Supporting Information
for

Flexible Random Resistive Access Memory with FeC-rGO Nanocomposites for Artificial Synapses

Jiankui Zhou,[a] Hanfang Feng,[a] Yadong Wang,[a] Qingqing Sun,[a] Yingliang Liu,[a] Xuying Liu*[a], Li Zhang*[a] and Shaokui Cao*[ab]

[a] School of Materials Science and Engineering, Zhengzhou University, Zhengzhou 450001, People's Republic of China

[b] Henan Key Laboratory of Advanced Nylon Materials and Application, Zhengzhou University, Zhengzhou 450001, People's Republic of China.

Characterization of FeC-rGO nanocomposite.

Fig. S1 shows the FTIR spectra of FeC, GO, rGO and FeC-rGO. Different from GO, the absorption peak of FeC-rGO disappears at 1723 cm^{-1}, suggesting the C=O in the FeC-rGO has been reduced.[S1, S2] There appear obvious absorption peaks at 1630 cm^{-1}, 1389 cm^{-1} and 1050 cm^{-1}, corresponding to the C=C stretching vibration of rGO, C-H bending vibration of ferrocene, and C-Fe stretching vibration, respectively.[S3] Moreover, C-H stretching vibration of FeC-rGO is red-shift relative to that of FeC, which is an evidence of the stronger interaction between FeC and rGO.[S4, S5]

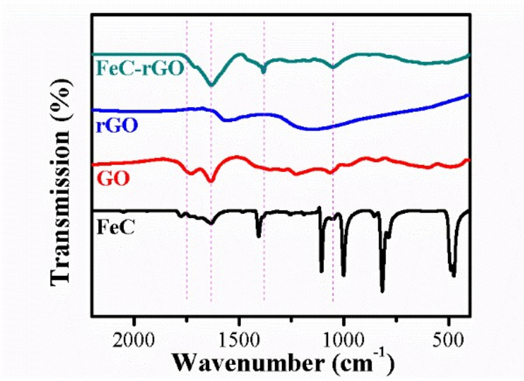

Fig. S1 FTIR spectra of FeC, GO, rGO and FeC-rGO.

Compared to GO, the π→π* absorption peak of rGO-FeC is red-shift to 269 nm, and the n→π* absorption peak (λ=304 nm) disappears, suggesting that the rGO is fully reduced by ascorbic acid.[S6] Additionally, there appears a wider shoulder peak at 300~600 nm for rGO-FeC, which is caused by the stronger electronic interaction between FeC and rGO.[S7] As shown in **Fig. S2**, the rGO-FeC can well disperse in ethanol. Unlike GO, the ethanol dispersion solution of FeC-rGO is black, suggesting that the graphene in the complex has been reduced.

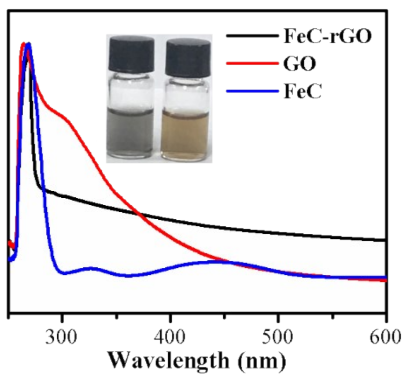

Fig. S2 UV-Vis spectra of FeC, GO and FeC-rGO.

Supplementary Figures

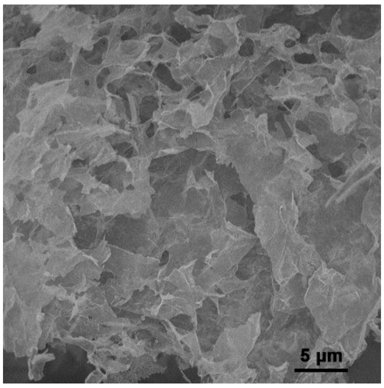

Fig. S3 SEM images of rGO.

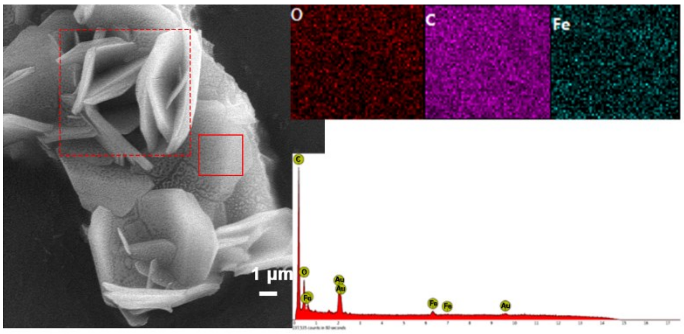

Fig. S4 SEM images of FeC-rGO and corresponding O, C, Fe mapping (rectangle area marked with red solid line) and EDS; The rectangle area using red dotted line shows the stacked lamellar structure of FeC-rGO.

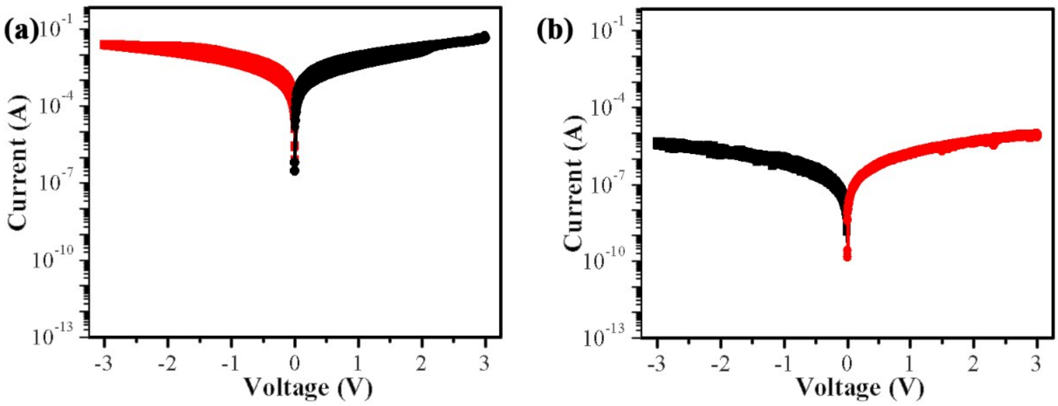

Fig. S5. Representative I-V curves of (a) Al/rGO/ITO and (b) Al/FeC/ITO devices.

Table S1 Comparison of memory performance of FeC-related memristor devices

Material	Device Structure	ON/OFF	V_{SET}/V_{RESET}	Retenion	WRER	Ref.
Ferrocenylphenyl-NHCO-GO	Al/FPArGO/ITO, 50nm	~10^3	2.1 / -1.9	1000s	1000	S8
PFT2-Fc	Al/PFT2-Fc/LiF/ITO, 50 to 60 nm	~10^3	-1.9 / 1.4	7h	100	S9
Ferrocene-containing metallopolymers*	Au/Polymer/ITO/Glass, 120 nm and 80 nm	<10^2	1.89, 1.54	-	-	S10
DAFcTPA/6FDA PI (PI-xFc)	Au/PI-xFc/ITO, ~50 nm	10^3	1.4 / -3.2	4000s	10^8	S11
Stb-1G-FcD	Al/dendrimer/LiF/ITO, 50nm	~10^3	-2.8 / 2.8			S12
Py-Fc	Al/Py-Fc/ITO, 50nm	>10^3	-1.6 / ~3	12000s	10^3	S13
Ferrocene-terminated hyperbranched polyimide (HBPI-Fc)*	Al/HBPI-Fc/ITO, 50nm	10^4	2.2 / -2.6	10^4s	-	S14
Ferrocene-terminated hyperbranched polyimide (HBPI-Fc)*	Al/HBPI-Fc/ITO, 50nm	10^4	2.5 to 3.2 / -3.3 to -2.7	10^4s	-	S15
Ferrocene-g-6FDA/DHTM PI	Au||ferrocene-g-6FDA/DHTM PI||ITO, ~50nm	10^3	~1.5 / -3.0	300s	-	S16
Non-conjugated ferrocene-containing copolymers FcCP	Al/FcCP/ITO, ~50nm	10^3 to 10^4	-0.5 to -2.5 / 3.2 to 3.7	10^4s	10^4	S17
Conjugated ferrocene-containing poly(fluorenylethynylene)s (PFcFE1–PFcFE4)	Al/PFcFE/ITO, ~50nm	10^3 to 10^4	~0.5 / ~3	3000 s	10^5	S18
PFTPA-Fc (redox active moieties of triphenylamine (TPA) and ferrocene (Fc) onto the pendants of fluorene skeletons) #	Pt/PFTPA-Fc/ITO, ~130 nm	-	-	-	-	S19
This work	Al/FeC-rGO/ITO, ~100 nm	10^8	-1.7/+2.1	10^5	500	

*Write Once Read Many times memory (WORM)

Memristors

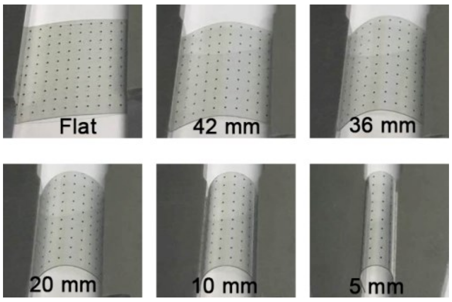

Fig. S6. Photographs of flexible Al/FeC-rGO/ITO/PET devices of flat and different bending radius.

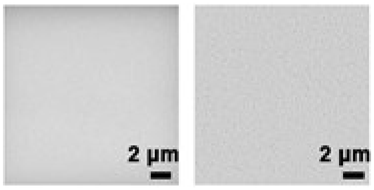

Fig. S7. SEM images of the surfaces of the flexible device before (Left) and after (Right) bending

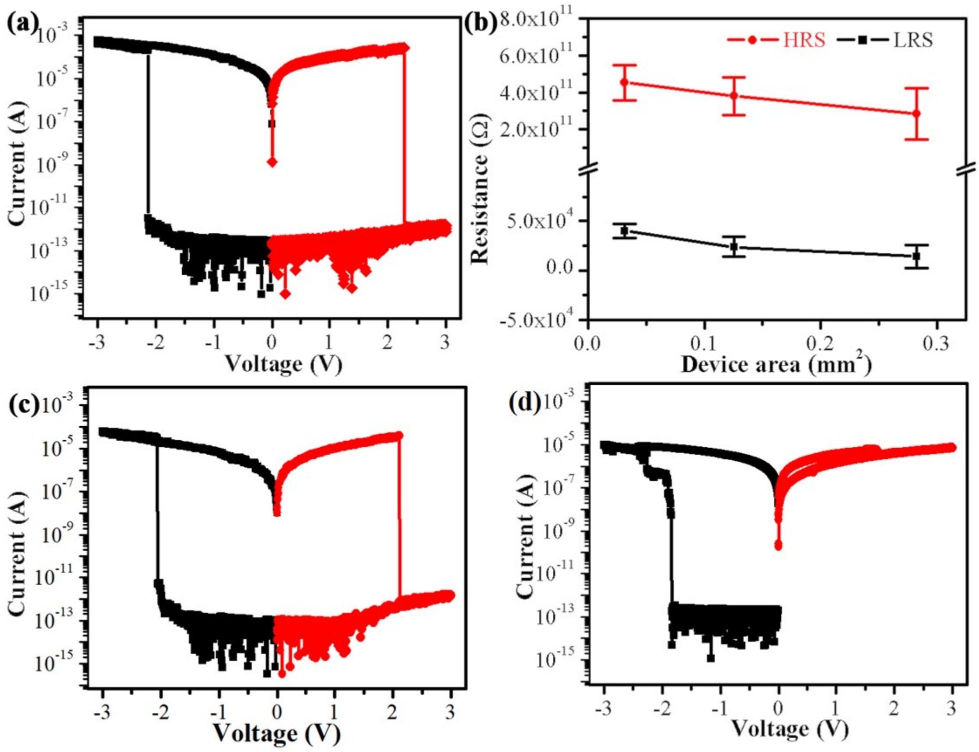

Fig. S8. (a) I-V curve of Au/FeC-rGO/ITO device; (b) LRS/HRS resistance of Al/FeC-rGO/ITO with different areas; (c) I-V curve of Al/FeC-rGO/Al device; (d) I-V curve of Al/GO-FeC/ITO device.

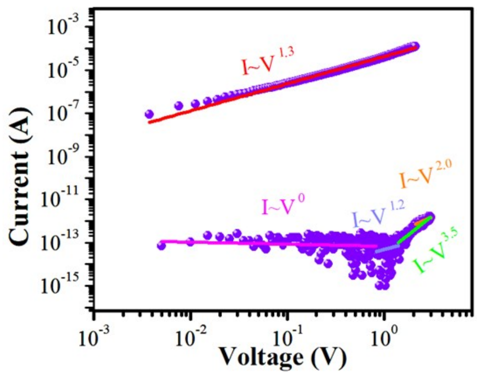

Fig. S9. log(I)–log(V) plot of RESET process

Table S2 coefficient and R_square of SET and RESET processes.

Processes		1	2	3	4	5
SET	Interval (V)	-0.005 to -0.175	-0.175 to -1.025	-1.025 to -1.440	-1.440 to -1.700	-3.000 to -0.005
	coefficient	0	1.2	2.8	24.6	1.3
	R_square	0.0227	0.1376	0.3357	0.8257	0.9804
RESET	Interval (V)	0.005 to 2.110	2.110 to 3.000	3.000 to 1.395	1.395 to 0.820	0.820 to 0.005
	coefficient	1.3	2.0	3.5	1.2	0
	R_square	0.9915	0.8474	0.9236	0.6628	0.1316

Reference

S1 M. Kudus, M. Zakari, H. Akil, F. Ullah and F. Javed, *J. King Saud University*, 2020, **32**, 910.

S2 Z. Hou, Y. Zhou, S. Wang, M. Wang, X. Hu, L. Zhou and G. Li, *Adv. Mater. Res.*, 2013, **807**, 2805.

S3 J. Enlow, H. Jiang, J. T. Grant, K. Eyink, W. Su and T. J. Bunning, *Polymer*, 2008, **49**, 4042.

S4 M. J. Deka and D. Chowdhury, *J. Phys. Chem. C*, 2016, **24**, 218.

S5 K. Deng, J. Zhou and X. Li, *Electrochim. Acta*, 2013, **95**, 18.

S6 A. D. Adhikari, R. Oraon, S. K. Tiwari, N. K. Jena, J. H. Lee, N. H. Kim and G. C. Nayak, *Chem.-Asian J.*, 2017, **12**, 900.

S7 C. Su, L. Ji, L. Xu, X. Zhu, H. He, Y. Lv, M. Ouyang and C. Zhang, *RSC Adv.*, 2015, **5**, 14053.

S8 C. Jin, J. Lee, E. Lee, E. Hwang and H. Lee, *Chem. Commun.*, 2012, **48**, 4235.

S9 T. Choi, K. Lee, W. Joo, S. Lee, T. Lee and M. Chae, *J. Am. Chem. Soc.*, 2007, **129**, 9842.

S10 X. Cheng, A. Md, H. Lian, Z. Zhong, H. Guo, Q. Dong and V. Roy, *J. Organomet. Chem.*, 2019, **892**, 34.

S11 R. Hao, N. Jia, G. Tian, S. Qi, L. Shi, X. Wang and D. Wu, *Mater. Design*, 2017, **139**, 298.

S12 C. Kim, W. Joo, E. S. Song, H. J. Kim, J. Kim, C. Park, H. L. Lee and C. Kiml, *Synthetic Met*., 2007, **157**, 640.

S13 Y. Li, X. Zhu, Y. Li, M. Zhang, C. Ma, H. Li, J. Lu and Q. Zhang, *ACS appl. Mater. interfaces*, 2019, **11**, 40332.

S14 H. Tan, H. Yao, Y. Song, S. Zhu, H. Yu and S. Guan, *Dyes Pigments*, 2017, **146**, 210.

S15 H. Tan, H. Yu, Y. Song, S. Zhu, B. Zhang, H. Yao and S. Guan, *J. Polym. Sci. Pol. Chem.*, 2018, **56**, 505.

S16 G. Tian, S. Qi, F. Chen, L. Shi, W. Hu and D. Wu, *Appl. Phys. Lett.*, 2011, **98**, 203302.

S17 J. Xiang, X. Li, Y. Ma, Q. Zhao, C. Ho and W. Wong, *J. Mater. Chem. C*, 2018, **6**, 11348.

S18 J. Xiang, T. Wang, Q. Zhao, W. Huang, C. Ho and W. Wong, *J. Mater. Chem. C*, 2015, **4**, 921.

S19 B. Zhang, F. Fan, W. Xue, G. Liu, Y. Fu, X. Zhuang, X. H. Xu, J. Gu, R. W. Li and Y. Chen, *Nat. Commun.*, 2019, **10**, 736.