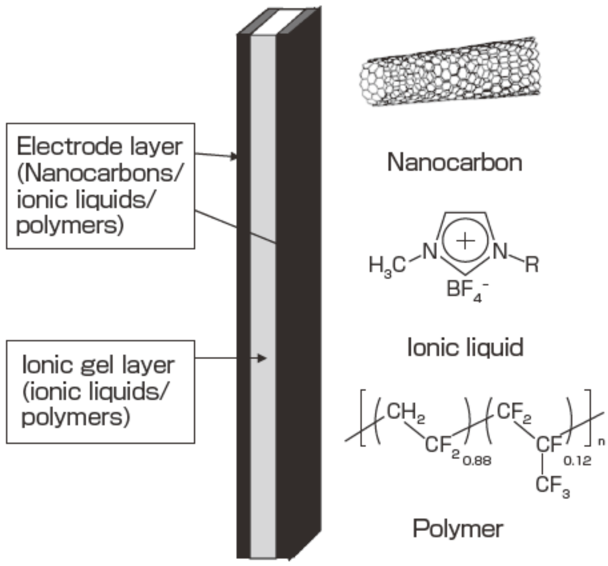


Fig. 3 Schematic diagram of the structure of the bucky gel actuator

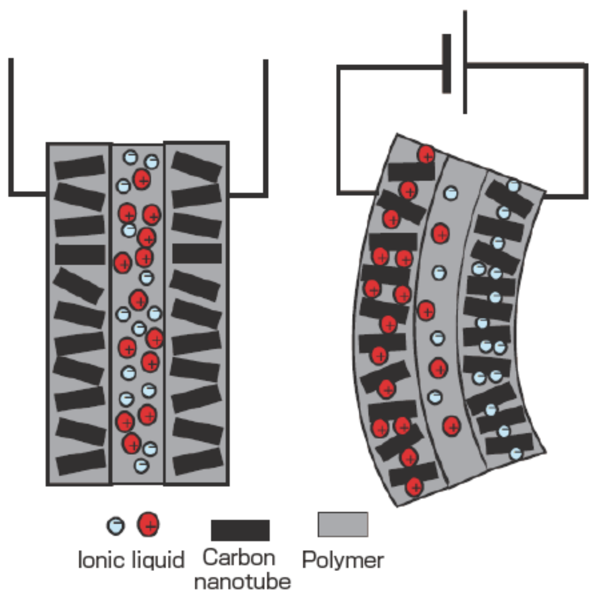


Fig. 4 Deformation response principle of the bucky gel actuator

were conducted. Then, from 2006–2007, the development to achieve a high performance actuator for human-friendly devices was conducted through joint development with a private company.

The human-friendly devices in which the polymeric actuators are needed include the devices worn on the body that require polymeric actuators that are soft, light, and low-voltage driven. The size of the devices can be divided into large human-sized devices such as Power Assist, and thin and light devices such as wearable micro-pumps and portable information display devices. The former was positioned as a future issue that necessitates upsizing technology through polymeric actuator stacking. For the latter, development of materials was continued to realize the application of polymeric actuators to thin and light devices by improving the bending strength, responsiveness, and bending displacement of the actuators.

6 Improvement of the performance of bucky gel actuators

Here, the points in improving the performance of bucky gel actuators are summarized from the perspective of materials development. Since the driving principle of this actuator is the polarization of ions to electrodes, the keys are the development of a material that increases polarization to improve the generative force and displacement, and the development of a material that increases the polarization speed to improve the response rate. Based on this basic way of thinking, the following materials development was started in 2007 jointly with corporations, toward application to thin and light human-friendly devices. Refer to Reference [11] for details of the materials development.

6.1 Improvement of nanocarbon dispersibility

As mentioned earlier, the dispersal of nanocarbons in the electrode layer is closely related to the conductivity, capacitance, and Young's modulus of the electrode layer. By increasing the nanocarbon content with good dispersibility, it is possible to manufacture electrodes with high conductivity and capacitance, and the actuator performance is expected to increase.

The single-layered CNTs have excellent performance but have poor dispersibility, and it was conventionally considered difficult to bring out the individual performance when forming them into electrodes or other products. We used the ionic liquids as dispersants, used various dispersing methods such as ultrasound, ball mill, or jet mill, developed the process for dispersing the single-layered CNTs and manufactured the electrode in which the CNTs were dispersed in high concentration. Also, we found that the actuator performance increased dramatically by adding carbon black (CB) and carbon nanohorn (CNH) to the CNT electrode layer. Therefore, we succeeded in manufacturing an excellent actuator electrode by seeking the optimal dispersing condition of such mixed electrodes.

From the above, it can be inferred that the properties required for electrode nanocarbons are not simply mechanical properties of single elements such as conductivity, initiation stress, expansion/contraction ratio, specific surface area, or Young's modulus, but also involve geometric factors such as aforementioned dispersibility or aspect ratio that appear as the properties of an assembly of elements. The improvements of actuator performance by the addition of CB and CNH are thought to be achieved through the contribution not only of the conductivity and capacitance of CB or CNH, but the contribution of their geometric factors such as enhanced dispersability of CNTs or increased density of the electrode structure.

We have sought the optimal electrode composition and its dispersal conditions through experience, but in the future, we wish to engage in research for quantitative evaluation of the dispersability in relation to actuator performance, to develop a nanocarbon polymeric actuator with higher performance.

6.2 Ionic liquids

The factor that determines the performance of this actuator along with the nanocarbon electrode is the ionic liquids used. Based on the deformation model shown in Fig. 4, we looked at the size difference of the cation and anion in terms of displacement, and looked at the ion conductivity for the displacement rate. Using five types of imidazole ionic liquids and two types of quaternary ammonium ionic liquids that were used frequently in electrochemical devices, we manufactured the actuator films, compared their performances, and sought the optimal ionic liquids. As a result, we obtained guidelines for the selection of the ionic liquids based on the deformation model of Fig. 4 as follows: 1) the rate of response of the actuator is determined by the ion conductivity and electrode conductivity, and 2) the degree of displacement response is dependent on the size difference of the cation and anion.

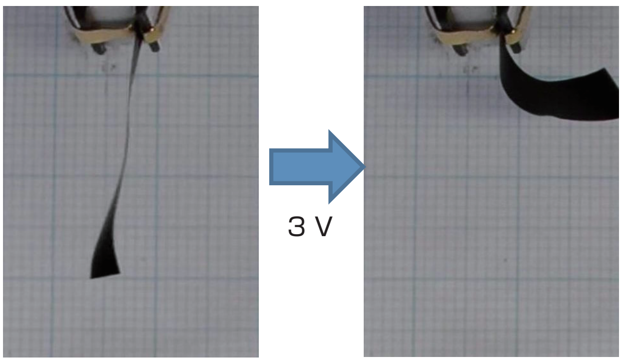


Fig. 5 Deformation of the bucky gel actuator at 3 V applied voltage

Table 1. Comparison of the performances of bucky gel and piezoelectric ceramics actuators

Actuator	Workability	Driving voltage	Displacement	Response	Generating force
Piezoelectric ceramics	×	100 V or more	2 μm	1 ms	100 gf
Bucky gel	○	3 V or less	1 mm	1 s	3 gf

Note: The performance values are representative values of a 5 mm × 5 mm size actuator. The values for a piezoelectric ceramic actuator are literature values of the bimorph type.

6.3 Comparison of the performances of a bucky gel actuator and a piezoelectric ceramics actuator

The bimorph-type actuator made of piezoelectric ceramics is known as the commercially available actuator that undergoes voltage-driven bending deformity. Here, we shall compare the actual values for a piezoelectric ceramics actuator and a bucky gel actuator (Table 1).

For workability, compared to piezoelectric ceramics that requires complex processes such as sintering and polling, the bucky gel can be mass produced by printing and other methods, and it is clear it has excellent workability. Looking at the actuator property, the bucky gel is low-voltage driven and has large deformation, while the piezoelectric ceramics excels in high response rate and large generative power. The bucky gel actuator can be used for products that require low-voltage-drive and large deformation that cannot be handled by piezoelectric ceramic actuators. We believe the issue of low generative power can be overcome in the future by developing the stacking technologies taking advantage of bucky gel's workability.

7 Application of the bucky gel actuator to light and thin braille displays

We worked on several applications in thin and light devices. Here, we shall describe the braille display development that reached the stages of prototype development and user evaluation.

The braille system where the letters are represented by six dots is the only writing system that can be used by the visually impaired. Currently, there are about 300 thousand visually impaired people in Japan, and there are 30 thousand braille users. Due to the advancement of digital information technology in recent years, braille displays have become commercially available as information terminals for the visually impaired. These use the piezoelectric system or solenoid actuators, and in principle, the size of the element becomes large. Therefore, such information terminal devices cannot be carried around conveniently. Moreover, only one line can be displayed. The users have requested for a device that is portable and can display multiple lines that allows easy reading.

Against the above background, under the FY 2009 R&D Project for Service and Support for Persons with Disabilities[12] and FY 2010 R&D Project for Service and Support for Persons with Disabilities[13] of the Ministry of Health, Labour and Welfare, we engaged in joint research for applying the bucky gel polymeric actuator to light and thin braille displays with the following organizations: the Sendai R&D Center, Alps Electric Co., Ltd.; Research and Education Center for Natural Sciences, Keio University; and School of Engineering, the University of Tokyo. If the braille can be presented by arranging the actuator filmstrips in planar configuration and by controlling each element independently with electric signals, it is possible to create a light and thin multi-line braille display. We continued the prototype development for braille displays and created a demo prototype for an ultra-thin light braille display that can display six letters (Fig. 6).

Through evaluation experiments by users using this demo prototype, high expectations were raised for various uses of such thin and light multi-line braille displays. For example, it may be used to replace the liquid display control panels of various home appliances and bank ATMs, or used for page displays through multi-line display. There are also expectations for displaying figures and tables. We also received inquiries from overseas such as Europe and Canada as well as throughout Japan.

In the evaluation experiment, several problems such as durability and reliability of the device as well as fluctuation in properties became apparent. We continued improvement of the actuator and the mechanical structure of the braille display, and saw a clear way for realizing thin and light braille display.

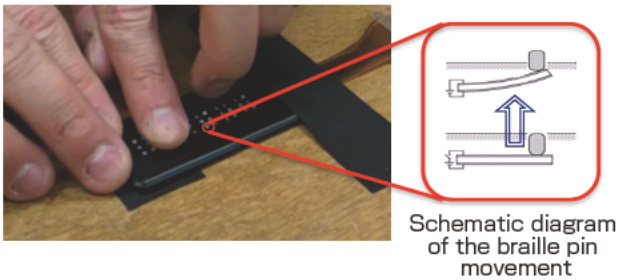


Fig. 6 Braille display using the bucky gel actuator

8 Current status of the polymeric actuator technology using nanocarbons and future prospects

For the practical issues of durability and reliability of the actuator that became apparent through the braille display project, many issues were solved through the NEDO grant and corporate joint research after the completion of the MHLW project. As a result, we succeeded in developing polymeric actuators using nanocarbons that are applicable to various kinds of utilization.[14]

In the future, we wish to realize the application to thin and light devices using this actuator element, as well as the development of a robot actuator such as Power Assist by upscaling through the development of stacking technology.

Acknowledgements

The contents presented in this paper are the results of joint research with the following people. I express my gratitude.

Professor Takanori Fukushima (Tokyo Institute of Technology); Professor Takuzo Aida (the University of Tokyo); Ms. Atsuko Kosaka (Tokyo Institute of Technology); Dr. Kenji Kiyohara, Dr. Takushi Sugino, Dr. Naohiro Terasawa, Dr. Ken Mukai, Mr. Ichiro Takeuchi, Dr. Kenji Hata, and Dr. Don Futaba (AIST); Sendai R&D Center, Alps Electric Co., Ltd.; Professor Takao Someya (the University of Tokyo); Professor Tsuyoshi Sekitani (Osaka University); Professor Yasushi Nakano and Mr. Tetsuya Arai (Keio University).

This paper includes parts of the results obtained through the grant from the following projects: NEDO R&D for the Utilization of Nanotech and Advanced Material (2006–2008); FY 2009 MHLW R&D Project for Service and Support for Persons with Disabilities; FY 2010 MHLW R&D Project for Service and Support for Persons with Disabilities; and NEDO Project for the Development of Innovative Carbon Nanotube Composite Materials to Realize Low Carbon Society (2012–2014).

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Discussions with Reviewers

1 Overall

Comment (Hiroaki Tao, AIST)

This paper presents the realization of a soft actuator that is also called an artificial muscle, by using carbon nanotubes, ionic liquids, and other advanced materials. The background of development, the working principle, and points of materials development are described appropriately. As an example of practical use, the application to braille displays is considered, manufacturing and evaluation are done through corporate joint research, and future points of improvement are reviewed. The flow of the R&D from the development of new technology, product realization, and future prospects are explained, and I think this paper will be particularly useful to the researchers in the field of materials development.

2 Meaning of "human-friendly"

Comment (Noboru Yumoto, AIST)

I think the term "human-friendly" in the title seems to be rather broad. The required characteristics of an actuator differ according to use, and the restrictions of materials and other factors greatly change depending on whether it is used outside or inside the body. Therefore, I think it is necessary to clarify what use you have in mind for the actuator developed in this paper.

Answer (Kinji Asaka)

Compared to other actuators, the nanocarbon polymeric actuator described in this paper has characteristics of being low-voltage driven and operable in air. We did not intend it to be "used inside the body," and I corrected the meaning of "human-friendly" as limited to "wearable on the human body."

3 Superiority of the developed actuator

Comment (Hiroaki Tao)

There is no quantitative description on the performances of the actuator developed in this paper, particularly in comparison with the conventional materials or the materials that serve as world benchmarks. I think this prevents communicating the superiority of your technology to the readers.

Answer (Kinji Asaka)

In Table 1, I added the comparison with the piezoelectric ceramics bimorph actuator that is widely used and commercially available. I also added an explanation of our actuator in the text.

4 Process of improving the performance

Question (Hiroaki Tao)

For improving the performance of the bucky gel actuator, you conducted various experiments and succeeded in developing useful materials and setting the guideline. Did you obtain these through experience of repeated trial-and-error, or did you set up a hypothesis that ion conductivity and molecular size are relevant, and then conduct experiments by selecting ionic liquids with different physical properties to prove your hypothesis?

Answer (Kinji Asaka)

For the optimal ionic liquids, we did a search by setting up a hypothesis based on the deformation model in Fig. 4. Specifically, we looked at the size difference of cation and anion for the degree of displacement, and we looked at the ion conductivity for the displacement rate. I added this process of investigation to the text.

5 Reason for improved performance

Question (Hiroaki Tao)

Why does the performance improve dramatically when you add carbon black or carbon nanohorn?

Answer (Kinji Asaka)

The properties required of electrode nanocarbons are not simply mechanical properties of single elements such as conductivity, initiation stress, expansion/contraction ratio, specific surface area, or Young's modulus, but the geometric factors such as dispersibility and aspect ratio become important as the properties of an assembly of elements. It is inferred that the improvement of actuator performance by the addition of carbon black or carbon nanohorn is due to the contribution of their geometric factors such as improved CNT dispersibility and increased density of the electrode structure, as well as their contribution to conductivity and capacitance. I added this inference to the text.