Supplementary Information (SI) for Journal of Materials Chemistry C.

Supplementary Information

Reservoir Computing Determined by Nonlinear Weight Dynamics in Gd-doped CeO_2/CeO_2 Bi-layered Oxide Memristors

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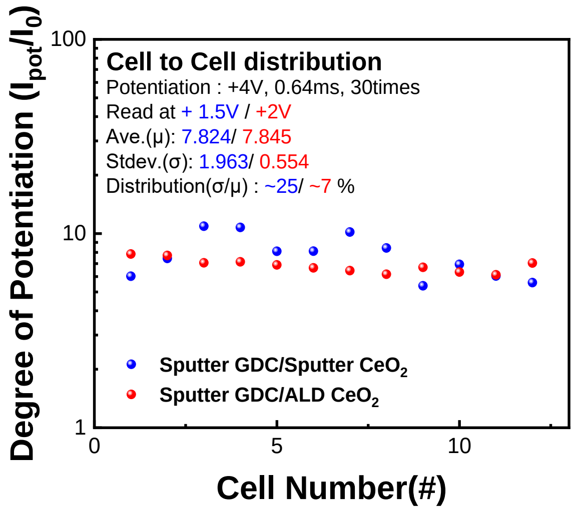


Fig. S1. Cell-to-cell distribution the degree of potentiation results of the device using sputter-deposited bilayers and the device using sputter GDC/ALD CeO_2.

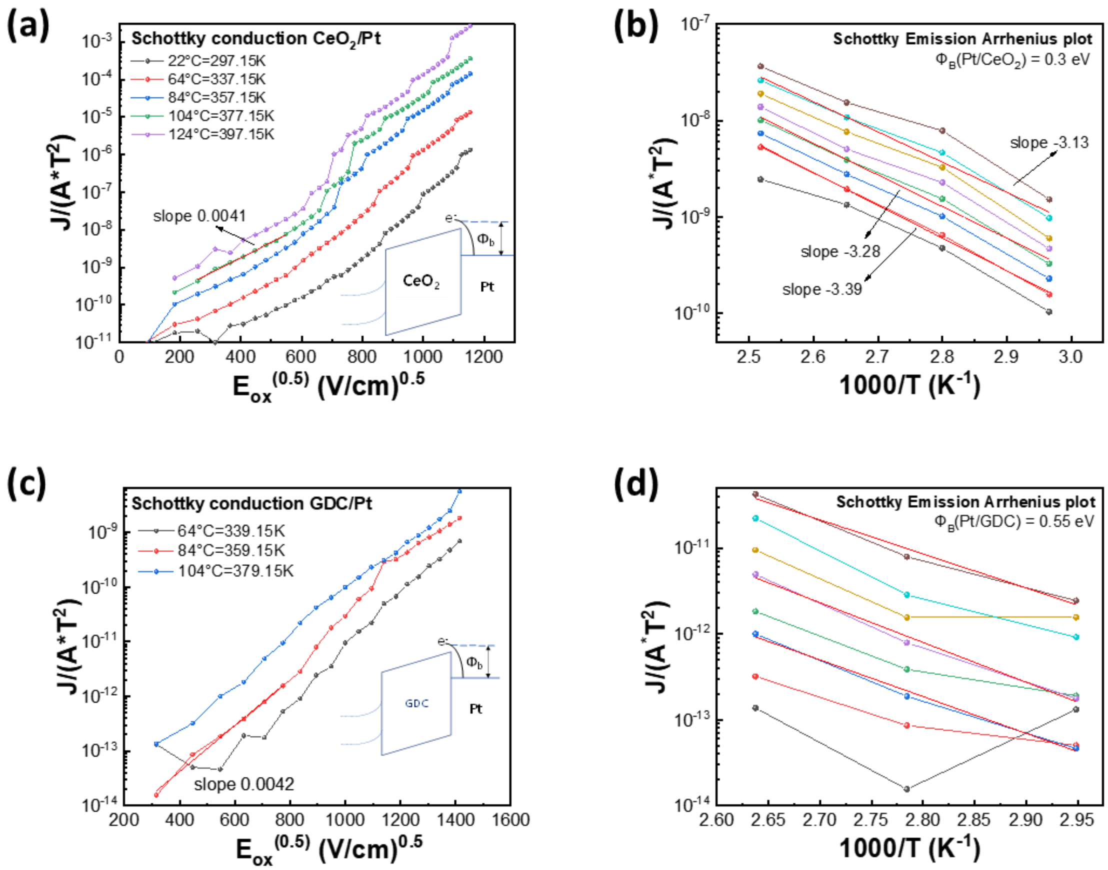


Fig. S2. Calculation of Schottky barrier height. *I*–*V* curves in the form of Schottky conduction with the relation of (**a**) log(J/A^*T^2) with respect to $E_{ox}^{0.5}$ at the temperatures of 297.15, 337.15, 357.15, 377.15 and 397.15 K with the band diagram of Pt/CeO_2 interface (inset) and (**b**) log(J/A^*T^2) with respect to 1/*T*. *I*–*V* curves in the form of Schottky conduction with the relation of (**c**) log(J/A^*T^2) with respect to $E_{ox}^{0.5}$ at the temperatures of 339.15, 359.15, and 379.15 K with the band diagram of Pt/GDC interface (inset) and (**d**) log(J/A^*T^2) with respect to 1/*T*.

For the Schottky conduction mechanism equation,

$$J = A^{*}T^{2}exp\left[-\frac{q\phi_B}{k_bT} + \frac{1}{k_bT}\left(\frac{q^3E_{ox}}{4\pi \mathrm{k_d}\varepsilon_o}\right)^{1/2}\right] \qquad \text{(S1)}$$

where J is the current density, A^* is effective Richardson constant, $q\phi_B$ is the Schottky barrier height, T is the absolute temperature, q is the elementary charge, ε_0 is the absolute dielectric permittivity of vacuum, E_{ox} is the electric field across the CeO_2 and GDC layer, k_b is the

Boltzmann's constant, k_d is dynamic dielectric constant of CeO_2 and GDC layer as extracted from the slope of the graphs in Fig. S2a and Fig. S2c. To calculate Schottky barrier height $q\phi_B$, the curves were replotted in the relation $\log(J/A^*T^2)$ with respect to $1/T$. From the slopes of these curves, the Schottky barrier heights of CeO_2 and GDC thin films were determined to be 0.3 eV and 0.55 eV.

Pulse / Interval	+5Vx10	+5Vx20	+7Vx20
5s	4	3	
10s		1	2

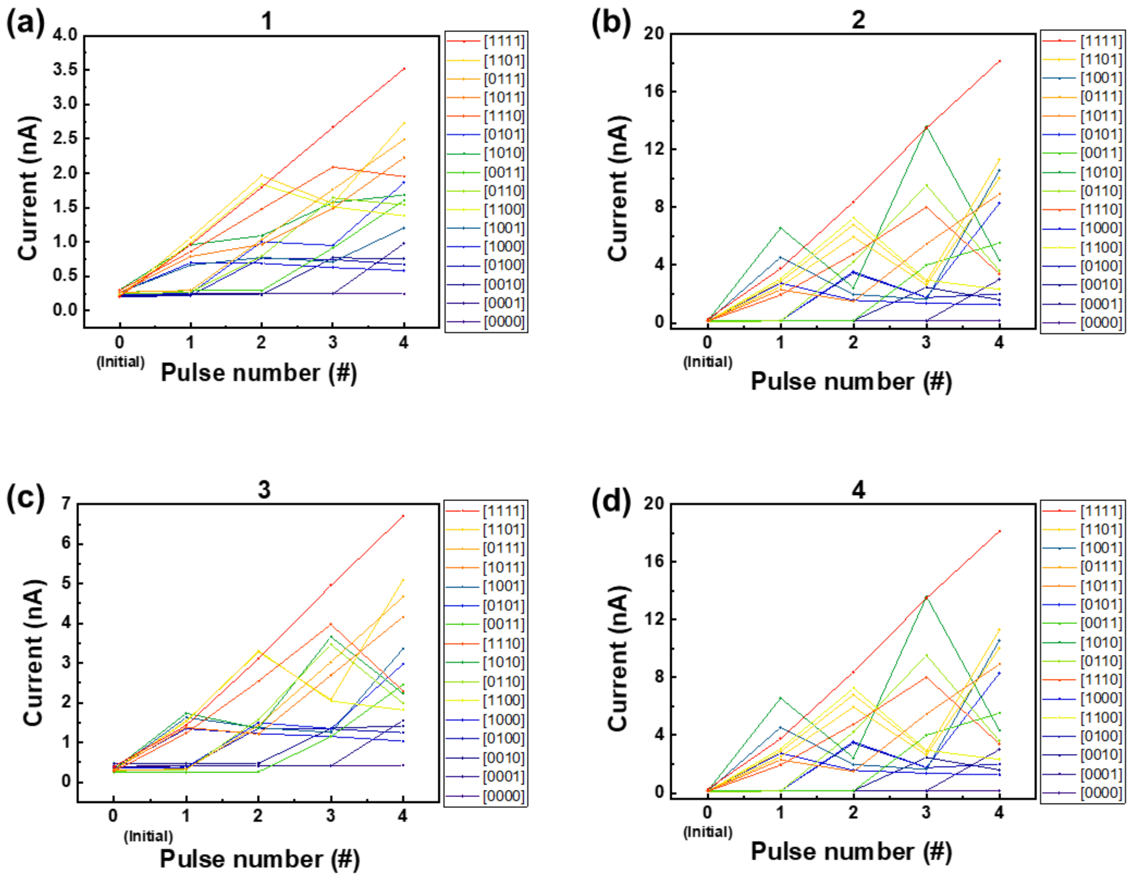
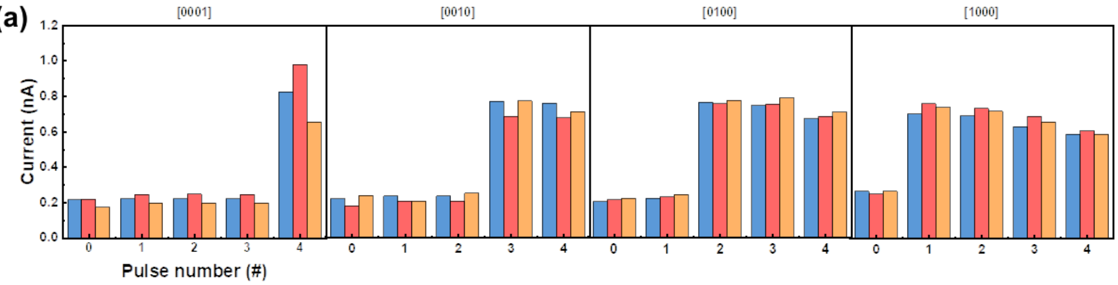


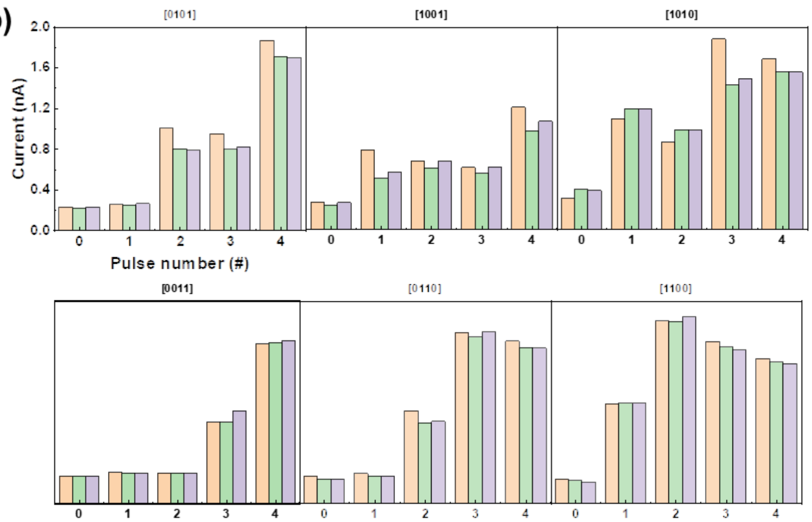
Fig. S3. Experimental results of binary 4-bit input pulse data discrimination processing at four different pulse conditions. Experimental results of binary 4-bit input data discrimination processing by applying a binary 4-bit pulse train with a total of 16 states ([0000] to [1111]) for each the condition shown in the table. The read pulse amplitude and width are +2 V and 0.64 ms, respectively. The pulse condition of (**a**) +5 V×20 read at +2V, interval 10 s (condition 1), (**b**) +7 V×20 read at +2V, interval 10 s (condition 2), (**c**) +5 V×20 read at +2V, interval 5 s (condition 3), and (**d**) +5 V×10 read at +2V, interval 5 s (condition 4).

Interval \ Pulse	+5Vx10	+5Vx20	+7Vx20
5s	4	3	
10s		1	2

(a)



(b)



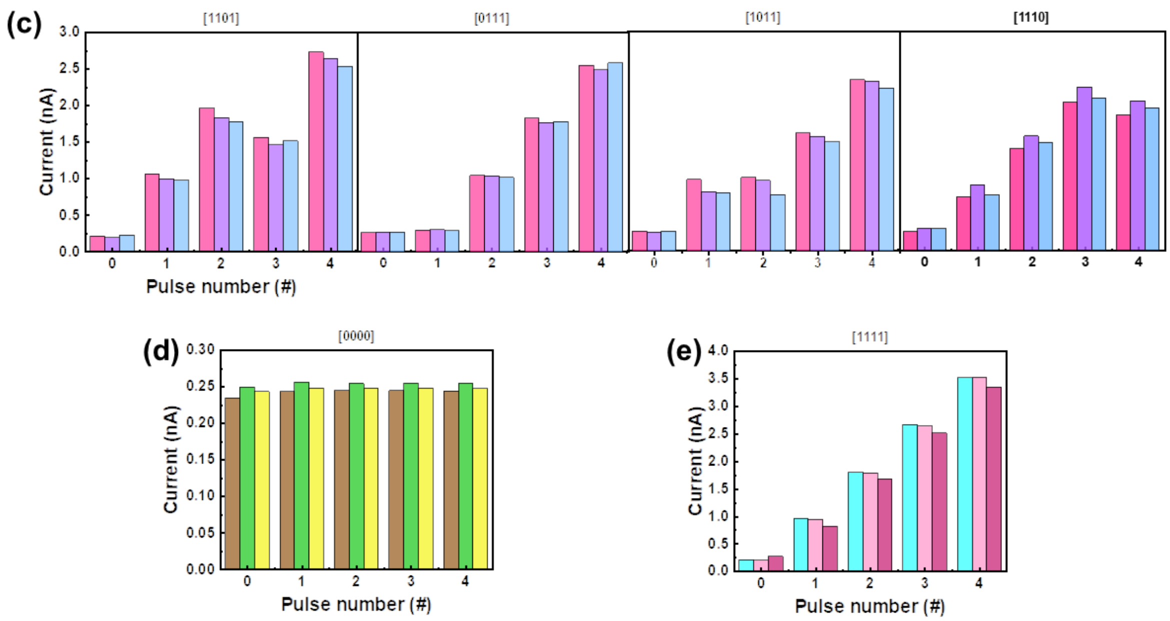
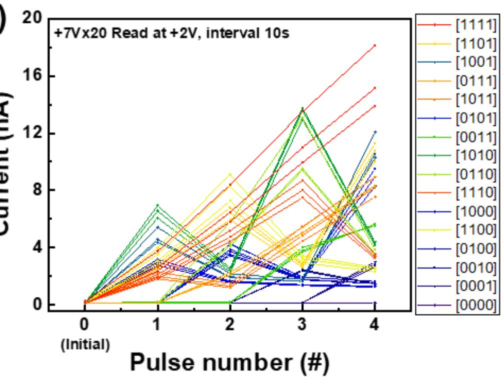


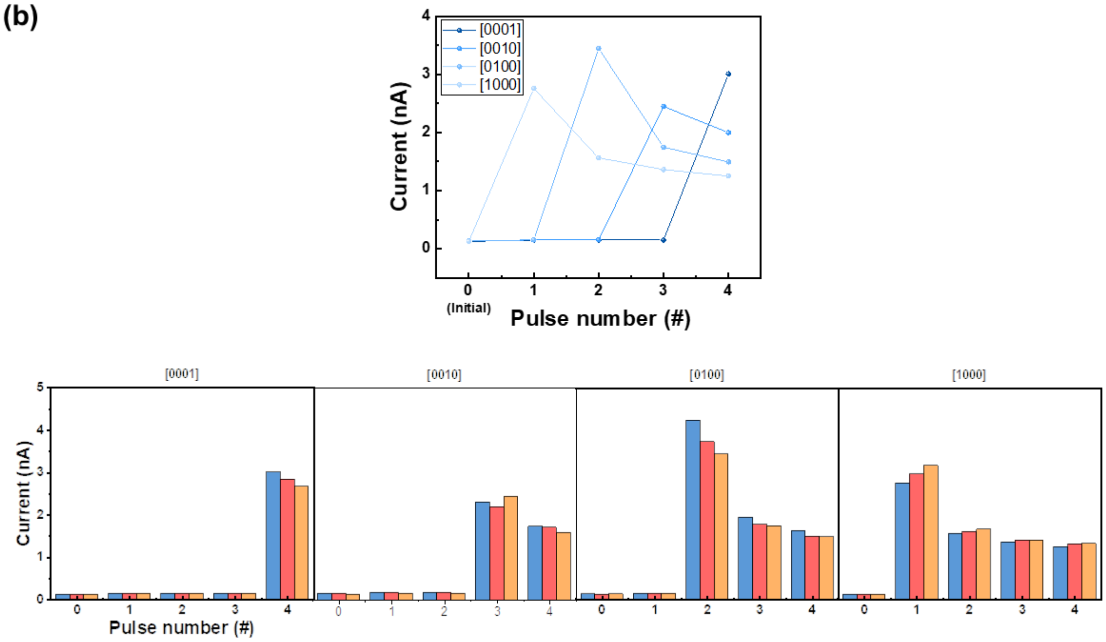
Fig. S4. In the case of pulse condition 1, response of GDC/CeO_2 memristors by repeatedly applying a pulse train to a binary 4-bit input for a total of 3 times with read pulse (+2 V, 0.64 ms) (**a**) when the pulse bit corresponding to "1" is one ([1000], [0100], [0010], [0001]), (**b**) when the pulse bit corresponding to "1" is two ([0101], [1010], [0011], [0110], [1100], [1001]), (**c**) when the pulse bit corresponding to "1" is three ([1101], [0111], [1011], [1110]), (**d**) when the pulse bit corresponding to "1" is zero ([0000]), and (**e**) when the pulse bit corresponding to "1" is four ([1111]).

Pulse / Interval	+5Vx10	+5Vx20	+7Vx20
5s	4	3	
10s		1	2

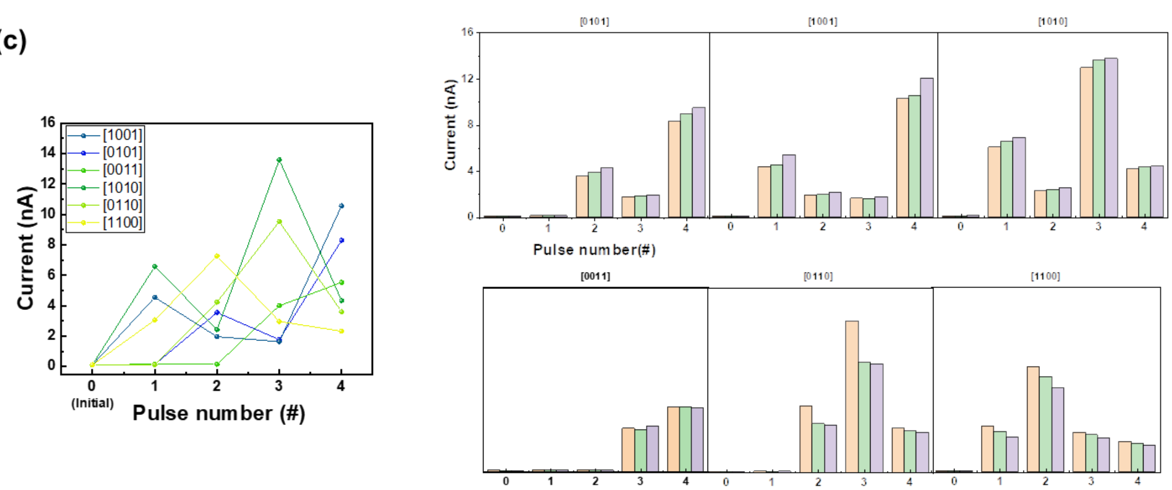
(a)



(b)



(c)



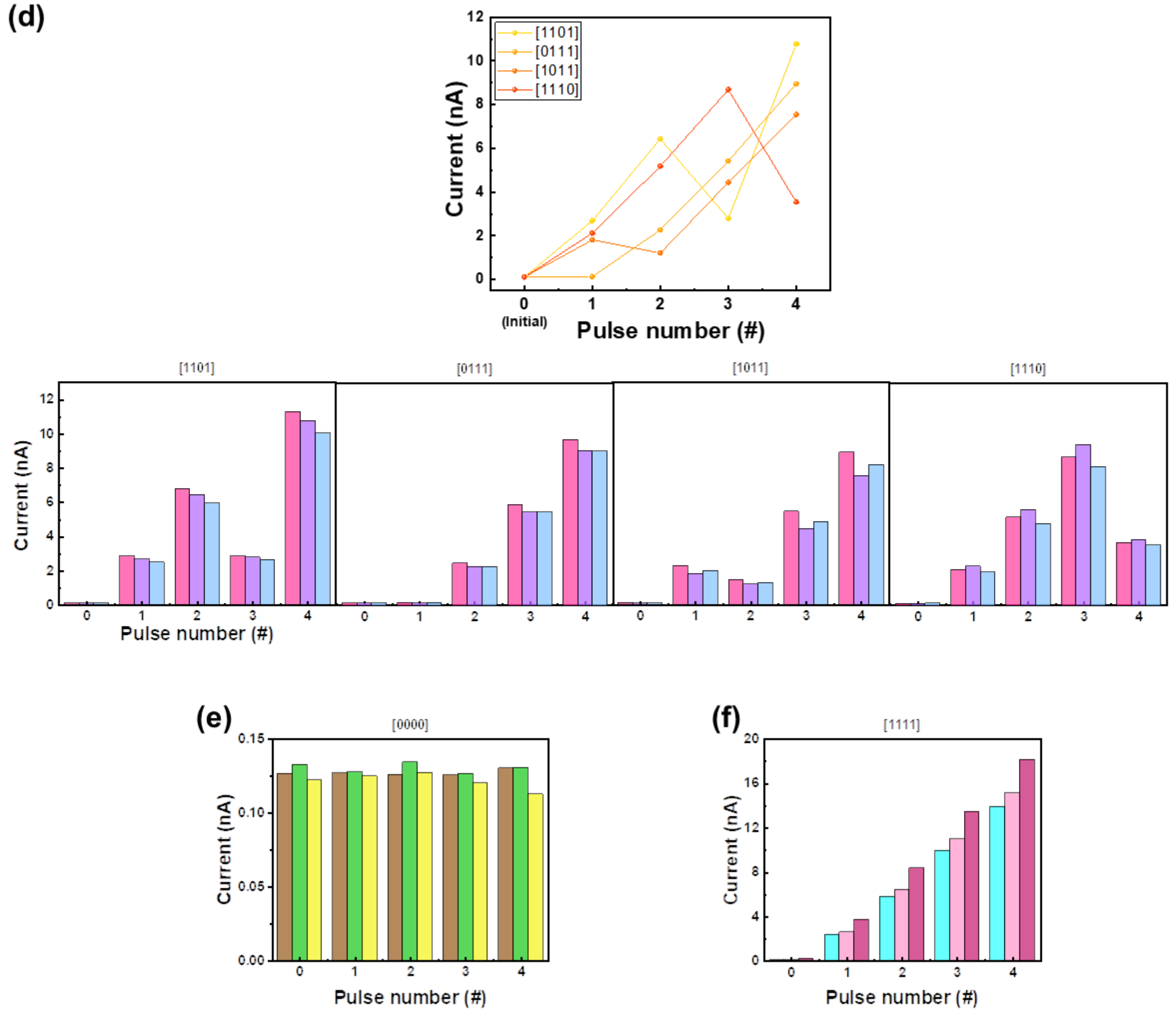
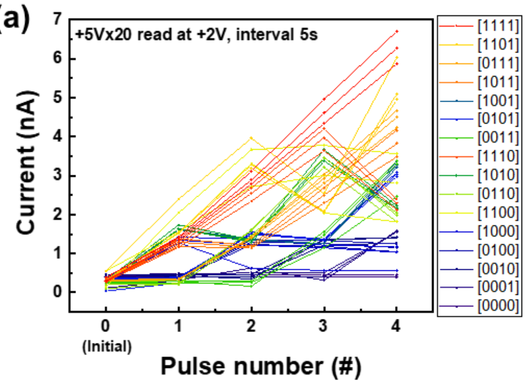


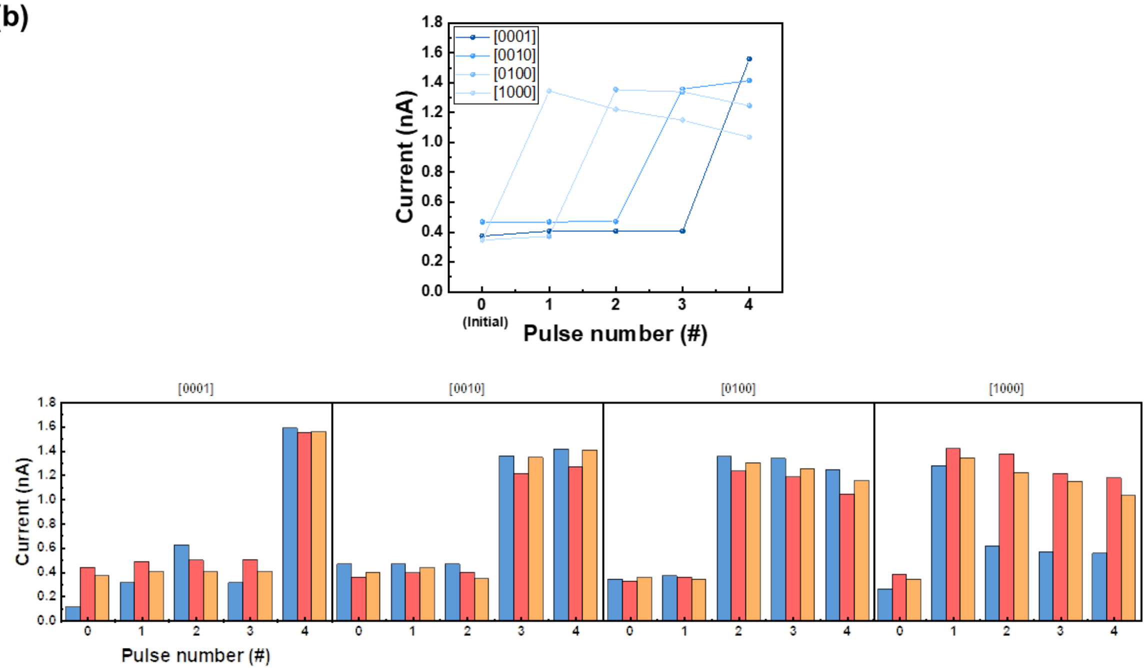
Fig. S5. In the case of pulse condition 2, response of GDC/CeO_2 memristors by repeatedly applying a pulse train to a binary 4-bit input for a total of 3 times with read pulse (+2 V, 0.64 ms) (**a**) when the pulse bit corresponding to “1” is one ([1000], [0100], [0010], [0001]), (**b**) when the pulse bit corresponding to “1” is two ([0101], [1010], [0011], [0110], [1100], [1001]), (**c**) when the pulse bit corresponding to “1” is three ([1101], [0111], [1011], [1110]), (**d**) when the pulse bit corresponding to “1” is zero ([0000]), and (**e**) when the pulse bit corresponding to “1” is four ([1111]).

Pulse / Interval	+5Vx10	+5Vx20	+7Vx20
5s	4	3	
10s		1	2

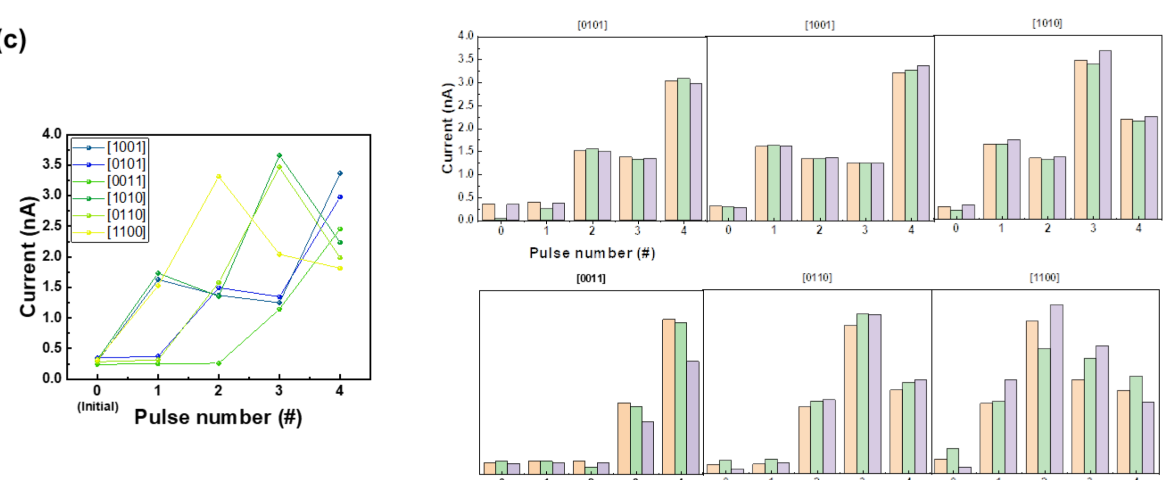
(a)



(b)



(c)



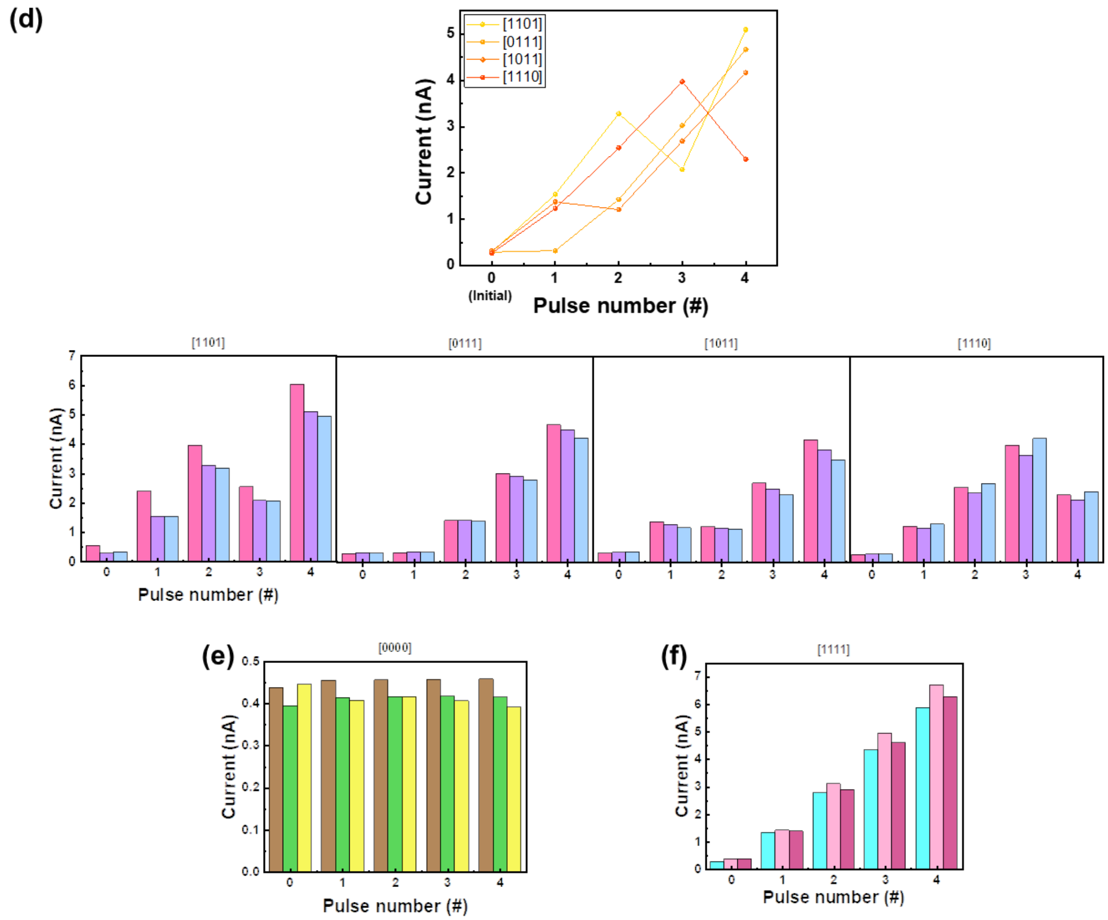


Fig. S6. In the case of pulse condition 3, response of GDC/CeO_2 memristors by repeatedly applying a pulse train to a binary 4-bit input for a total of 3 times with read pulse (+2 V, 0.64 ms) (**a**) when the pulse bit corresponding to "1" is one ([1000], [0100], [0010], [0001]), (**b**) when the pulse bit corresponding to "1" is two ([0101], [1010], [0011], [0110], [1100], [1001]), (**c**) when the pulse bit corresponding to "1" is three ([1101], [0111], [1011], [1110]), (**d**) when the pulse bit corresponding to "1" is zero ([0000]), and (**E**) when the pulse bit corresponding to "1" is four ([1111]).

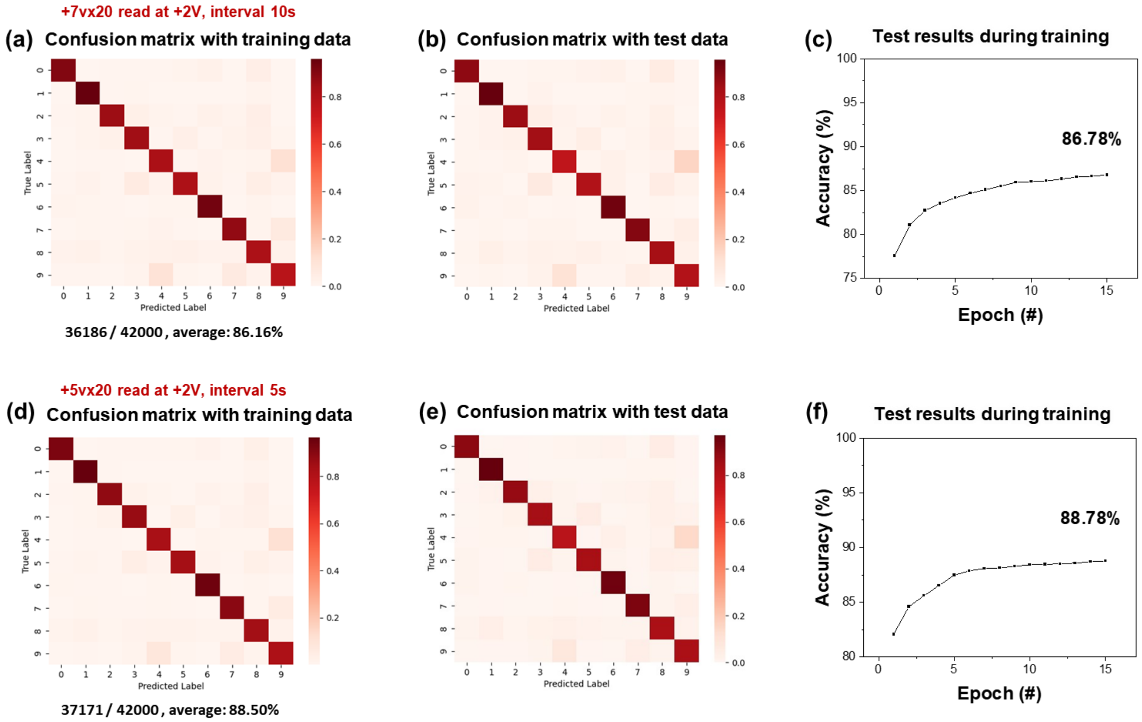


Fig. S7. Recognition of handwritten digit images using a memristor-based RC system. (**a**) Confusion matrix with training data, (**b**) confusion matrix with test data, and (**c**) the simulation results of accuracy at the 15th epoch of 4-bit classification for RC system in the pulse conditions +7 V×20 read at +2V. (**d**) Confusion matrix with training data, (**e**) confusion matrix with test data, and (**f**) the simulation results of accuracy at the 15th epoch of 4-bit classification for RC system in the pulse conditions +5 V×20 read at +2V.

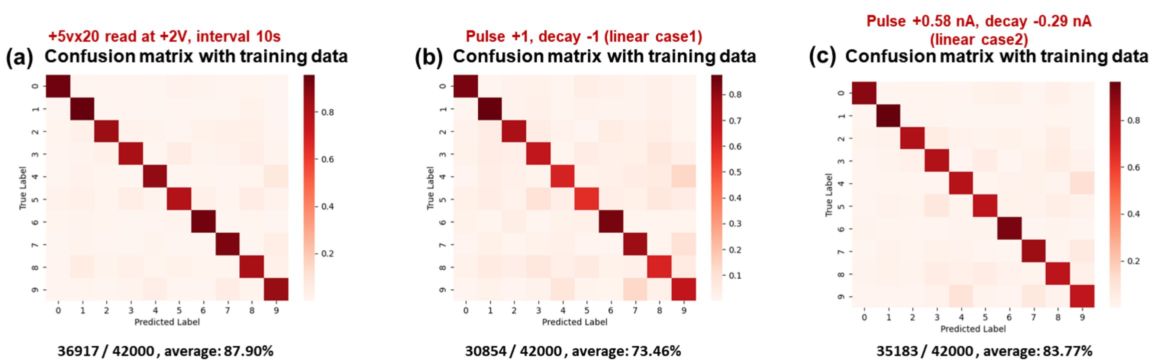


Fig. S8. Recognition of handwritten digit images using a memristor-based RC system. Confusion matrix with training data in conditions (**a**) +5 V×20 read at +2V, interval 10 s (condition 1), (**b**) linear case 1, and (**c**) linear case 2.