Supporting Information for

A facile solution processable self-rectifying and sub-1 V operating memristor via oxygen vacancy gradient within TiO₂ single layer

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Fig. S1 Schematic structure of the a-TiO₂ based memristor. (a) Dot type device. (b) Crossbar type device.



Fig. S2 (a) HR-TEM image of a-TiO₂ on the ITO substrate with 100 nm scale bar. (b-f) EDS mapping images of various elements (O, Ti, C, In, Si) in the a-TiO₂ film on the ITO substrate.



Fig. S3 XPS depth profile for Ti 2p (a) in g-TiO₂ film and (b) in s-TiO₂ film as the functions of Ar^+ sputtering time.



Fig. S4 XPS spectra of O 1s of (a,d,g) a-TiO₂,(b,e,h) g-TiO₂ and (c,f,i) s-TiO₂ films under different etching conditions.



Fig. S5 Summarized ratio of deconvolution area of a-TiO₂, g-TiO₂ and s-TiO₂ films XPS O 1s spectra.



Fig. S6 *I-V* curves of memristors across 5 cycles, where positive voltage (0 V \rightarrow 0.5 V \rightarrow 0V)

were applied.



Fig. S7 *I-V* curves of a-TiO₂ based dot type memristors using various concentrations of solution.



Fig. S8 UPS spectra of ITO, a-TiO₂, g-TiO₂ and s-TiO₂. (b) Secondary electron cut-off region.(c) Valance region.



Fig. S9 Schematic energy band diagram of Ag/TiO₂/ITO memristors with aligned Fermi level.



Fig. S10 $\log(I)$ - $\log(V)$ curves of the a-TiO₂, s-TiO₂ and g-TiO₂ to investigate the conduction mechanism.



Fig. S11 *I-V* characteristics of a-TiO₂ based dot type memristor with PEDOT:PSS top electrode.



Fig. S12 *I-V* characteristics of a-TiO₂ based memristor in the 3×3 crossbar array.



Fig. S13 (a) *I-V* curves, and (b) rectification ratio of dot type and crossbar type devices.



Fig. S14 The data retention of HRS and LRS recorded for an a-TiO₂ based memristor in the 3 \times 3 crossbar array, with error bars.

	Structure	\mathbf{V}_{set}	Rectification	Rectification ratio	On/off ratio	Size	Crossbar	Туре	Ref
1	Pt/Ti/HfO _x /TaO _x /TiN	4 V	No		> 10	$4{\sim}100~\mu m^2$		Interface	67
2	Pt/NbO _x /TiN	$1.5 \sim 2 \text{ V}$	No		$> 10^{2}$	$0.16{\sim}16~\mu m^2$		Filament	68
3	Ag/CTS-MoO ₃ /Mo	$1 \sim 2 V$	Restrictive	$> 4 \times 10^{3}$	$> 4 \times 10^{3}$	NA		Filament	69
4	Pt/HfO _x /SiO _x /TiN	3 V	No	NA	NA	NA		Filament	70
5	Au/MAPb ₃ /Au	2 V	No		$> 10^{8}$	$2500 \ \mu m^2$	8 × 8	Filament	71
6	Au/HfSe ₂ /Ti	0.74 V	No		$> 10^{2}$	$25 \ \mu m^2$	3 × 3	Filament	72
7	Pt/TiO _x /HfO ₂ /Pt	-5 V	Yes	> 10 ³	NA	4 nm^2	3 × 3	Interface	73
8	Au/TiO ₂ /Au	3 V	Restrictive	NA	$> 10^{3}$	$25{\sim}100~\mu m^2$	32 × 32	Interface	74
9	Ag/TiO ₂ /FTO	5 V	No		> 10	12.55 mm^2		Interface	75
10	Ag/TiO ₂ /FTO	2 V	No		> 30	NA		Filament	76
This work	Ag/TiO ₂ /TiO _x /ITO	0.9 V	Yes	$> 10^{4}$	$> 4 \times 10^3$	$0.1 \sim 1 \text{ mm}^2$	3 × 3	Interface	

Table. S1 Comparative analysis of a-TiO₂ memristor performance relative to previous studies.