Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

Two-Stage Filamentary Mechanism in High-Performance Organic Resistive Switch

Arti Bisht[1,2], Nitish Saini[1,2], Komal Bhardwaj[1,2], Rachana Kumar[1,2,3], Ajeet Kumar [1,2*]

[1]CSIR-National Physical Laboratory, Dr. K. S. Krishnan Marg, New Delhi 110012, India
[2]Academy of Scientific and Innovative Research (AcSIR), Ghaziabad-201002, India
[3]CSIR-Indian Institute of Toxicology Research, Lucknow 226001, India
*Corresponding Author Email: *kumarajeet@nplindia.org*

Table S1. A summary of resistive switching characteristics of organic-ReRAM devices and governing switching mechanisms.

Device	Retention	Endurance	On/OFF	Voltage	Memory	Mechanis	Devi	Ref

structure	[s]	[Cycle]		range [V]	behaviour	m	ce measured	.
Al/4C_ZIPN-P_3HT/ITO	10^4	150	10^5	-6/3.3	Non-Volatile	Trapping and de-trapping of charges	-	[1]
Ag/Pectin/FTO	10^8	27	10^4	3/-2.8	Non-Volatile	Filamentary switching	-	[2]
Al/D–A–A/ITO	10^4	60	10^4	-3.5/1.8	Non-Volatile	Trap-based hole transfer	-	[3]
Al/F8BT/ITO	- [a)]	-	10	-5/+5	Non-Volatile	The transition of trap-limited SCLC to trap-filled SCLC	-	[4]
Al/Carbazole/ITO	10^3	-	10^3	-1.5/+4	Non-Volatile	Formation and dissociation of a charge-transfer state under the applied voltage	-	[5]
Mg/Pectin/Mg	10^6	160	20	Current bias	Nonvolatile	Filamentary switching	-	[6]
Au/PEDOT:PSS/Ag clusters/PMMA/ITO	10^4	50	10^2	-10/+10	Nonvolatile	Filamentary switching	10	[7]
Ag/2DImine/ITO	10^3	200	10^3	-3/+1	Flash	Filamentary switching	-	[8]
Au/MQDs-PVP/ITO	10^3	-	10^2	−3.8/−2.2	Flash and WORM	Trapping and De-trapping of charge carriers	20	[9]
Al/PVK/ITO	10^4	15	10^3	-2.7/+1.4	Flash	Filamentary switching	-	[10]
Cu/Par	10^4	10^3	10^3	-6/+1	Flash	Copper	8	[11]

ylene/ITO						filament formation		
Al/ PDA coated yarn	10^6	-	10^3	-1.5/+1.5	Nonvolatile	carbon-based electric filament	25	[12]
Ag/PVCi/Au	5000	300	-	-1.8/+1.8	Nonvolatile	Conductive filament	10	[13]
Al/PDI/ITO	**10^5**	**>250**	**10^3**	**-1.5/+2**	**Non-Volatile**	**Al and oxygen vacancy filaments**	**10**	**This work**

a) No data in this publication.

+

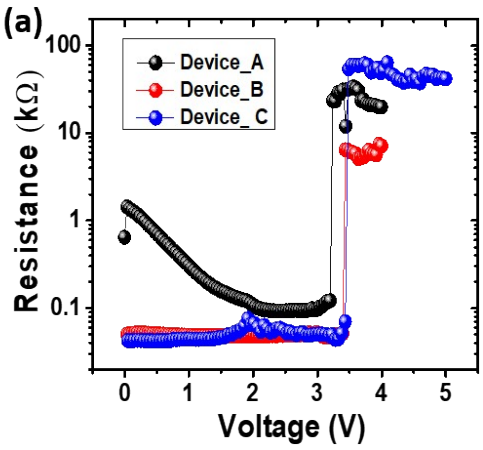


Figure S1. Reproducible electroformation of three devices.

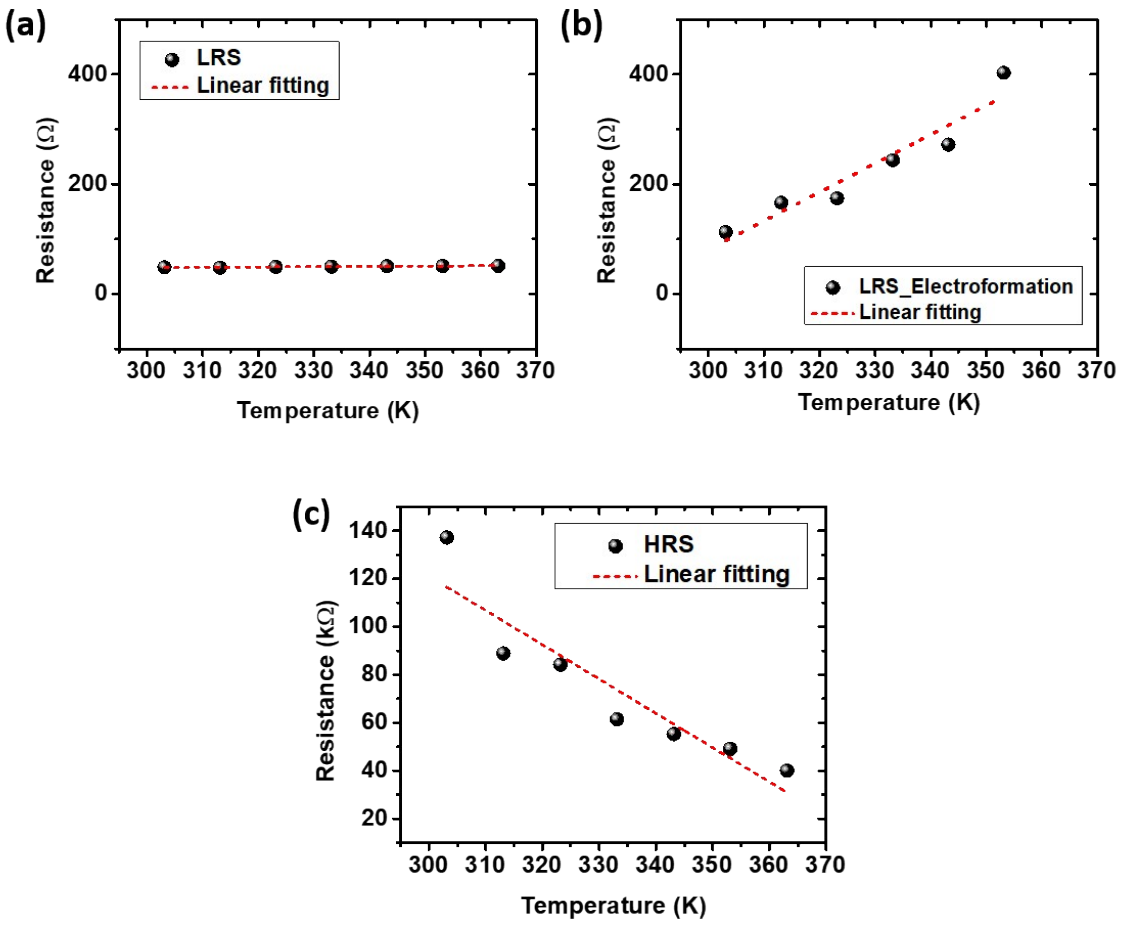


Figure S2. Temperature dependence of the resistance states of the device: (a) LRS, (b) LRS just after electroformation, (c) HRS.

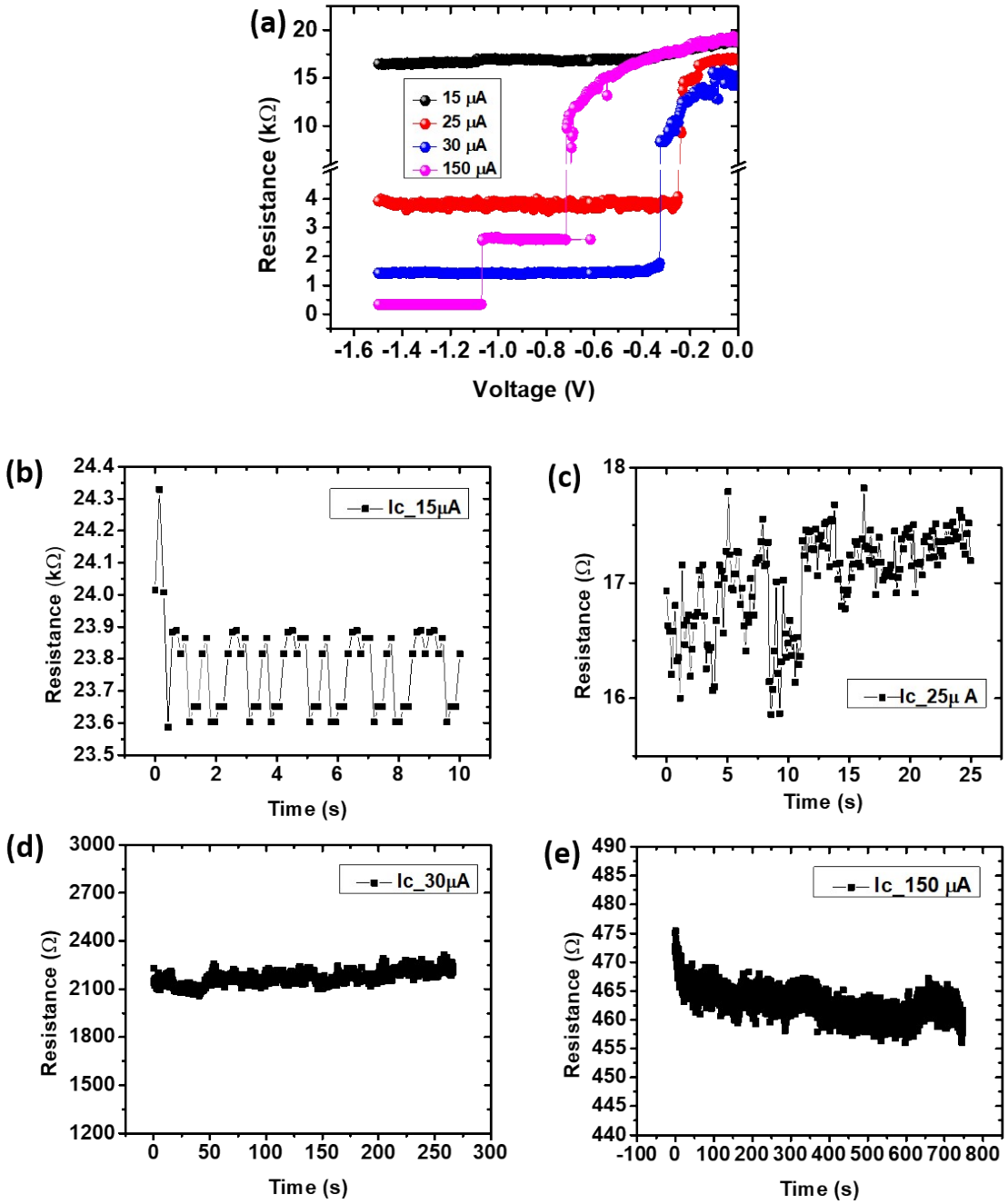


Figure S3. (a) Compliance current (I_{CC}) dependent switching and stability of the filament of the Al/PVK/ITO device. state of the resistance after switching at (b) 15μA I_{CC}, (c) 25μA I_{CC}, (d) 30μA I_{CC}, (e) 150μA I_{CC},

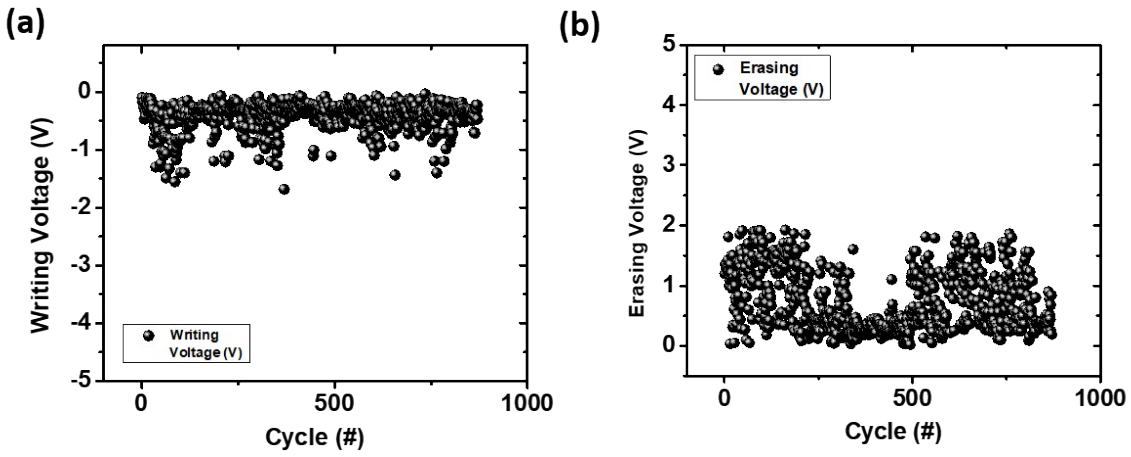
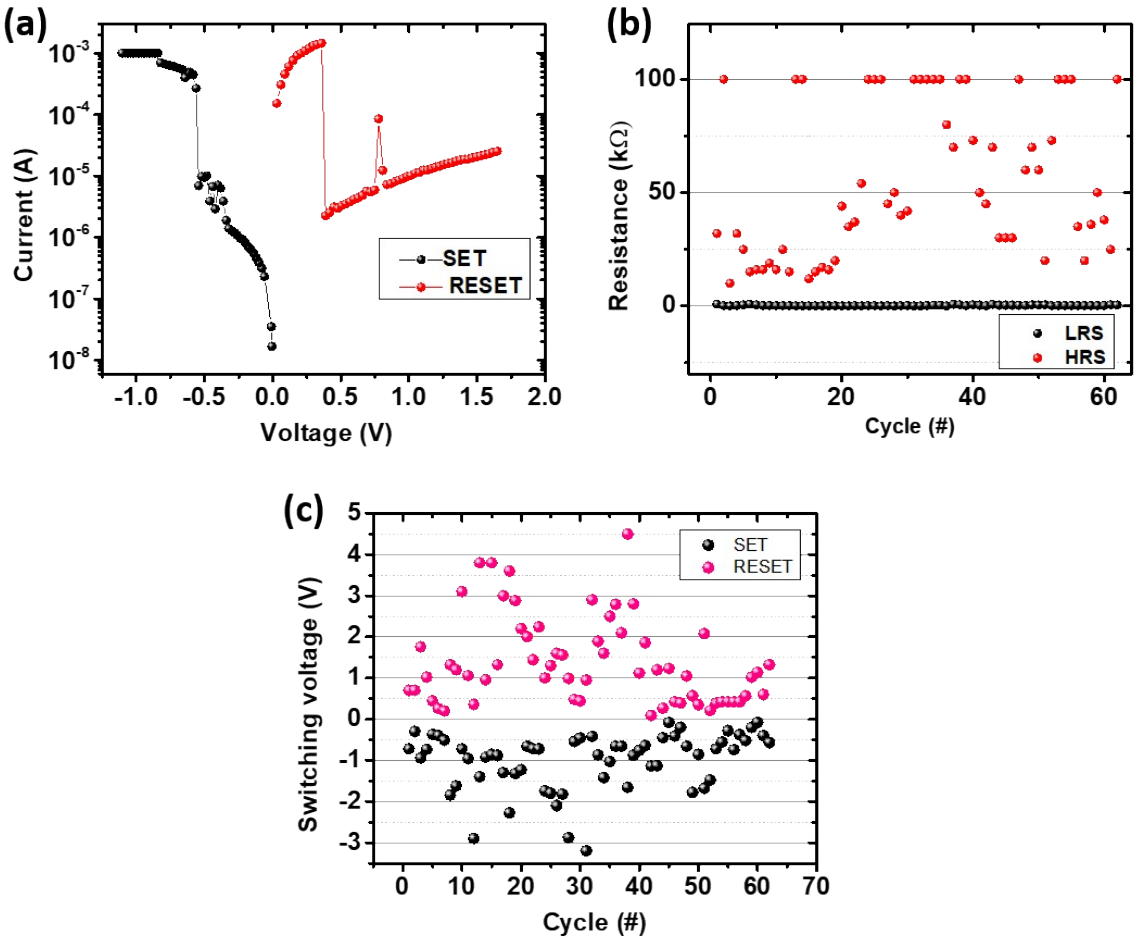


Figure S4. (a), (b) switching voltage distribution for cumulative ~880 cycles measured from



10 different devices.

Figure S5. (a) I-V characteristic of the voltages measured after ~2years (b) Endurance of the device after ~2 years. (c) Switching voltage distribution of the device.

References:

1 and B. W. Wei Li, Huiwen Zhu, Tong Sun, Wenshan Qu, Xiaxia Fan, Zhixiang Gao*, Wei Shi*, *J. Phys. Chem. C*, 2022, **126**, 12897–12905.

2 N. Arshad, M. S. Irshad, M. S. Abbasi, S. Ur Rehman, I. Ahmed, M. Q. Javed, S. Ahmad, M. Sharaf and M. D. Al Firdausi, *RSC Advances*, 2021, **11**, 4327–4338.

3 S. Pan, Z. Zhu, H. Yu, W. Lan, B. Wei and K. Guo, *Journal of Materials Chemistry C*, 2021, **9**, 5643–5651.

4 M. N. Awais, M. Mustafa, M. N. Shehzad, U. Farooq, M. T. Hamayun and K. H. Choi, *Micro and Nano Letters*, 2016, **11**, 712–714.

5 W. Lin, H. Sun, S. Liu, H. Yang, S. Ye, W. Xu, Q. Zhao, X. Liu and W. Huang, *Macromolecular Chemistry and Physics*, 2012, **213**, 2472–2478.

6 and Y. L. Xiaoning Zhao, Jiaqi Xu, Dan Xie, Zhongqiang Wang, Haiyang Xu,* Ya Lin, Junli Hu, 2021, 2104023.

7 H. L. Park, M. H. Kim and S. H. Lee, *Advanced Electronic Materials*, 2020, **6**, 1–8.

8 J. Liu, F. Yang, L. Cao, B. Li, K. Yuan, S. Lei and W. Hu, 2019, **1902264**, 1–7.

9 H. Mao, C. Gu, S. Yan, Q. Xin, S. Cheng, P. Tan and X. Wang, 2019, **1900493**, 1–8.

10 H. F. Ling, M. D. Yi, M. Nagai, L. H. Xie, L. Y. Wang, B. Hu and W. Huang, *Advanced Materials*, 2017, **29**, 1–9.

11 A. A. Minnekhanov, B. S. Shvetsov, M. M. Martyshov, K. E. Nikiruy, E. V Kukueva, M. Yu, P. A. Forsh, V. V Rylkov, V. V Erokhin, V. A. Demin and A. V Emelyanov, *Organic Electronics*, 2019, **74**, 89–95.

12 S.-B. J. Hagyoul Bae, Daewon Kim, Myungsoo Seo, Ik Kyeong Jin, K. Y. Hye Moon Lee, Soo-Ho Jung, Byung Chul Jang, Gyeongho Son and and Y.-K. C. Sung-Yool Choi, 2019, 1900151.

13 H. L. Park, M. H. Kim, M. H. Kim and S. H. Lee, *Nanoscale*, 2020, **12**, 22502–22510.