Electronic Supplementary Information

Reservoir computing with the electrochemical formation and reduction of gold oxide in aqueous solutions

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1. Experimental details

Electrochemical measurement

Electrochemical cell was a 30 mL glass Erlenmeyer flask. Electrodes (gold wires) were immersed in the solution through a stopper made of silicon rubber. The experiment was carried out in a box filled with N_2. A reference electrode is prepared by electrochemical oxidation of a gold wire in the solution used for the measurements.

Electrochemical measurements were carried out with a Potentiostat (PS–06, Toho Technical Research, Japan). Voltage wave form is generated by digital-to-analog converter (USB-6002, National Instruments) and current signal is recorded by analog-to-digital converter (USB-6002, National instruments).

Image classification task

General Explanation

Image classification task using neural network is one of the most frequently used evaluation of supervised machine-learning model. Here, we will explain an example of handwritten digit recognition.

First of all, hand-written images of digits with “True Label” which indicates the number represented by the image are prepared (examples are shown in Fig. ICT1). The standard image sets are available from *The MNIST DATABASE* (http://yann.lecun.com/exdb/mnist/index.html).

Fig. ICT1. Example of the MNIST handwritten digit images.

Then, image data is fed to the input node of neural network (NN) model. A scheme of the NN is shown in Fig. ICT2. Usually, image pixel is used as input data. In case of MNIST, an image consisted of 28 × 28 = 784 pixels. So, 784 input nodes are prepared. When the digit is from 0 to 9, 10 output nodes, representing digits from 0 to 9, are prepared. The NN is tuned so that the input is related to the output nodes representing the correct digit. The tuning of NN is carried out to thousands of image data. Construction and training of NN can be carried out with an open-source neural network library such as Keras (https://keras.io/).

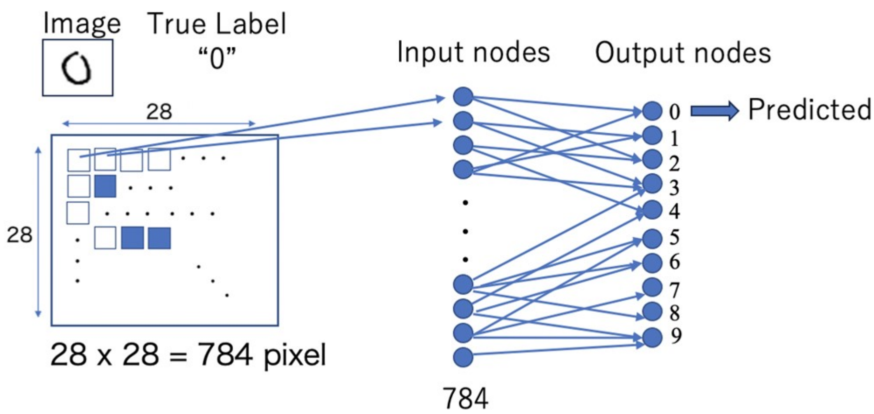


Figure ICT2. Scheme of data flow in image classification task using NN.

Procedure for reservoir computing

The scheme of the image-classification using electrochemical reservoir is shown in Fig. ICT3.

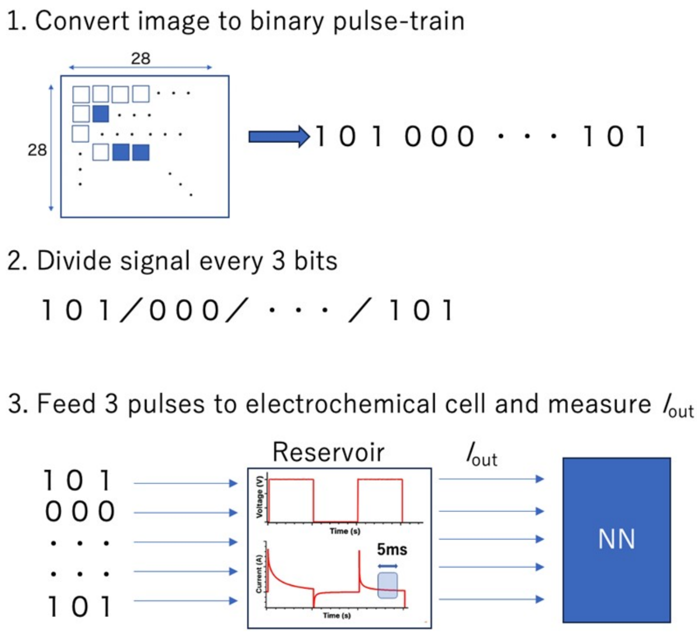


Figure ICT 3. Scheme of data flow in image classification task using NN.

The gray-scale images of the MNIST data were converted to black and white. The 28 × 28 pixels data were converted to 1-dimensional 784 pulse-train. The pulse signals were fed to the electrochemical cell every three bits and the I_{out} was measured. As a result, the image data was converted to the vector consisted of 262 I_{out}. A super-vised learning was executed using these 262 I_{out}.

The machine-learning code attached as ESI was built by referring tutorial for Keras. The readout layer consisted of a single layer perceptron with 262 input nodes and 10 output nodes which had a softmax function in the output nodes, based on the error backpropagation that uses the RMSprop method to minimize the cross-entropy loss by updating each weight between the input and output. Each output node corresponds to the label number from 0 to 9. The epoch number and batch size were 20 and 8, respectively. Overtraining was checked by comparing accuracy of the training data with that of the test data. Google Collaboratory was used to conduct machine-learning.

The source code and data used were attached as separated files.

2. Relationship between Au oxidation-reduction and short-term memory

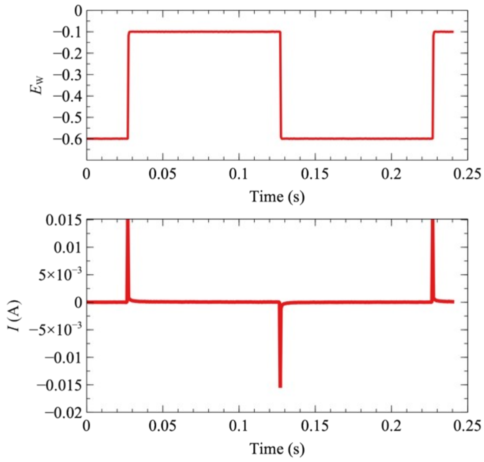


Figure S1 The *I* transient in the order of 0.1 s was not observed when the voltage for [1] and [0] were set to -0.1 V and -0.6 V, respectively, where *gold oxidation-reduction* does not occur. This result indicates that Au oxidation-reduction is required to produce current transient in the order of 0.1 s as shown in Figure 2.

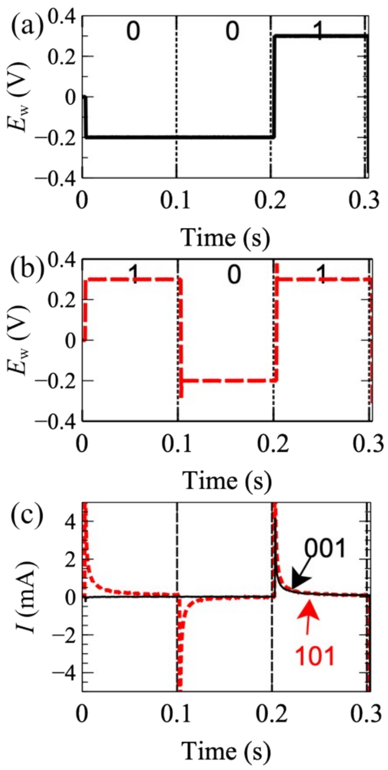
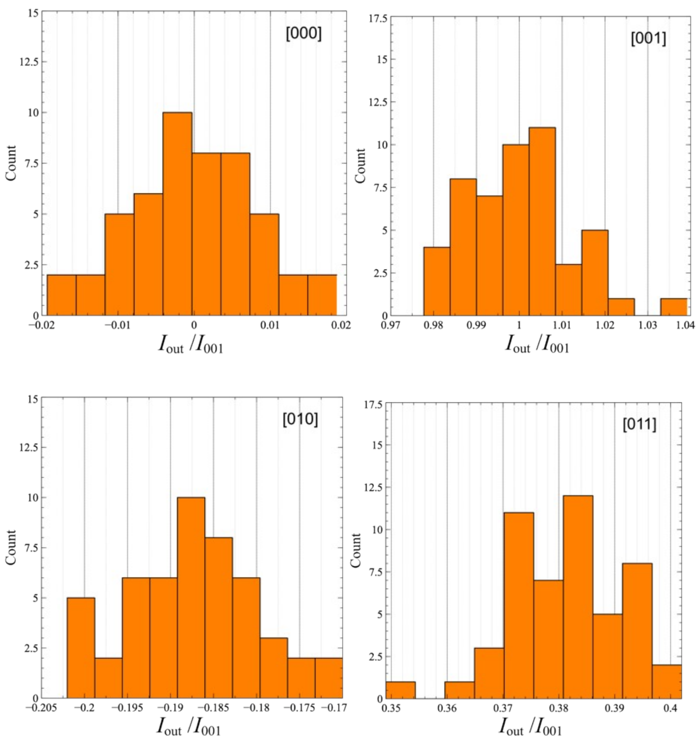


Figure S2 The I transient when the [1] and [0] were set to 0.3 V and -0.2 V at which gold oxide was reduced. No difference in current transient was observed in I. This result indicates that gold oxide left on the surface is required for the short-term memory.

3. Statistics and histogram of I_{out}/I_{001}

Table S1 Statistics of I_{out}/I_{001}

Bit pattern	Average	Standard Deviation
000	-0.00019	0.0082
001	1.0	0.012
010	-0.19	0.0074
011	0.38	0.010
100	-0.10	0.0084
101	0.41	0.0094
110	-0.17	0.0085
111	0.23	0.0086



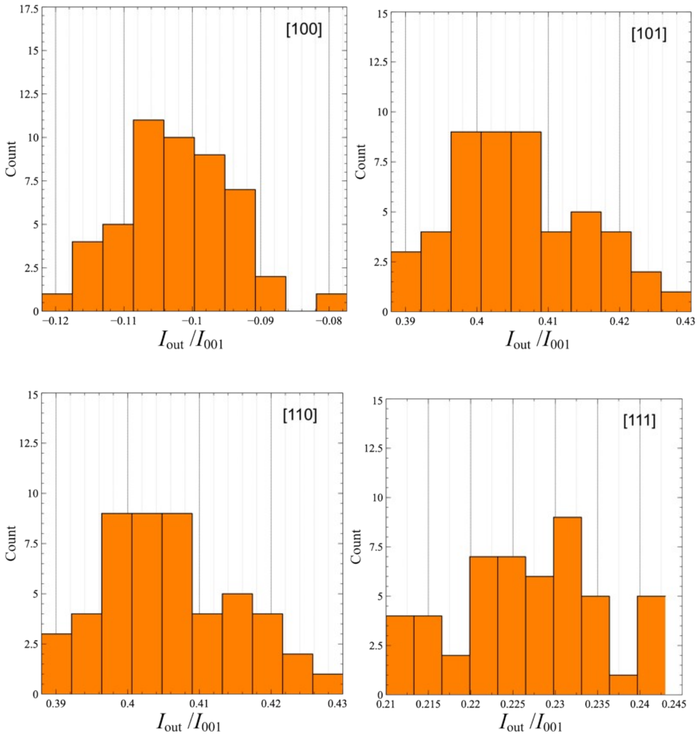
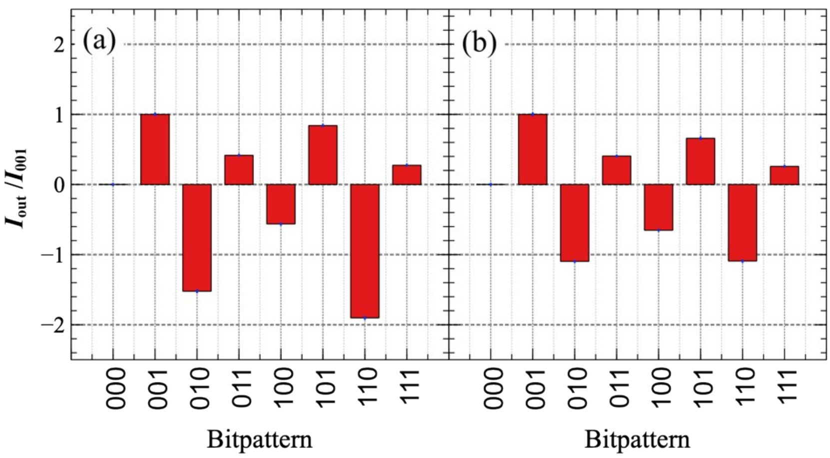
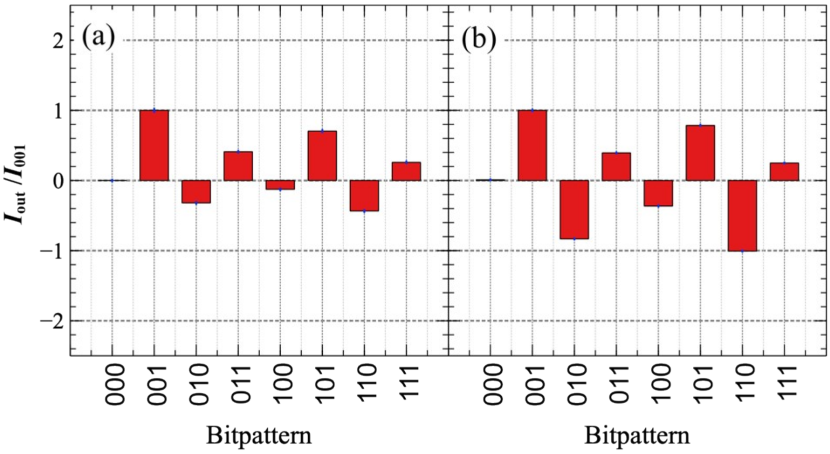


Figure S3 Histograms of I_{out}/I_{001}.

4. Dependence of the pulse condition on the 3bit recognition



Figures S4 I_{out}/I_{001} dependence on the pulse condition, showing the tunability of I_{out}. [1] = +0.3 V, [0]= (a) -0.15 V and (b) -0.1 V. As the [0] was set to more negative voltages, the I_{out} for the patterns ending [10] becomes relatively larger, representing the reduction current of gold oxide increased.



Figures S5 show I_{out}/I_{001} dependence on the pulse condition, showing the tunability of I_{out}. [0] = -0.1 V, [1] = 0.1 V(a) and 0.4 V(b).

5. Concentration dependence of the current response

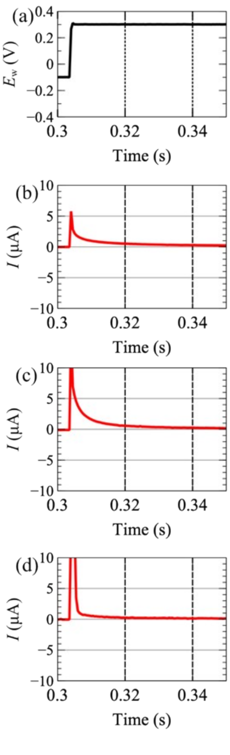


Figure S6 The I transient at the final bit [1] in [101] where [1] = 0.3 V and [0] = 0 in (a) 0.01 mol/L, (b) 0.1 mol/L and 1 mol/L of $HClO_4$. The peak current become larger due to the fast double layer charging at higher concentration. The decay becomes faster as the concentration increased, showing the tunability of response speed.