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Short-term plasticity and long-term potentiation mimicked in single inorganic synapses

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Memory is believed to occur in the human brain as a result of two types of synaptic plasticity: short-term plasticity (STP) and long-term potentiation (LTP; refs 1–4). In neuromorphic engineering[5,6], emulation of known neural behaviour has proven to be difficult to implement in software because of the highly complex interconnected nature of thought processes. Here we report the discovery of a Ag_2S inorganic synapse, which emulates the synaptic functions of both STP and LTP characteristics through the use of input pulse repetition time. The structure known as an atomic switch[7,8], operating at critical voltages, stores information as STP with a spontaneous decay of conductance level in response to intermittent input stimuli, whereas frequent stimulation results in a transition to LTP. The Ag_2S inorganic synapse has interesting characteristics with analogies to an individual biological synapse, and achieves dynamic memorization in a single device without the need of external preprogramming. A psychological model related to the process of memorizing and forgetting is also demonstrated using the inorganic synapses. Our Ag_2S element indicates a breakthrough in mimicking synaptic behaviour essential for the further creation of artificial neural systems that emulate characteristics of human memory.

Neuroplasticity, where changes in the strength of synaptic connections (or weights) are caused by memorization events, underlies the ability of the brain to memorize. STP is achieved through the temporal enhancement of a synaptic connection, which then quickly decays to its initial state. However, repeated stimulation causes a permanent change in the connection to achieve LTP; shorter repetition intervals enable efficient LTP formation from fewer stimuli. Although synaptic behaviour has been imitated by hardware-based neural networks such as hybrid complementary metal–oxide–semiconductor analogue circuits or other artificial neural devices[9–20], no inherent hardware-based memorizing ability has been implemented and is currently achieved through software programming.

Recently, electrically induced switching phenomena have been shown in nanoscale ionic-based devices, which have been proposed as a basis for future non-volatile memories[21–23]. In non-stoichiometric materials with ionic and electronic conductivity, ion migration is coupled to reduction and oxidation processes that result in large electrical conductance changes. Artificial synaptic devices operated by ion migration have been reported[24], and some of these devices[25,26] have shown evidence of spike-timing-dependent plasticity[27] characteristics. Spike-timing-dependent plasticity is an important memorization mechanism related to the synaptic strength of connections in biological circuits and synthetic devices that emulate the spike-timing-dependent plasticity model. They require precise control of the relative timing between the signals applied to the two electrodes, to mimic the pre- and post-synaptic potentials in biological systems.

In previous work, we reported an atomic switch[7,8], which is a two-terminal electroionics-based device, where formation and annihilation of a metallic atom bridge is controlled in a nanogap situated between two electrodes (typical current–voltage characteristics are presented in Supplementary Fig. S1). It uses a solid-state electrochemical reaction, which indicates the presence of some characteristics of memory formation related to a synaptic connection[28,29]. Namely, the switch showed two types of conductance state: one that rapidly fades away after weak signal inputs, analogous to STP, and a second long-lived stable state requiring a strong signal for erasure, conceptually analogous to LTP. Changes in conductance, including a transition between these two states, were observed on application of specific bias voltages, where conductance was determined by the history of previous input signals. As the adjustment of the timing of pre- and post-synaptic potential is not required, a new approach for emulating synaptic behaviour is available for exploration. However, a time dependence was not observed in that work, although it is an essential characteristic of neuroplasticity.

Here, we report that the decay of conductance occurred in STP without any application of bias voltage, resulting in both time-dependent STP and LTP behaviour in a single device, which we find is dependent on stimulation rate. In a Ag_2S inorganic synapse, we relate the temporal enhancement of conductance to STP, and find that it occurs before the complete formation of a metallic atomic-sized bridge, and that the decay in conductance is explainable by deformation of an incomplete bridge. Once a robust bridge is formed, the enhancement persists for a long period of time, that is, corresponding to LTP, as schematically illustrated in Fig. 1a. Moreover, we present a psychological model related to memorizing and forgetting, that was duplicated through a rehearsal process of the inorganic synapse by using the decay behaviour in measured electrical conductance.

STP and LTP appeared during inorganic synapse operation, where input pulses with an amplitude (V) of 80 mV, a width (W) of 0.5 s and repetition intervals (T) of 2 s or 20 s were applied. When these input pulses were applied with a lower repetition rate, at intervals of 20 s, the system did not maintain the higher-conductance state of approximately one quantized channel (77.5 μS) that was achieved directly after each input pulse, but

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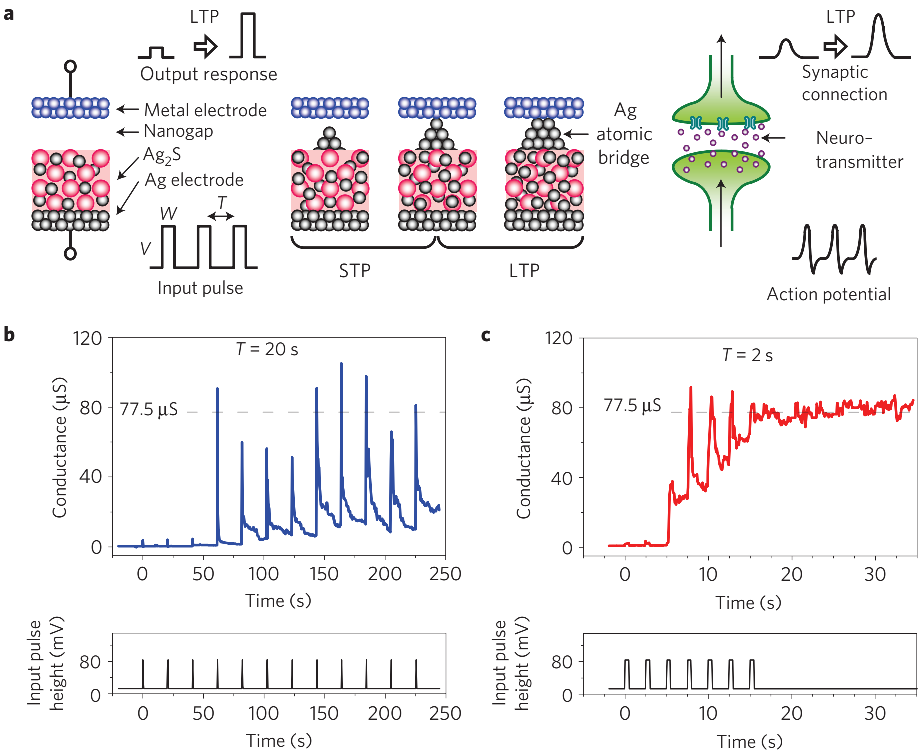

Figure 1 | Inorganic synapse showing STP and LTP, depending on input-pulse repetition time. a, Schematics of a Ag_2S inorganic synapse and the signal transmission of a biological synapse. Application of input pulses causes the precipitation of Ag atoms from the Ag_2S electrode, resulting in the formation of a Ag atomic bridge between the Ag_2S electrode and a counter metal electrode. When the precipitated Ag atoms do not form a bridge, the inorganic synapse works as STP. After an atomic bridge is formed, it works as LTP. In the case of a biological synapse, the release of neurotransmitters is caused by the arrival of action potentials generated by firing, and then a signal is transmitted as a synaptic potential. Frequent stimulation causes long-term enhancement in the strength of the synaptic connection. **b**,**c**, Change in the conductance of the inorganic synapse when the input pulses ($V = 80$ mV, $W = 0.5$ s) were applied with intervals of $T = 20$ s (**b**) and 2 s (**c**). The conductance of the inorganic synapse with a single atomic contact is $2e^2/h$ (=77.5 μS), where e is the elementary charge, and h is Planck's constant.

which decreases with time back to its initial low conductance value (Fig. 1b). This behaviour relates to the STP mechanism observed in biological synapses. The decay phenomenon appears after each input pulse, until the application of the next pulse. Importantly, this decay occurs without any application of voltage, a feature that is in contrast to other ionic devices, where an applied signal is required to cause a change in conductance[23–26]. The inorganic synapse also showed a long-lived transition to the higher-conductance state when the repetition rate of the stimulation pulses was increased, that is, when a shorter interval time ($T = 2$ s) between inputs was used, as shown in Fig. 1c. Here, a permanent transition to higher-conduction states is clearly observed with repeated application of input pulses, successfully mimicking the LTP mechanism of a biological synapse. The higher conductance range observed is over 77.5 μS, corresponding to a single atomic contact[30], supporting a synaptic behaviour where STP is achieved by incomplete bridge formation whereas LTP is achieved by a complete atomic bridge formation. This resulting output response stability is also observed to be enhanced by application of input pulses, resembling a persistent increase in synaptic connection following higher-repetition stimulation by action potentials found in the biological nervous system[2], as shown schematically in Fig. 1a.

We investigated the stability of LTP as a function of input-pulse repetition rate, where we defined 'LTP' as a state maintaining a conductance higher than 77.5 μS after a time exceeding 20 s from the last electrical stimulus (see Supplementary Section S2). We also define 'unstable potentiation' as a temporary increase in conductance over 77.5 μS that is metastable and rapidly decays to a value smaller than 77.5 μS after an input pulse. After application of a total of 12 input pulses ($V = 80$ mV, $W = 0.5$ s) with two different pulse intervals, it was observed that 97% and 78% of the recorded events corresponded to unstable potentiation with both shorter ($T = 2$ s) and longer ($T = 20$ s) intervals, respectively (Fig. 2a). The 2-s-interval pulses, however, caused 93% of the inorganic synapses to move to an LTP state whereas the 20-s-interval pulses gave only 19% of events resulting in LTP. The relatively high value of 19% of events resulting in LTP with longer interval was found to statistically relate to particular inorganic synapses that suddenly increased rather than gradually built up conductance as a function of input pulses. Each inorganic synapse operating in the STP state also showed a certain distribution of the threshold number of input pulses required for unstable potentiation (see Supplementary Fig. S5).

In Fig. 2b, we show that the conductance of an inorganic synapse after the 12th input pulse is smaller than 77.5 μS, and its subsequent decrease in conductance is characteristic of STP. Conversely, it was observed that its relatively easy to maintain an enhanced conductance higher than 77.5 μS, corresponding to the formation of a multi-atom metallic bridge. We found that the decay time of the STP state also maintains a history of the previous input pulses. This is shown in Fig. 2c, where the exponential decay time of STP is observed to increase with the number of input pulses. This history dependence indicates that a non-stoichiometry of Ag_2S, supporting the Ag atomic bridge, rearranges[31] with subsequent pulses. The lack of Ag^+ ions in the Ag_2S region, adjacent to the Ag cluster, can then recover with

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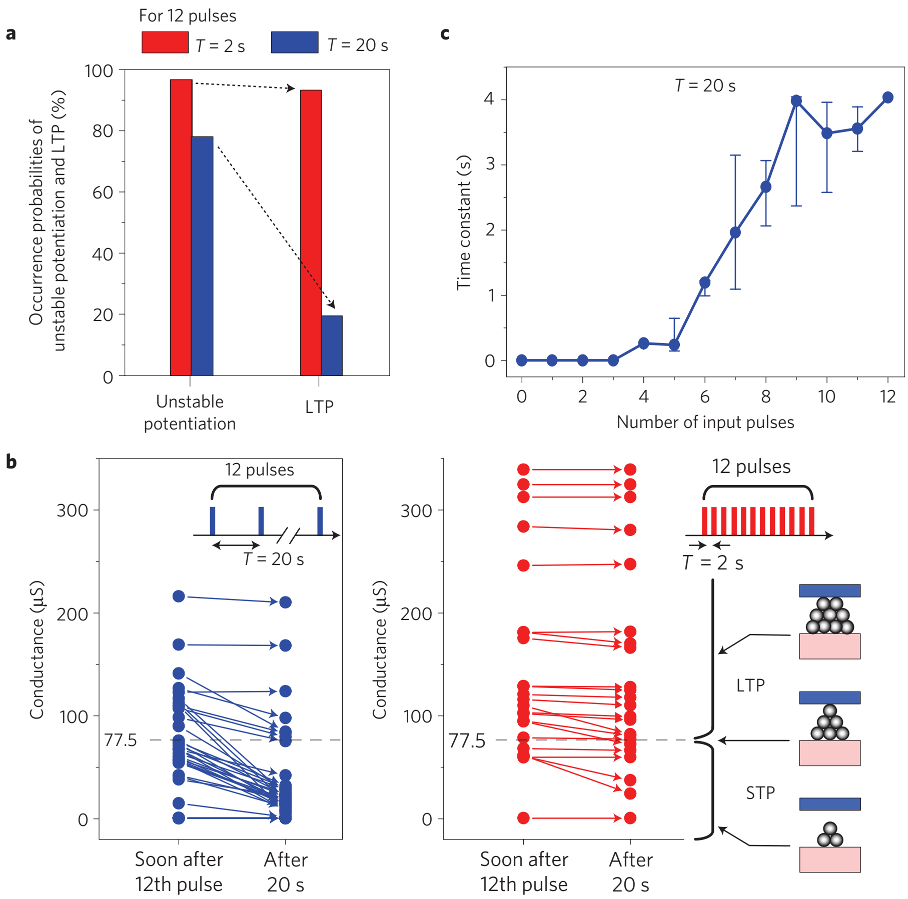

Figure 2 | LTP formation depending on input-pulse repetition time. a, The occurrence probabilities of inorganic synapses in which an 'unstable potentiation' state occurred during stimulation pulses and that formed an 'LTP' state at the end of the stimulation events. Input pulses ($V = 80$ mV, $W = 0.5$ s) were applied 12 times at intervals of $T = 2$ s or 20 s. **b**, The change in conductance after application of 12 input pulses with pulse intervals of $T = 20$ and 2 s. The dashed line shows a conductance of 77.5 μS. Each circle represents an individual inorganic synapse. **c**, Typical change in the time constant of an inorganic synapse for the decay in the STP state.

an increase in the number of input pulses. The measured time constant is of the order of seconds, indicating that the phenomenon is not purely electronic, but rather electroionic, because electrical capacitance effects have an estimated timescale of nanoseconds (see also Supplementary Section S4).

The inorganic synapse was observed to go beyond purely mimicking synaptic-like biological behaviour, and may also be useful in psychology for implementing a model of human memory in the brain. It is believed that human memory is created by the dynamic change of neural circuits based on the synaptic connections, and it is accepted that some architecture for human memory exists in the brain, although its mechanism has not yet been fully elucidated. In 1968, Atkinson and Shiffrin proposed 'the multistore model' of human memory[32], which is still the most accepted model in psychology. In this model, new information from the external environment is stored for a very short period of time in the sensory register as a sensory memory (SM), and then selected information is transferred from temporary short-term memory (STM) in the short-term store to a permanent long-term memory (LTM) in the long-term store, as illustrated in Fig. 3a. Importantly, Atkinson and Shiffrin assumed that STM can become LTM through a process of rehearsal, and that the probability of transfer to LTM increases with rehearsal repetition. It should be noted that STP and LTP are terms used in neuroscience whereas STM and LTM are terms used to describe psychological phenomena. In Fig. 3b, we present a memorization model inspired by the multistore model. The memorization level in SM mode increases slightly with initial rehearsals, and then, in STM mode, is temporarily enhanced before decaying. Subsequent frequent rehearsals are observed to result in LTM, leading to a long-term memorization. We propose that the conductance value of the inorganic synapse is analogous to the memorization level in this model of human memory, as shown in Fig. 1b,c, where the behaviour of the inorganic synapse corresponds to the simplified memorization model.

The decay phenomenon of conductance in STM mode also suggests a surprising similarity to 'the forgetting curve'. Since Ebbinghaus developed the first approach to forgetting in 1885 (ref. 33), forgetting (or retention) curves have been derived[34,35], and it is clear that the repetition rehearsal based on active recall is an appropriate method for increasing memory strength. Figure 3c shows the experimental decay curve of conductance of the inorganic synapse in STM mode. The decay rate, which corresponds to the rate of the power function (m), generally used in psychology, decreased and the ratio of memory retention increased with the increase in the number of rehearsals. This result correlates well to the forgetting curve in psychology and is well reproduced by the inorganic synapse.

To demonstrate concrete psychological behaviour, the memorization of two images into a 7×7 inorganic synapse array was carried out, as shown in Fig. 4 and Supplementary Movie. An image of the numeral '2' was stored using ten inputs with longer intervals ($T = 20$ s), and an image of the numeral '1' was simultaneously stored using the same number of inputs, with equal

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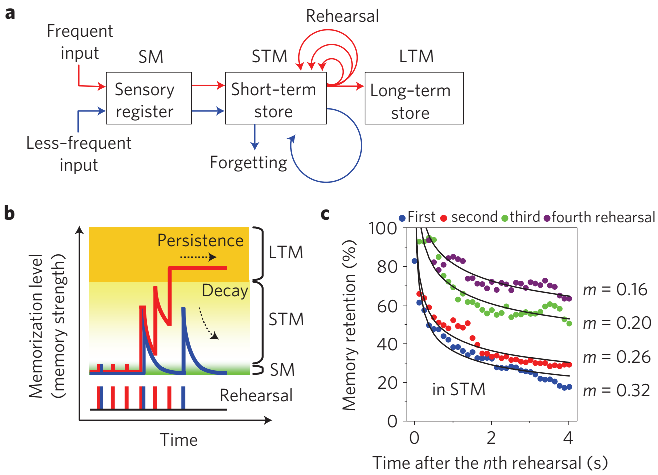

Figure 3 | The multistore model and the human-memory forgetting curve. **a**, The psychological model of human memory proposed by Atkinson and Shiffrin. This multistore model provides for three types of memory, that is, sensory memory (SM), short-term memory (STM) and long-term memory (LTM). **b**, Simplified memorization model in the inorganic synapse, which was inspired by the multistore model. After storing new information as SM, information is stored in STM for short periods of time, whereas repeated rehearsal events result in LTM. At a higher repetition rate, rehearsal before complete decay in memorization level forms LTM, as shown by the red line. Rehearsal at lower repetition rate cannot form LTM, as shown by the blue line. **c**, Typical change of memory retention in the inorganic synapse for the decay in STM mode. A power function, used to analyse psychological behaviour such as STM (ref. 35), $y = b \times t^{-m}$, was used to fit the conductance curves, where y is the memory retention, b is the fit constant for scaling, t is the time from the nth rehearsal and m is the power function rate. Memory retention was normalized using a conductance value of 77.5 μS. The conditions of the input pulse were $V = 80$ mV, $W = 0.5$ s and $T = 4$ s.

amplitude and width, but with shorter intervals ($T = 2$ s). At first, the numeral '2' (and '1') emerged slightly on the first few inputs, corresponding to SM mode. Following this, the numerals '1' and '2' were temporarily stored in the inorganic synapse array while repeating the concurrent enhancement of conductance and spontaneous decay in conductance up to the last (tenth) input. This process corresponds to repeated rehearsal in STM mode. The numerals '1' and '2' appeared with higher conductance soon after the last input, which made it difficult to distinguish them from each other. However, the numeral '1' was observed to persist after 20 s from the last input, owing to the forgetting of the numeral '2', demonstrating that only the numeral '1' was transferred to LTM mode. As the total numbers of inputs of images were the same for memorizing both images, the numerals '1' and '2' should have been stored at the same conductance level in a conventional switch array. However, our result indicates that a multistore model best described the observed behaviour. This experimental demonstration clearly shows implementation of a multistore model, which is a unique function of the inorganic synapse, and that it has three types of memory (SM, STM and LTM), including forgetting. The data indicate that we may apply a psychological memory model simultaneously with the emulation of biological synaptic-like behaviour.

Here, we demonstrate that synaptic behaviours of an Ag_2S inorganic system resemble some of the key features of a biological synapse, with clear experimental evidence of STP and LTP characteristics in a single device. A temporary increase in conductance and its spontaneous decay over time was observed using input stimuli at a lower repetition rate, and persistent enhancement was easily achieved by frequent input repetition. The results reported show that individual inorganic synapse elements may enable a new functional element suitable for the design of neural systems that can work without the need of the poorly scalable software and pre-programming currently employed in artificial neural network systems, with clear potential for hardware suited to artificially and physically intelligent systems. In addition, the wiring problem[6], caused by the requirement for a much larger number of elements to imitate the human brain, may be solved by using inorganic synapses with previously proposed but experimentally unconfirmed architectures for hardware-implemented artificial neural networks[36,37].

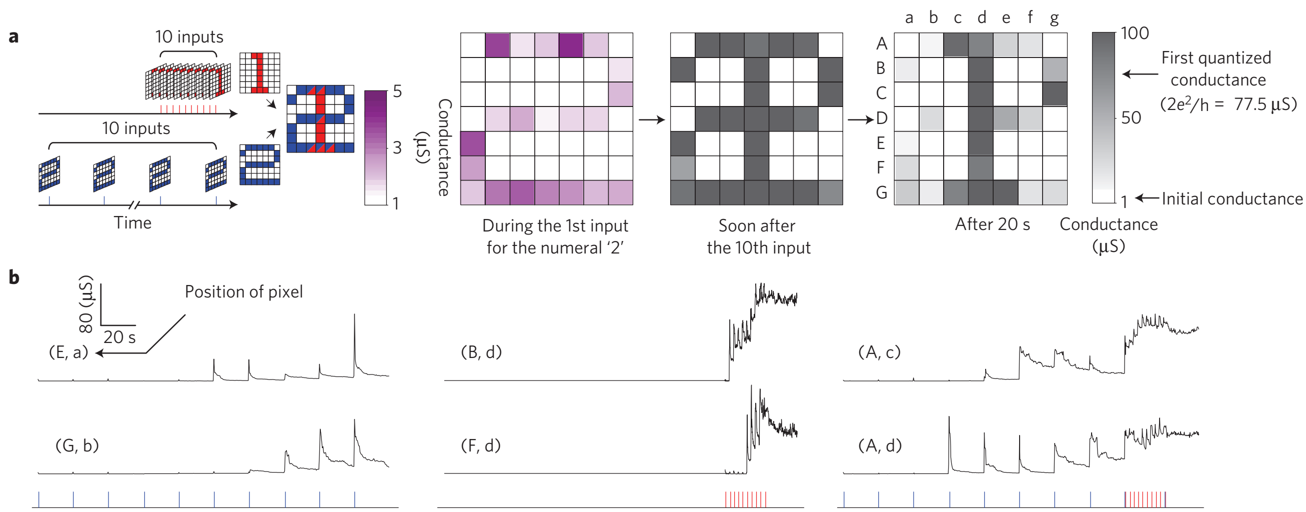

Figure 4 | Image memorizing into an inorganic synapse array. **a**, Images of the numerals '1' and '2' were memorized simultaneously into a 7 × 7 inorganic synapse array by inputting the image ten times with intervals of $T = 2$ and 20 s, respectively. The image of the numeral '2' was stored into STM mode and that of the numeral '1' into LTM mode, which caused the emergence of the numeral '1' after 20 s from the last input. The numeral '2' also emerged slightly by the first few inputs, which corresponds to SM mode (in this figure, the response by the first input was shown, and its scale bar is also different from the other two). The change in conductance at each pixel corresponds to the result for a single inorganic synapse, and the respective conductance profiles in the pixels are shown in **b**. **b**, Typical change in the conductance of an individual inorganic synapse, depending on input-pulse repetition time. The x axis is time and the two upper plots correspond to conductance (see the scale bar on the left). The bottom y axis shows the input pulse sequences.

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Methods

The inorganic synapse devices consisted of a nanoscale Ag_2S-coated Ag electrode and a counter platinum electrode. Ag_2S, which is a mixed ionic and electronic conductor[38,39], was made by sulphurizing a Ag substrate in sulphur vapour at 150 °C for 70 min. A scanning tunnelling microscope, working at room temperature, was used to make the nanometre gap between the two electrodes. The initial conductance of each inorganic synapse was set to 1 μS, which corresponds to the initial conductance state. After formation of the nanogap, application of the voltage input pulse to the inorganic synapse was carried out. The conductance of the inorganic synapse was measured under an applied voltage of 10 mV using a series-connected reference resistance of 10 kΩ.

The demonstration of the memorizing of two images into a 7 × 7 Ag_2S inorganic synapse array was also carried out using a device structure with a nanogap formed by a scanning tunnelling microscope. The change in conductance at each pixel was obtained by the voltage pulse input–output measurement of each point on the Ag_2S/Ag substrate. The row/column of the array was addressed by the moving of a counter platinum electrode (platinum tip) 49 times with an interval of 100 μm.

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Author contributions

T.O. and T.H. designed the experiments. T.O., T.H. and J.K.G. wrote the paper. T.O. also carried out the experiments and analysed the data. T.T. and K.T. contributed to the materials and analysis. All authors discussed the results and commented on the manuscript. T.H. and M.A. directed the projects.

Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper on www.nature.com/naturematerials. Reprints and permissions information is available online at http://www.nature.com/reprints. Correspondence and requests for materials should be addressed to T.O. or T.H.

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