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Influence of metal plating treatment on the electric response of Nafion

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A potentially promising material as a high performance electroactive polymer gel actuator, Nafion, is known for its fast and large bending upon an applied voltage. Not long ago, it was reported that copper plating on Nafion enhances the degree of its bending. We performed the combinational metal plating treatment on Nafion surfaces with silver, copper and nickel, and the performances of metal plated Nafions—bending curvature and generated force—upon an applied voltage were quantitatively evaluated. From the obtained results, it was speculated that the hydrated mobile ions play substantial roles for the large bending of Nafion as has been widely believed and for the enhancement of its generated force. In addition to them, nickel plating on Nafion surfaces was found to enable Nafion to exhibit a large force without a significant force decay owing to the low-elastic property of nickel layers.

1. Introduction

Gel actuator has attracted researchers attention for the past decades [1–19]. Its expected soft motion, large shape change and low energy consumption by far exceed the conventional metal and ceramics actuators performance. Nafion is well-known material as the most promising future actuator material [9–15, 19]. It exhibits a relatively fast and large bending upon the low voltage as low as a few volts. The bending is reversible by the polarity change of applied voltage and this reversible bending can be continued thousands times. Some applications of Nafion actuator are a catheter, a dust wiper and etc. However, nobody has realized such Nafion products yet. Some of the difficulties lie in the occurrence of bending relaxation, the low generated force and the force decay.

Uchida and Taya reported that they successfully improved the bending performance of Nafion upon a voltage by forming copper layers on its surfaces [19]. We performed some experiments modifying their experiments, and investigated the bending curvature and the generated force of Nafion along with the current flowing through its body as a function of time. From the results obtained through these experiments, we could see a substantial role of mobile ions for the enhancement of bending. And we theoretically found a strong correlation of Nafion bending and its Young's modulus to the force it generates.

2. Experiment

2.1. Specimen preparation

Metal plating on the Nafion surfaces was performed, which primarily serves as a flexible electrodes on Nafion. Three different kinds of metal plating was applied on Nafions surfaces, silver, silver & copper and silver & copper & nickel.

2.1.1. Silver plated Nafion

Nafion surfaces were roughened with a sandpaper. The silver plating solutions, Solution-A and Solution-B, were prepared. Solution-A consists of silver nitrate (0.6 g), 5.0 N ammonium hydroxide (3.0 g) and deionized water (30.0 g). Solution-B consists of D-(+)-glucose (0.4 g), sodium hydroxide (0.8 g) and deionized water (20.0 g). A Nafion sheet was placed in Solution-A followed by the pouring of Solution-B. This process was repeated at least four times to do the better plating on Nafion surface. Silver nitrate and ammonium hydroxide were purchased from Aldrich (Miwaukee, WI). D-(+)-glucose and sodium hydroxide were purchased from NACALAI TESQUE, Inc. (Kyoto, Japan).

The silver plated Nafion sheet was cut into the strips, 20 mm-length × 1 mm-width and dried in the atmosphere for 1 day. Then they were immersed in two different solvents, deionized water and 0.1 M HCl aqueous solution for a day. Nafion swollen with deionized water and 0.1 M HCl were was designated as N-DIW and N-H, respectively.

2.1.2. Silver & copper plated Nafion

First, N-DIW was prepared, and it was plated with copper by following procedure. Copper plating solution consists of copper (II) sulfate pentahydrate (20.0 g), sulflic acid (7.5 g) and deionized water (100.0 g). Nafion

and a small piece of a copper plate connected to the power supply were placed in the copper plating solution, and 1 A-current and 3 V-voltage was applied for 3 s. This plated Nafion piece was cut into the strips, 20 mm-length × 1 mm-width. It was designated as N-SC. Copper (II) sulfate pentahydrate was purchased from Aldrich (Miwaukee, WI). Sulflic acid was purchased from Wako Pure Chemical Industries, Ltd. (Osaka, Japan).

2.1.3. Silver & copper & nickel plated Nafion

First, N-SC was prepared, and immediately nickel plating was performed on its surfaces. Nickel plating solution consists of nickel (II) sulfate hexahydrate (15.0 g), ammonium chloride (1.5 g), boric acid (1.5 g) and deionized water (100.0 g). Nafion and a nickel wire connected to the power supply were placed in the copper plating solution, and 2 A-current and 7 V-voltage was applied for 7 s. This plated Nafion piece was cut into the strips. It was designated as N-SCN. All chemicals were purchased from Wako Pure Chemical Industries, Ltd. (Osaka, Japan).

2.2. Measurement

2.2.1. Bending curvature and current of Nafion

The curvature of all Nafion specimens was measured upon 1 V, as a function of time using an experimental setup depicted in Fig. 1, where Nafion was in the free bending state.

2.2.2. Force and current of Nafion

The force generated by Nafion bending upon 1 V, was measured as a function of time using an experimental setup depicted in Fig. 2. Nafion strip was placed on the fixture attached on the jack. The tip of Nafion was placed on the balance. The force generated by this bending Nafion was measured with this balance, where Nafion length excluding a part of it grabbed by the fixture was 15 mm. As is obvious, the downward movement of Nafion tip is restrained by the balance, namely, Nafion is in the restraint bending state.

2.2.3. Young's modulus of Nafion

Young's modulus of Nafion was obtained. Fig. 3 is an experimental setup. A straight shape Nafion strip was placed on the fixture. On the tip of Nafion, a thread with a small weight (W) was attached. Nafion bent due to this weight, and the coordinate values of this bent Nafion (x, y) defined as in Fig. 3 were obtained experimentally. The coordinate values of Nafion have a relationship with W represented by Equation 1.

$$y = W(x^3 - 3\ell^2 x + 2\ell^3)/6EI \qquad (1)$$

where E, I and ℓ are Young's modulus, the moment of inertia of cross section, and the length of Nafion in the straight shape, respectively. I is defined by $I = bh^3/12$ where b and h are the width and thickness of Nafion specimen, respectively. Since all the values in Equation 1 except for E can be easily obtained experimentally, E can be calculated.

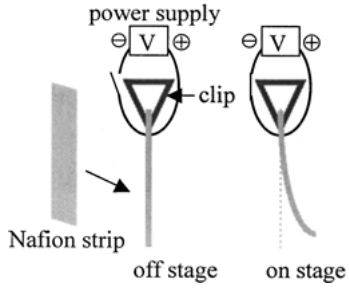

Figure 1 Experimental setup for the measurement of Nafion curvature.

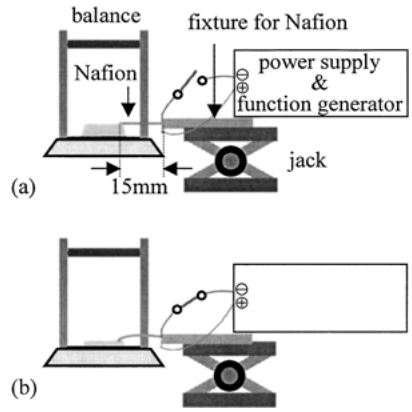

Figure 2 Experimental setup for the measurement of generated force of Nafion: (a) No generated force because of no bending of Nafion upon 0(V) and (b) Occurrence of force generation by Nafion bending upon applied voltage.

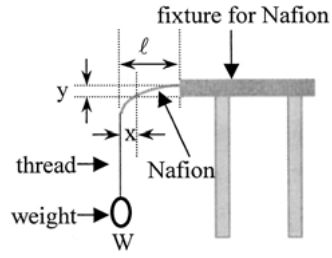

Figure 3 Experimental setup for the measurement of Young's modulus of Nafion W, E and ℓ are the force exerted on the tip of Nafion, Young's modulus of Nafion, and the length of straighten Nafion, respectively.

3. Results and discussions

3.1. Bending curvature and current of Nafion

Fig. 4 shows time dependence of curvature of Nafion specimens upon 1 V, where the positive value of curvature is defined as the bending toward positive electrode direction. All Nafion specimens but N-SCN exhibit quite large bending curvatures immediately after starting imposing a voltage. Especially N-H exhibits an extremely large bending, but it's followed by a large bending relaxation. It's a big issue to be overcome for the purpose of making Nafion actuator.

The bending of Nafion is widely believed to be caused by the shift of hydrated mobile ions contained in its body [9, 15, 19]. If such a concept is right, even a

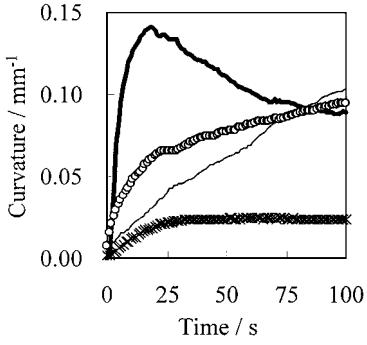

Figure 4 Time dependence of Nafion curvature upon 1 V where the positive value of curvature is defined as the bending of Nafion toward the positive electrode direction. Thin line, fat line, circle, and × mark represent the data of N-DIW, N-H, N-SC, and N-SCN, respectively.

neutral membrane could be bent upon a voltage, when it contains an ionic solution. We performed a following experiment: A neutral dialysis membrane was plated with silver and immersed in the copper plating solution prepared following the description in Section 2.1.2 in order to import mobile ions in it. This dialysis membrane exhibited a bending upon 1 V. Therefore almost undoubtedly the mobile hydrated ions contained in N-DIW, N-H, N-SC, and N-SCN, play a dominant role for the bending. However, the exact mechanism of their bending could not be elucidated fully. Then we performed additional investigations described below, and we'll show our speculations on the cause of these Nafions bending along with our finding obtained from those investigations.

We prepared N-DIW, N-H, N-SC, and N-SN, where N-SN was prepared simply by skipping a copper plating process for N-SCN detailed in Section 2.1.3. Using an experimental setup depicted in Fig. 1, we performed curvature and current measurement for them, where the applied voltage was monotonously raised as given by Equation 2.

$$V(t) = 10\,(\mathrm{mV/s}) \cdot t(\mathrm{s}) \qquad (2)$$

where V and t represent the applied voltage and time, respectively.

Time dependences of the current flowing through Nafion and of their curvature are shown in Fig. 5a and b, respectively. We have to make some comments on this experiment on N-SC as next. The smaller specimens of N-SC were used for the measurement of current, since its value is extremely high out of range of our equipment. Therefore the value of current using a normal size of N-SC is expected to be quite higher than that in Fig. 5a.

Current of all Nafion specimens abruptly increase before reaching $t = 25$(s), and the curvature of them but N-SN exhibit abrupt increase, too, correspondingly. It's in line with the concept of the occurrence of shift of hydrated mobile ions as a cause of Nafion bending. Since all of them have H^+ as mobile ions in common, their bending except for N-SN is speculated to be due to the shift of hydrated H^+. And the extremely large bending of N-H must be due to the import of H^+ through the immersion of it in 0.1 M HCl solution in its preparation

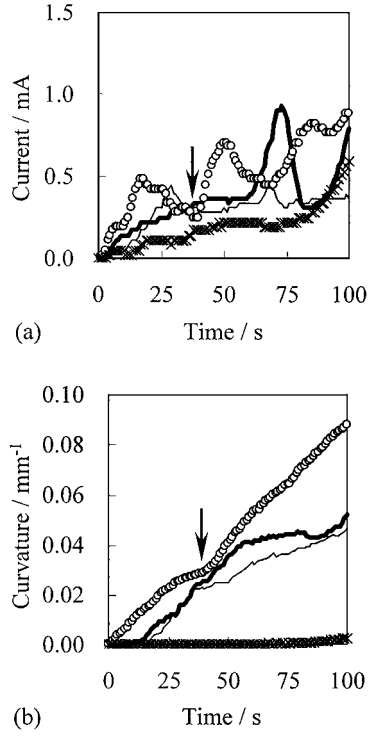

Figure 5 Time dependence of Nafion current and curvature upon $V(t) = 10(\mathrm{mV}) \cdot t(\mathrm{s})$ where thin line, fat line, circle, and × mark represent the data of N-DIW, N-H, N-SC, and N-SN, respectively. (a) Current and (b) curvature.

stage. The cause of no bending of N-SN is explained in the latter part of this section.

The current of N-SC exhibits a quite unique behavior, it has three current peaks. Especially the onset point of second current peak indicated by an arrow in Fig. 5a attracted our attention, where the applied voltage at this onset point is around 0.35 (V). At the very same moment, a discontinuous bending indicated by an arrow in Fig. 5b was observed. This must be due to the occurrence of the reaction $Cu^{2+} + 2e^- \rightarrow Cu$. Namely, N-SC was doped with Cu^{2+} during the copper plating stage. Doped Cu^{2+} ion got hydrated in N-SC and continuously transferred to the negative side around at 0.35 (V). They were converted into Cu atoms by receiving two electrons at the negative side. This reaction process must cause the discontinuous bending as in Fig. 5b. As a matter of fact, it has been already widely known that the reaction $Cu^{2+} + 2e^- = Cu$ occurs at $V = 0.337$(V) (vs NHE) [20]. Since 0.337(V) for the occurrence of $Cu^{2+} + 2e^- \rightarrow Cu$ is measured against NHE, one may say that it is not right to claim that this reaction occurs in N-SC. However, this reaction appears to be even visible in a certain condition as described next. A silver plated Nafion containing copper plating solution was prepared. We observed the color change of its negative surface into red around upon 0.35 (V). This red color must be the color of reduced Cu created by the reaction $Cu^{2+} + 2e^- \rightarrow Cu$. So this reaction must occur to N-SC, too, around at $V = 0.35$ (V), although it is invisible for N-SC due to its originally dark color. Cu^{2+} can be regarded as a strong promoter for the Nafion bending. Here we add some words on this phenomenon. The enhancement of bending of copper plated Nafion, N-SC, must be primarily due to Cu^{2+} doped into N-SC rather

than Cu^{2+} created from copper layer on N-SC. Detail analysis on it is explained in reference [21] written by some of the authors of this pape.

Up to $V = 0.75$(V), virtually no bending was observed for N-SN. Since it exhibits a considerable value of current as in Fig. 5(a), it must contain some mobile ions such as H^+ and Ni^{2+} and etc, where Ni^{2+} was doped into N-SC through the nickel plating treatment. Occurrence of no bending must be due to the large stiffness of N-SN by the nickel plating. Nickel layers on N-SN are quite stiff. Therefore a not-quite-yet-relatively-large bending observed for N-SCN stiffened with nickel layers shown in Fig. 4 must be primarily owing to existence of Cu^{2+} in its body, which leads to the occurrence of reaction $Cu^{2+} + 2e^- \rightarrow Cu$ as described above and cause a bending.

Around above $V = 0.75$(V), all the Nafion specimens including even N-SN exhibit bending. The current behavior corresponding to their bending behavior at this voltage region is quite complicated, and hard to identify what's going on. Yet, Ag^+ must also come to play a role for the bending along with H^+, or H^+ and Cu^{2+} to some extent. All the Nafion specimens have silver layers on their surfaces, and it's been widely known that the reaction of $Ag^+ + e^- = Ag$ occurs at 0.799(V) (vs NHE) [20]. Namely, Ag on the positive surface is ionized into Ag^+ and hydrated at this voltage region. Then it is transferred to the negative side and converted into Ag atoms by receiving one electron. This process must be continuously occurring, resulting in the bending. Although 0.799(V) for the reaction $Ag^+ + e^- = Ag$ is measured against NHE, the reaction $Ag \rightarrow Ag^+ + e^-$ is strongly speculated to occur in Nafion, since $Cu^{2+} + 2e^- \rightarrow Cu$ also appears to occur in Nafion at the close voltage against NHE as described earlier.

3.2. Force, current, and Young's modulus of Nafion

Even though Nafion can exhibit a large bending in the free bending state as in Fig. 4, it's not worth using as an actuator material, unless it generates a large enough force and exhibits no force decay as well as exhibits a relatively large bending. Time dependence of force generated by Nafion specimens upon 1 V is shown in Fig. 6, where it was measured by using

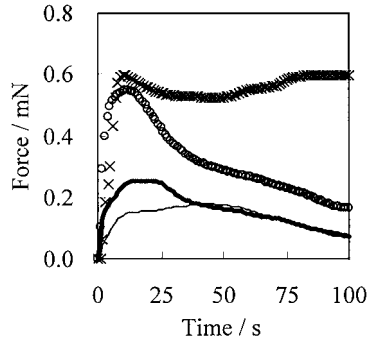

Figure 6 Time dependence of force generated by Nafion upon 1 V where thin line, fat line, circle, and × mark represent the data of N-DIW, N-H, N-SC, and N-SCN, respectively.

TABLE I Young's modulus (E) of Nafion

	N-DIW	N-H	N-SC	N-SCN
E (GPa)	0.09	0.12	0.14	1.24

an experimental setup depicted in Fig. 2. N-SC and N-SCN generate quite large force. Especially, N-SCN did not exhibit a force decay, while N-SC exhibited. It is an attractive facet of N-SCN, yet a long duration time of applied voltage results in the uncontrollability on its force.

As described earlier, the bending of N-SCN is not so large as that of the other even in the free bending state as in Fig. 4, although it exhibited a quite large force as in Fig. 6. Therefore the enhancement of bending is not a sole cause of the generation of large force. There should exist another cause of large force. The most likely cause of large generated force of N-SCN is its large stiffness. Young's modulus of Nafion measured using an experimental setup depicted in Fig. 3 is summarized in Table I. While the bending curvature of N-SCN is far smaller than those of N-DIW or N-H, its Young's modulus is obviously far larger than N-DIW and N-H. Therefore stiffer matrix of N-SCN must play a role of enhancement of its generated force.

Here we discuss the relationship among the force (f), Young's modulus (E), and the bending moment (M_o) of Nafion theoretically. Three of the authors of this paper have proposed a simple evaluation method for the dynamic behavior of Young's modulus of Nafion in the restraint bending state as depicted in Fig. 2 [22]. The theoretical part of it is described next. Fig. 7 is a model of Nafion in the restraint bending state. In this model, the bending of Nafion is assumed to be small, and the vertical force exerted to the tip of Nafion in the process of bending is assumed to be the same as the force, f, generated by Nafion.

$$EI(d^3y/dx^3) + [f - \rho g(\ell - s)] = 0 \qquad (3)$$

ρ, g, ℓ, and I are the mass per unit length of Nafion, gravitational acceleration, the length of Nafion, and the moment of inertia of cross section of Nafion, respectively.

Under the boundary conditions, $y(0) = 0$, and $(dy/dx)(0) = 0$, Equation 4 is obtained from Equation 3.

$$y = -[(f - \rho g\ell)x^3/6 + \rho g x^4/24 + Cx^2]/EI$$
$$C = \text{const} \qquad (4)$$

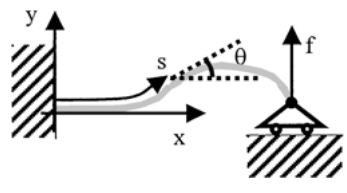

Figure 7 A model of Nafion exerting a force in the process of self-bending upon applied voltage.

Since $y(\ell) = 0$, $y'(\ell) = -\alpha$, and $M_o = EI\alpha$, Equation 5 is obtained from Equation 4, where α is the curvature of Nafion.

$$f = 6M_o/\ell^2 + \rho g\ell/2 = 6EI\alpha/\ell^2 + \rho g\ell/2 \qquad (5)$$

Equation 5 suggests that the force of Nafion has a strong correlation to its Young's modulus and curvature. Even though the bending curvature of Nafion is quite small, the large Young's modulus can make up for it. It agrees with the observation of a large force by N-SCN that is less deformable than N-SC. Another thing, the force depends merely on the bending moment (M_o). Since the maximum forces of N-DIW and N-H are close each other and those of N-SC and N-SCN are almost the same each other, the maximum bending moments of N-DIW and N-H are speculated to be close each other and those of N-SC and N-SCN are speculated to be almost the same.

Since the validity of this theory is limited to the elastic small bending of Nafion [21], it cannot apply to the whole bending behaviors of Nafions. However, the early stage of Nafion bending is speculated to be dominated by the elastic deformation from our observations. Therefore this theory is valid at least in the early bending stage till we can observe the fast increase of curvature as in Fig. 4 and the sharp increase of force as in Fig. 6.

In order to see in detail what's going on in their force generation process of Nafion, we performed force and current measurement for N-DIW, N-H, N-SC, and N-SN, where the applied voltage is given by Equation 2. The results are shown in Fig. 8, where we have to bear in mind that the size of N-SC specimen was intentionally made smaller than normal size due to the same reason described in Section 3.1. However, even small specimens of N-SC exhibit large current out of range of our equipment, yet a few of them exhibit small enough current. We intentionally chose such data of small current and corresponding generated force. Therefore concerning N-SC, we can discuss only the trend of current and force behaviors and cannot discuss their values.

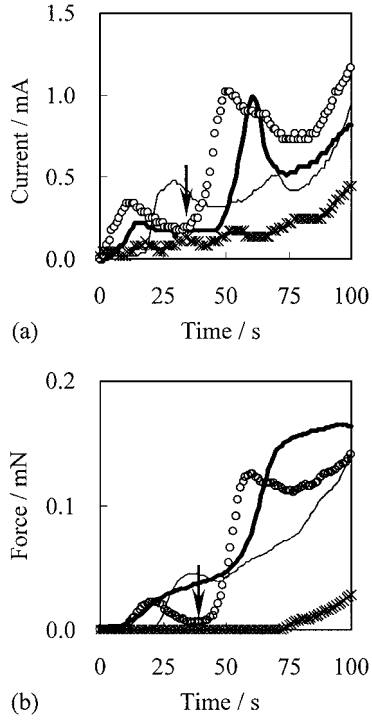

Figure 8 Time dependence of current and force by Nafion upon $V(t) = 10\ (\text{mV}) \cdot t(s)$ where thin line, fat line, circle, and × mark represent the data of N-DIW, N-H, N-SC, and N-SN, respectively. (a) Current and (b) force.

Making comparison of Fig. 8a and b, we can observe unambiguously a direct correlation between the current and force. Abrupt increase of generated force is mostly observed at the moment when abruptly current increases. It suggests that shift of mobile hydrated ions enhances the force generation. The current of N-SC again attracts our attention, especially at the onset point of its second peak of current, its force also abruptly starts increasing, where both indicated by arrows in Fig. 8a and b, respectively. This must be due to the reaction of $Cu^{2+} + 2e^- \rightarrow Cu$.

N-SN did not exhibit a large force as in Fig. 8b, although it was stiffened with nickel layers, which suggests that N-SN tended to bend only to a slight degree. On the other hand, N-SCN exhibits a quite large force as in Fig. 6, which suggests that N-SCN tended to bend more largely than N-SN. From these observations, it is speculated that the large force by N-SCN is due to the strong tendency of bending by the shift of primarily hydrated Cu^{2+} upon applied voltage along with its stiffened matrix, which agrees with our speculation on the cause of free bending of N-SCN described in Section 3.1.

One may think that simply extending the duration time of copper plating for N-SC beyond 3 s could bring us another type of N-SC which is well stiffened with thicker copper layers and highly doped with Cu^{2+}. Such N-SC is expected to exhibit a more effective bending and generate a larger force. In order to answer this question, a copper plated Nafion was prepared by extending the copper plating time of N-SC up to 9 s, three times as long as a normal N-SC, and the time dependence of its curvature and force upon 1(V) was measured, where this specimen was designated as N-SC3. The results are shown in Fig. 9. We could not see any improvement on the bending curvature or the generated force, compared with those of N-SC in Figs 4 and 6, respectively. Furthermore, the copper plating layer is not stable enough unlike a nickel plating layer. It easily comes off the N-SC surfaces. However, doping itself is important for the effective bending of Nafion, since its bending is strongly speculated to be dominated by the mobile hydrated ions contained in it as described earlier. For instance, N-S immersed in the copper plating solution for a day absorbs a large amount of Cu^{2+} and bends largely extremely upon 1(V). But N-SC3 does not appear to be doped more than N-SC judging from the results on its force curvature behaviors. Probably the thicker copper layer on N-SC3 prevents the heavy doping.

Additional thing, there appears to be a trend difference in the current of Nafions between in the free bending and in the restraint bending states especially as for

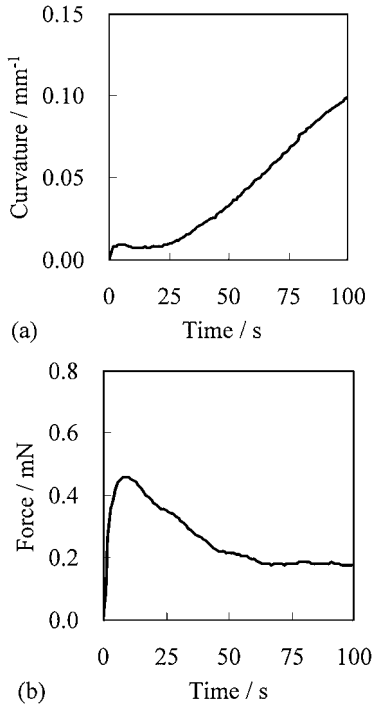

Figure 9 Time dependence of curvature and force by N-SC3 upon 1 V. (a) Curvature and (b) force.

N-SC. Figs 5a and 8a show the current of Nafions in the free bending and in the restraint bending states, respectively. Although we cannot make a direct comparison of the absolute values of current of N-SC because of the reduction of their size and etc, obviously their trend is different each other. The current behavior of Nafion might be quite susceptible to the external force and the degree of its bending. It demands a further investigation in detail.

4. Conclusion

The fundamental properties for the actuator—a relatively large bending and generated force—were given to Nafion through the combination of metal plating with silver, copper, and nickel. It gives Nafion an additional benefit—no force decay. However, we found another problem of N-SCN—uncontrollability of its bending. Overcoming this uncontrollability is one of the important next tasks for us.

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References

1. T. TANAKA, I. NISHIO, S.-T. SUN and S. UENO-NISHIO, *Science* **218** (1982) 467.
2. T. SHIGA, Y. HIROSE, A. OKADA and T. KURAUCHI, *Koubunshironbunshu* **46** (1989) 709.
3. T. SHIGA and T. KURAUCHI, *J. Appl. Poly. Sci.* **39** (1990) 2305.
4. S. UMEMOTO, T. MATSUMURA, T. SAKAI and N. OKUI, *Polymer Gels and Networks* **1** (1993) 115.
5. T. HIRAI, H. NEMOTO, M. HIRAI and S. HAYASHI, *J. Appl. Polym. Sci.* **53** (1994) 79.
6. T. SHIGA, Y. HIROSE, A. OKADA and T. KURAUCHI, *J. Mater. Sci.* **29** (1994) 5715.
7. M. HIRAI, T. HIRAI, A. SUKUMODA, H. NEMOTO, Y. AMEMIYA, K. KOBAYASHI and T. UEKI, *J. Chem. Soc. Faraday Trans.* **91** (1995) 473.
8. K. SALEHPOOR, M. SHAHINPOOR and M. MOJARRAD, in Proceedings of SPIE Smart Materials and Structures, San Diego, 1996, Vol. 2716, p. 116.
9. K. ASAKA, K. OGURO, Y. NISHIMURA, M. MIZUHARA and H. TAKENAKA, *Polym. J.* **27** (1995) 436.
10. R. KANNO, S. TADOKORO, T. TAKAMORI and M. HATTORI, in Proceedings of the 1996 IEEE International Conference on Robotics and Automation, Minneapolis, 1996, p. 219.
11. K. SALEHPOOR, M. SHAHINPOOR and M. MOJARRAD, in Proceedings of SPIE Smart Materials and Structures, San Diego, 1997, Vol. 3040, p. 192
12. M. SHAHINPOOR, M. MOJARRAD and K. SALEHPOOR, in Proceedings of SPIE Smart Materials and Structures, San Diego, 1997, Vol. 3041, p. 829.
13. Y. BAR-COHEN, T. XUE, M. SHAHINPOOR, K. SALEHPOOR, J. SIMPSON, J. SMITH and P. WILLIS, in Proceedings of SPIE Smart Materials and Structures, San Diego, 1998, Vol. 3324, p. 218.
14. M. SHAHINPOOR, Y. BAR-COHEN, T. XUE, J. SIMPSON and J. SMITH, in Proceedings of SPIE Smart Materials and Structures, San Diego, 1998, Vol. 3324, p. 251.
15. K. OGURO, K. FUJISAWA, K. ASAKA, K. ONISHI and S. SEWA, in Proceedings of SPIE Smart Materials and Structures, San Diego, 1999, Vol. 3669, p. 64.
16. T. HIRAI, J. ZHENG and M. WATANABE, in Proceedings of SPIE Smart Materials and Structures, San Diego, 1999, Vol. 3669, p. 209.
17. H. TAMAGAWA, S. POPOVIC and M. TAYA, in Proceedings of Materials Research Society Symposium Electroactive Polymers (EAP), Boston, 2000, Vol. 600, p. 273.
18. H. TAMAGAWA, S. POPOVIC and M. TAYA, *Polymer* **41** (2000) 7201.
19. M. UCHIDA and M. TAYA, *ibid.* **42** (2001) 9281.
20. T. OOSAKAI, K. KANOU and S. KUWABATA, in "Beisikku Denkikagaku" (Basic Electrochemistry) (Kagakudoujin, Kyoko, 2000) p. 189 (in Japanese).
21. H. TAMAGAWA, F. NOGATA, T. WATANABE, A. ABE and K. YAGASAKI, in Proceedings of IEEE ICIT'02, Bangkok, 2002.
22. H. TAMAGAWA, K. YAGASAKI and F. NOGATA, *J. Appl. Phys.*, accepted.

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