

Inorganic Thin Film-based Ionic Decision-maker for Adaptive Artificial Intelligence Technology

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Abstract

In order to achieve highly-integrated device fabrication in an ionic decision-maker (IDM) for solving multi-armed bandit problems (MBPs), mesoporous silica (MS) proton conducting thin film has been applied to its fabrication, where a thick Nafion proton conducting polymer electrolyte has previously been employed. Whereas the proton conductivity of the MS thin film ($1.7 \cdot 10^{-4}$ S/cm) was lower than that of Nafion (approx. 10^{-2} S/cm), the MS-IDM showed good solving properties, which are similar to Nafion-IDM and CPU-based conventional computing. Furthermore, fluctuations in the correct selection rate (CRS), which was previously observed with Nafion-IDM, and reached CRS were significantly improved with MS-IDM.

1. Introduction

Artificial intelligence (AI) systems that are capable of quickly making optimum decisions (selections) in changing environments are required in various fields. While the usefulness of CPU-based AI has been verified, one critical drawback is inherent in such method: *i.e.* exponential increases in the amount of computational resources required for processing the vast streams of information required. Such conventional methodology for computations is hence reaching its limit, leading to the intensive exploration of breakthrough computational methodologies.

We previously reported an ionic decision-maker solving a multi-armed problem (MBP) through the inherent nonlinearity and nonvolatility observed in the charge-voltage relationships of electrochemical cells [1]. Said ionic decision-maker successfully solved MBPs, with good levels of performance similar to those achieved with conventional CPU-based computation methods. However, the device configuration, consisting of 100- μ m-thick polymer electrolyte with a Pt/C catalyst layer, was not advantageous for integrated device fabrication. Furthermore, undesirable and mysterious fluctuations in correct selection rate (CSR) were found in its learning behavior. In the present study, mesoporous SiO₂ thin film has been applied to the development of an inorganic thin film-based ionic decision-maker (IDM). The solving properties of MS-IDM were compared with a Nafion-IDM and conventional CPU-based computations.

2. Experimental

Multi-armed bandit problems

MBPs are mathematical problems in which a gambler facing many slot machines (SM) with various reward probabilities has to determine, on the basis of his experience (trials), which SM to play so as to maximize the total reward. Figure 1 illustrates the MBP for one user and two SMs, where the user can choose only a single SM at any given time. Suppose that at a certain time (t), the user chooses either SM A or B, which give rewards with probabilities P_A and P_B and no rewards with probabilities $1 - P_A$ and $1 - P_B$, respectively. In advance of the trials, the user has zero knowledge of the value of P_A and P_B . The results of trials are used to calculate the correct selection rate, which is the main concern of this study.

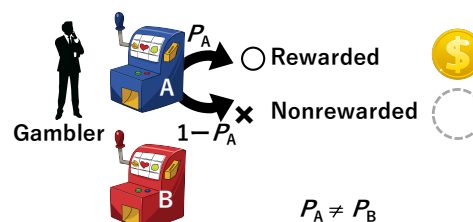


Fig. 1. Illustration of MBP in which a gambler needs to select a SM.

Ionic decision-maker fabrication and operating principle

Figure 2 illustrates our ionic decision-maker, which consists of a 500-nm-thick mesoporous SiO₂ (MS) thin film. A MS thin film was deposited using a sol-gel method using Si(OC₂H₅)₄ (TEOS) and CH₃(CH₂)₁₅N⁺(CH₃)₃Br⁻ (CTAB) as a starting material and template organics, respectively [2]. To achieve MS thin films, the MS precursor was spin-coated onto the flat surface of a SiO₂ glass substrate, then subjected to heat treatment. Special care was taken to prepare a viscous MS precursor, with a long aging time before spin-coating, in

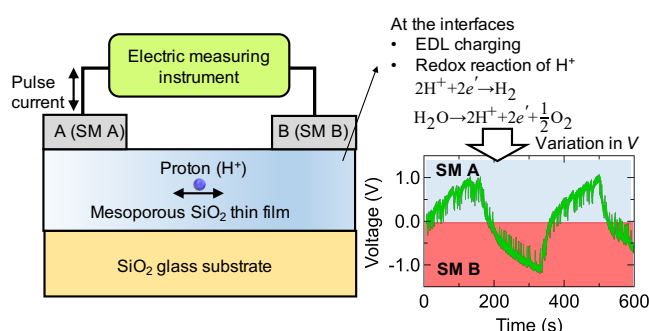


Fig. 2. Illustration of our ionic decision-maker, consisting of mesoporous SiO₂ thin film (MS-IDM).

order to achieve a relatively thick MS with low in-plane resistance. Platinum electrodes were deposited on the surface of the MS using RF sputtering with a stencil mask.

First, in the open circuit condition, the sign V of the MS-IDM is judged (see inset in Fig. 2). When V is positive (negative), SM A (B) with P_A (P_B) is played. In accordance with the results [rewarded or nonrewarded, as illustrated in Fig. 1], a pulse current of 500 nA, or -500 nA, is applied to electrode A for 500 ms. The circuit is then opened, and V is measured. This sequence of these steps is defined as one play. As shown in Fig. 2 inset, repeated plays resulted in a digital-like V variation, which was used to make a correct decision and calculate a correct selection rate. Refer to reference 1 for the operation details [1].

3. Results and discussion

MS-IDM proton conductivity

An AC impedance spectrum of the device, measured at 298 K, is shown in Fig. 3. The semicircle in the high frequency region and the electrode response in the low frequency region were good indications of proton conduction in the device. By fitting of the spectrum with a typical equivalent circuit of electrochemical cells, proton conductivity was calculated to be $1.7 \cdot 10^{-4}$ S/cm, which is a typical value for MSs at RT.

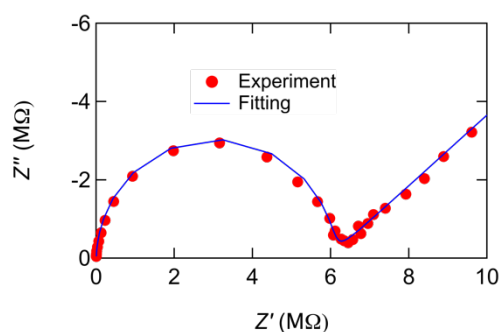


Fig. 3. AC impedance spectrum of MS-IDM and the fitting result assuming a serial connection of two unit cells.

Solving properties of MS-IDM with dynamic MBPs and comparison with Nafion-IDM and CPU-based computing

Figure 4(a) shows the variation in CSR of the MS-IDM device. The experiment started from the initial condition $(P_A, P_B) = (0.6, 0.4)$. During the experiment, P_i inversions were applied [e.g., $(0.6, 0.4)$ was inverted to $(0.4, 0.6)$]. At the beginning of the experiment (i.e., the number of plays was zero), CSR was about 0.5. This was reasonable because the device had no information about channels A and B; the ionic decision-maker randomly selected channels. However, as the number of selections increase, CSR showed a gradual increase toward 1.0 (i.e., 100%), which is indicative of empirical learning on the basis of the results of plays. The learning process corresponds to an electrical response of the cell, in which repetition of selections made V take a positive or negative value. The V variation is caused by modulation in the concentration distribution of protons, gas and water molecules near electrodes A and B. By comparing the performance with a conventional CPU-based computation using ϵ -

greedy algorithm, which is a typical algorithm for solving MBPs, it can be seen that MS-IDM successfully solves the MBP.

Figure 4(b) shows MS-IDM performance compared with Nafion-IDM [1]. Whereas the adaptation seen in the MS-IDM was slightly slower than in the Nafion-IDM, fluctuation in CSR, which was significant in Nafion-IDM, was much improved in the MS-IDM. Furthermore, with MS-IDM, the 0.96 average reached CSR after 200 plays is higher than that for Nafion-IDM (0.92). Although the mechanism of the improvement achieved by the MS is not clear at present, one possible reason is a chemical stability in the MS SiO_2 matrix, which is superior to that of the Nafion matrix with various functional groups.

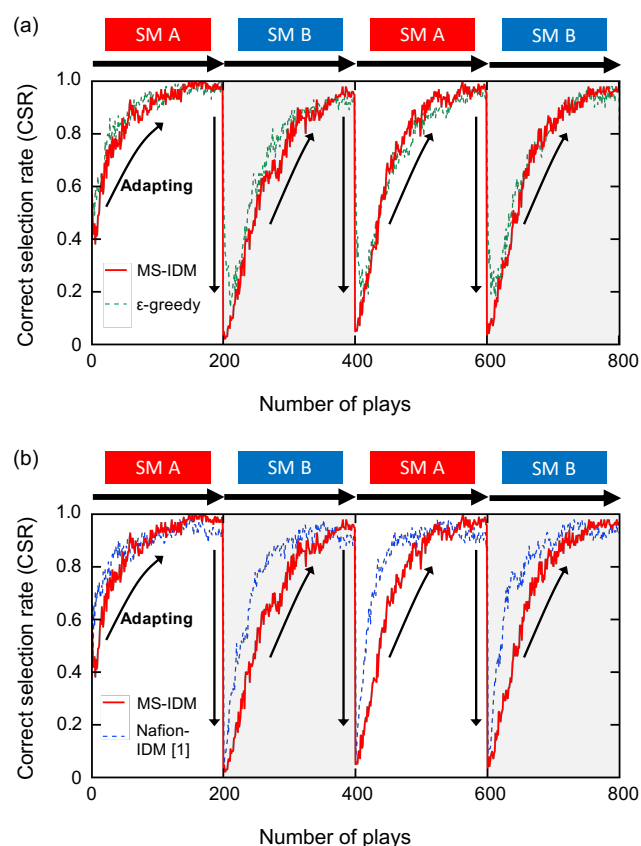


Fig. 4. Comparison of variation in CSR of MS-IDM (a) with (a) ϵ -greedy algorithm, and (b) with Nafion-IDM.

4. Conclusions

An inorganic thin film-based IDM has been developed, on the basis of the protonic conduction of MS thin films, for highly-integrated device fabrication. Whereas the proton conductivity of MS was lower than that of Nafion, fluctuations in CSR and reached CSR were much improved, indicating that the chemically-inert nature of the SiO_2 matrix is highly suited to enduring the repetitive application of pulse currents and to the achievement of better decision-making abilities.

References

- [1] T. Tsuchiya, T. Tsuruoka, S.-J. Kim, K. Terabe, M. Aono, *Sci. Adv.* **4**, (2018) eaau2057.
- [2] T. Tsuchiya, T. Tsuruoka, K. Terabe, M. Aono, *ACS Nano* **9**, (2015) 2102-2110.