Molecular Devices **Hot Paper**

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Alcohol-Mediated Resistance-Switching Behavior in Metal–Organic Framework-Based Electronic Devices

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Abstract: *Metal-organic frameworks (MOFs) have drawn increasing attentions as promising candidates for functional devices. Herein, we present MOF films in constructing memory devices with alcohol mediated resistance switching property, where the resistance state is controlled by applying alcohol vapors to achieve multilevel information storage. The ordered packing mode and the hydrogen bonding system of the guest molecules adsorbed in MOF crystals are shown to be the reason for the alcohol mediated electrical switching. This chemically mediated memory device can be a candidate in achieving environment-responsive devices and exhibits potential applications in wearable information storage systems.*

Metal–organic frameworks (MOFs), a kind of porous crystalline materials constructed by metal centers and organic linkers, have been widely utilized as significant platforms for catalysis,[1] gas storage, and separation, as well as sensing of chemical vapors.[2] Recently, increasing attention has been paid to applications in the electronic industry, owing to the various assemblies by functional components and diverse intermolecular interactions between guest molecules and the host framework.[3] Although the insulating nature would always limit their utilities in constructing electronic devices, some achievements have already been made to improve the conductivity of MOFs by loading conductive guest molecules and ions,[4] which inspired a chemical means to control the electrical properties in the device. However, there are still challenges for MOFs to work as functional units in common electronics, such as the mismatch between the crystallinity of MOFs and device manufacturing techniques.[5] Herein, we report that a kind of MOF thin film with dense nanostructure can be easily involved in device fabrication, and present reliable resistance-switching behavior, which is widely used in realizing artificial memories. Meanwhile, by adsorbing small molecules through host–guest interactions, the resistance-switching performance can be chemically modulated, exhibiting the potential to overcome the gap between chemical information and electrical properties.[6]

Resistance-switching memory has gained increasing attention as a promising candidate for next-generation memory devices owing to its non-volatility, stable performance, high density, and easy fabrication process.[7] The fundamental characteristic of such a memory device is repeatable resistance switching in an active material, triggered and modulated by different physical operating parameters, such as electrical stimuli,[8] magnetic field,[9] temperature,[10] or light.[11] However, learning and memory, which are crucial aspects of the information storage systems in human brain, are chemically mediated processes that are usually triggered and regulated by chemical molecules and ions.[12] To extend the functionality and adaptability of resistance-switching devices, memories with chemical responsiveness would be promising and attractive when constructing smart devices. Thus, owing to the adsorption property of MOFs, we hypothesized that MOFs could not only work as active material to perform resistance switching, but also endow the memory behavior with chemical responsiveness to guest molecules, such as gases and organic vapors. Therefore, the adsorbed molecules in MOFs would provide a valuable platform to connect the intermolecular interaction and electrical performance, and realize the chemically mediated properties in electrical memory devices.

As a proof of concept, we present a promising design for an alcohol-mediated multilevel resistance-switching device based on MOFs (Figure 1a). A zeolitic imidazolate framework-8 (ZIF-8) crystal (Figure 1b), that has permanent porosity and shows high chemical and thermal stability among MOF materials,[2a,13] was selected to fabricate the memory device. A uniform ZIF-8 thin film was sandwiched between the electrodes to form the Ag/ZIF-8/Si device, and alcohol vapors were used to demonstrate the chemically mediated memory effect. Notably, the device can also exhibit a multilevel resistance-switching property.[14] Moreover, by fabricating the device on a soft substrate, we can realize a flexible as well as chemically responsive memory device. This strategy is proved to be a promising and effective design in the construction of wearable information storage devices with environment-responsive nature, which would be a prospective candidate for next-generation devices[5b,15] and synapses in neuromorphic computation systems.[16]

First, we fabricated ZIF-8 thin films[17] to work as the insulator layer in a typical sandwiched memory device

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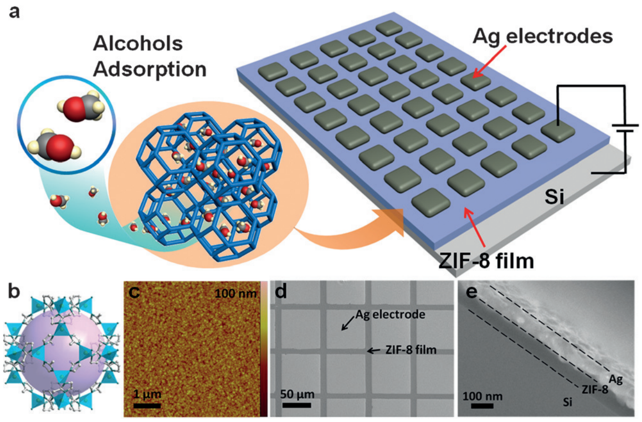

Figure 1. Strategy and morphology of the ZIF-8-based memory devices. a) ZIF-8-based memory device arrays with alcohol-mediated properties. b) Crystal structure of ZIF-8. c) AFM image of ZIF-8 film on silicon wafer. SEM images of the top (d) and cross view (e) of the fabricated device arrays. The thickness of one layer ZIF-8 film is around 60 nm.

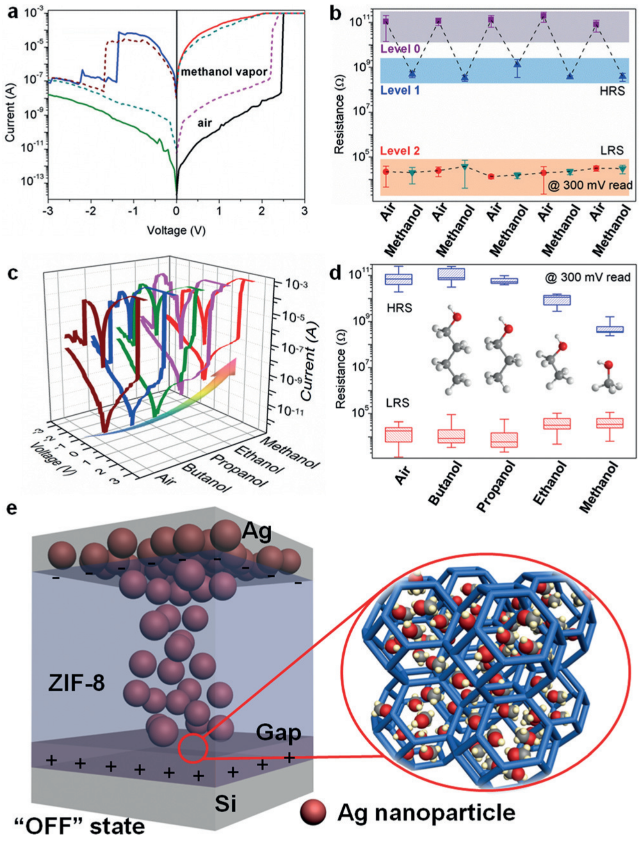

Figure 2. Methanol-mediated resistance-switching property of ZIF-8 based memory devices. a) Comparison of I-V curves of the Ag/ZIF-8/Si device in voltage sweeping mode in saturated methanol vapor (dashed line) and air (rigid line) at room temperature. The compliance current is set to 1×10^{-3} A. b) Repeated cycles of the resistance-switching of the devices working in air and saturated methanol vapor under a 300 mV read voltage. c) Typical I-V curves of ZIF-8-based memory devices in air and different saturated alcohol vapors. d) Statistical analysis of the two resistance states in different saturated alcohol vapors under a 300 mV readout voltage at room temperature. e) Schematic diagram of the ZIF-8 based device in the OFF state. While applying negative voltage on the top electrode, a positive charged gap would form at bottom interface,[18c,d] and the methanol molecules adsorbed in ZIF-8 crystals would play an essential role in tuning the HRS and OFF/ON ratio.

structure (Figure 1 d,e; Supporting Information, Figure S1). Under ambient conditions, I-V curves of the Ag/ZIF-8/Si device showed typical resistance-switching characteristics with the non-volatile nature (Figure 2 a, Figure S2 a), when a voltage bias was applied to the top Ag electrode while the silicon electrode was grounded. Device-to-device distributions of the high resistance state (HRS) and low resistance state (LRS) ranged from 1.91×10^{10} to 2.54×10^{11} Ω, and 1.34×10^{3} to 4.55×10^{4} Ω under a 300 mV read voltage, and the V_{set} and V_{reset} were located from 1.80 to 2.65 V, and −0.80 to −2.45 V (Figure S2 c, d). The endurance and retention tests conducted on the memory devices showed that the LRS and HRS were well separated and the resistance OFF/ON ratio remained at 10^{7} for repeated voltage sweeping cycles or under a continuous read voltage (Figure S2). These results suggest that ZIF-8 based devices have potential in nonvolatile memory applications.

The mechanism of resistance switching relies on the combined action of the formation of Ag nanoparticles in the insulator layer through electro-migration and the electron hopping effect in between the nanoparticles that leads to LRS, which would be fractured by application of a reversed electric field in the reset process, causing the resistance state of the memory device to switch back to HRS[18] (see the Supporting Information for details, Figure S3–S6).

Next, we chose methanol, which has a large dipole moment and abundant hydrogen bonding interactions,[19] as a prototype molecule to pursue the chemically mediated memory property. We tested the ZIF-8-based devices in saturated methanol vapor conditions, and obtained a typical nonvolatile resistance-switching property (Figure 2 a and Figure S7). Although this process was similar to that exhibited by the memory device operated in air, the data level of the memory device was noticeably changed by the methanol vapor. The HRS resistance of the ZIF-8-based memory devices decreased dramatically when the device working condition changed from air into saturated methanol vapor, and the corresponding resistance OFF/ON ratio decreased to around 10^{4} (Figure 2 b and Figure S7, S11). We conducted a tracking test that revealed that the resistance of HRS for the device in methanol vapor gradually decreased to another stable resistance state, and the curve of changing resistance fit the adsorption isotherm of methanol by ZIF-8 crystals,[20] which present the chemically mediated nature of ZIF-8-based memory devices (Figure S8). Moreover, upon desorption of the methanol molecules in ZIF-8 crystals, the HRS resistance reverted back to the initial state, and this process could be repeated by adsorbing and desorbing methanol molecules (Figure 2 b), which demonstrated the chemically responsive nature of the devices. This reliable resistance-switching property, which exhibits well-separated resistance states and the endurance of switching behavior between them triggered by electrical and chemical stimuli, can be further applied in achieving multilevel data storage.[21]

To elucidate the structural and dynamic properties of the methanol molecules adsorbed in ZIF-8 crystals, and understand how the adsorbed molecules influence the resistance state, we performed molecular dynamics simulations (Figure 3 a,b). We hypothesized that the decrease of HRS resistance may be attributed to the close-packed mode of methanol molecules in ZIF-8 pores and the consequent hydrogen bonding system among the molecules.[22] The simulation results showed that, before voltage was applied on the ZIF-8 crystals, the adsorbed methanol molecules arranged into a relatively free packing mode in the cage of ZIF-8 crystals, and the angles of the hydrogen bonds with the c-axis showed irregular distribution (Figure 3 a,c). By applying an electric field in the c-axis direction of a ZIF-8 single crystal, the methanol molecules could be arranged in an ordered fashion with the dipole of methanol aligning largely along the direction of the external field (Figure 3 b).[23] The distribution of the angles of hydrogen bonds with the c-axis mainly located in the range of 27.5–47.5 degrees with increasing strength of electric field. Under the applied electric field, with strength of 100 mV nm^{-1} (corresponding working voltage was about 1.7 V, Figure S9), more than 70 % of the methanol molecules were well-packed in an ordered mode along the direction of the electric field (Figure 3 c). Moreover, in saturated methanol vapor conditions, the average number of hydrogen bonds in a single cell of ZIF-8 crystal (N_C) and the average number of hydrogen bonds per adsorbed molecule (N_M) without applied electric field was calculated to be 21.7 and 0.79, respectively. These two values gradually decreased to 19.4 and 0.71 when the strength of electric field was increased to 100 mV nm^{-1} (Figure 3 d, Figure S12), suggesting that only a small number of the hydrogen bonds broke during the ordered rearrangement of the methanol molecules. The ordered methanol in ZIF-8 pores with extensive hydrogen bonded network would work as electron transfer pathways to decrease the HRS resistance of ZIF-8-based memory device and finally form a new resistance state.[5a]

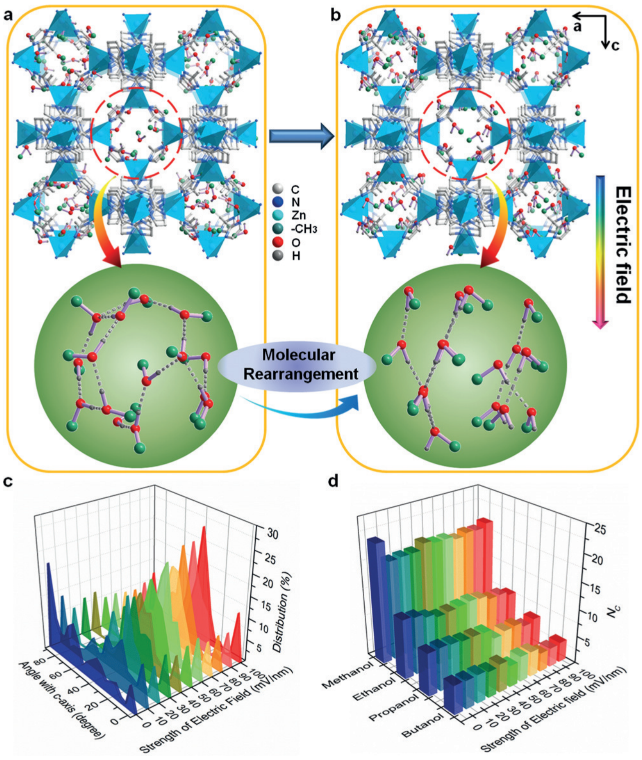

Figure 3. Schemes and statistic analysis of molecular dynamics simulations of alcohol molecules adsorbed in ZIF-8 crystals. Molecular dynamics simulation results showed the packing mode of methanol molecules in ZIF-8 crystals at saturation vapor pressure without (a) and with (b) external electric field (100 mV nm^{-1}). The enlarged figures below show the arrangement of methanol molecules and hydrogen bonds between them. c) Distributions of angles of the hydrogen bonds between methanol molecules with the c-axis under an electric field. d) Statistical analysis of the average numbers of hydrogen bonds among different alcohols adsorbed in a single cell of ZIF-8 crystal (N_C) under an electric field.

To verify the role of hydrogen bonding systems in the memory performance, a control experiment was performed using acetonitrile as the guest molecule. Compared with methanol, acetonitrile has similar polarity and loading amount in ZIF-8 crystals (Figure S10), but cannot form hydrogen bonds with each other. In saturated acetonitrile vapor, the resistance-switching property of ZIF-8-based memory did not show a significant difference compared to the one operated in air (Figure S10 a). In particular, the range of HRS resistance was nearly the same as that in air (Figure S10 b), and did not show any decrease in resistance like the devices working in methanol vapor, indicating that the abundant hydrogen bonding interactions between methanol molecules is an essential factor in achieving this chemically mediated memory performance.

Based on the methanol-mediated resistance-switching properties, we confirmed that alcohols with different length of carbon chains, which show regularity in physicochemical properties,[22] would endow a series of regular memory characteristics in chemically mediated memory devices (Figure 2 c). Figures 2 d and S11 show the statistical analysis of the HRS, LRS resistance and OFF/ON ratio of ZIF-8-based memory devices operated in saturated vapors of respective alcohols. The LRS resistance did not show much difference between conditions. But, as the number of carbon atoms in alcohols decreased, an obvious trend of decreasing HRS resistance and OFF/ON ratio was observed. In saturated methanol or ethanol vapor, the HRS resistance and OFF/ON ratio were, respectively, hundreds- or tens-of-times lower than the one in air. But for butanol and propanol, which have longer carbon chains, the relative memory performances of ZIF-8-based memory devices were quite similar to that in air.

We also introduced molecular dynamics simulations to reveal the relationship between the memory performance and alcohol structures (Figures 3 d and S12). The two values N_C and N_M of the four alcohols showed an obvious decreasing trend with increasing number of carbon atoms in their molecular structures, which matched well with the results of the memory performance tested in the corresponding alcohol vapor condition. Under an external electric field of 100 mV nm^{-1}, each butanol or propanol molecule possessed 0.31 or 0.41 hydrogen bonds, and each cell of ZIF-8 crystal had only 3.64 or 5.53 hydrogen bonds between butanol or propanol molecules, respectively. In contrast, for methanol, 19.4 hydrogen bonds in each ZIF-8 cell would dramatically decrease the HRS. For ethanol, these two values are 0.47 (N_M) and 7.84 (N_C), revealing that ethanol molecules would form more hydrogen bonds in ZIF-8 cages than butanol or propanol, causing a small decrease in HRS resistance (Figure 2d). So the inability of alcohols with longer chains to form efficient electron transfer pathways in the pores of ZIF-8 and tune the memory performance can possibly be attributed to the decline in the number of hydrogen bonds between the molecules in the ZIF-8 cells.

The chemically mediated resistance-switching property and the simulation results can be further illustrated by the adsorptive characteristics of different alcohols in ZIF-8 crystals. Firstly, the variation trend of HRS resistance and OFF/ON ratio fits the regularity of the loading amount of different alcohols in ZIF-8 crystals under each saturated vapor condition.[20,24] In particular, the significantly higher saturation-loading amount of methanol (10 mmol g^{-1}) compared to other alcohols might be the most indicative for the specificity of methanol.[22] Secondly, the limit for the packing mode of alcohol molecules with longer carbon chains in small ZIF-8 cages would impede the formation of efficient hydrogen bonding system, owing to the stronger steric hindrance and affinity with hydrogen binding interactions and show the most significant hydrophobic ZIF-8 skeleton.[19,22,25] Thus, the methanol molecules, with the highest loading amount in ZIF-8 crystals and the weakest interaction with the framework among these four alcohols, present the most abundant influence on the resistance-switching properties of Ag/ZIF-8/Si devices.

Finally, we demonstrated ZIF-8-based devices on soft substrates, which would make this chemically mediated memory device a candidate in constructing flexible and wearable electronics.[26] The alcohol-mediated resistance-switching property can still perform well in the bending state (Figures S13, S14b). In addition, we incorporated ZIF-8-based flexible memory devices on a wrist strap to demonstrate their prospective application in wearable electronics (Figure S14a). The chemically mediated memory properties were well-exhibited by the devices on the minimal bending radius (14.2 mm) part on this elliptically shaped wrist strap (Figure S14c,d). Moreover, tuned by electrical and chemical stimuli, the resistance-switching property between different data levels was shown in a continuous reading process, which presents the integrated effect of the responsiveness of ZIF-8-based memories in alcohol vapor and the electrical resistance-switching behavior (Figure S15). Thus, the reliability of the alcohol-mediated nature of ZIF-8-based flexible memory supports the potential application of chemically mediated resistance-switching devices in environment-responsive wearable electronics.

In summary, we have successfully fabricated alcohol-mediated memory devices based on ZIF-8-films with reliable resistance-switching property. Notably, the chemical responsiveness of the devices can be attributed to the supramolecular interactions between the guest molecules (alcohols) in the ZIF-8 crystals, as validated by molecular dynamics simulations and the adsorption characteristics of the different alcohol vapors. Furthermore, we achieved a flexible ZIF-8-based resistance-switching device while maintaining its chemically responsive property, which exhibits potential applications in wearable electronics. Recently, the development of memory devices with tunable resistance-switching properties, which could expand their applicability to environment-responsive devices and sensors, has begun to draw considerable attention.[27] This work will be instructive in the study of chemically mediated memory processes, and the cooperation of device engineering and supramolecular chemistry are expected to provide more opportunities in the construction of functional synapses and smart devices.

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[1] a) G. Lu, S. Li, Z. Guo, O. K. Farha, B. G. Hauser, X. Qi, Y. Wang, X. Wang, S. Han, X. Liu, J. S. DuChene, H. Zhang, Q. Zhang, X. Chen, J. Ma, S. C. Loo, W. D. Wei, Y. Yang, J. T. Hupp, F. Huo, *Nat. Chem.* **2012**, *4*, 310–316; b) C. M. McGuirk, M. J. Katz, C. L. Stern, A. A. Sarjeant, J. T. Hupp, O. K. Farha, C. A. Mirkin, *J. Am. Chem. Soc.* **2015**, *137*, 919–925; c) C. H. Kuo, Y. Tang, L. Y. Chou, B. T. Sneed, C. N. Brodsky, Z. Zhao, C. K. Tsung, *J. Am. Chem. Soc.* **2012**, *134*, 14345–14348; d) W. Zhang, Y. Liu, G. Lu, Y. Wang, S. Li, C. Cui, J. Wu, Z. Xu, D. Tian, W. Huang, J. S. DuCheneu, W. D. Wei, H. Chen, Y. Yang, F. Huo, *Adv. Mater.* **2015**, *27*, 2923–2929.

[2] a) G. Férey, *Chem. Soc. Rev.* **2008**, *37*, 191–214; b) S. Shimomura, M. Higuchi, R. Matsuda, K. Yoneda, Y. Hijikata, Y. Kubota, Y. Mita, J. Kim, M. Takata, S. Kitagawa, *Nat. Chem.* **2010**, *2*, 633–637; c) R. Banerjee, A. Phan, B. Wang, C. Knobler, H. Furukawa, M. O'Keeffe, O. M. Yaghi, *Science* **2008**, *319*, 939–943; d) H. Sato, W. Kosaka, R. Matsuda, A. Hori, Y. Hijikata, R. V. Belosludov, S. Sakaki, M. Takata, S. Kitagawa, *Science* **2014**, *343*, 167–170.

[3] a) A. Stroppa, P. Jain, P. Barone, M. Marsman, J. M. Perez-Mato, A. K. Cheetham, H. W. Kroto, S. Picozzi, *Angew. Chem. Int. Ed.* **2011**, *50*, 5847–5850; *Angew. Chem.* **2011**, *123*, 5969–5972; b) Y. Cui, Y. Yue, G. Qian, B. Chen, *Chem. Rev.* **2012**, *112*, 1126–1162; c) H. Y. Ye, Y. Zhang, D. W. Fu, R. G. Xiong, *Angew. Chem. Int. Ed.* **2014**, *53*, 6724–6729; *Angew. Chem.* **2014**, *126*, 6842–6847.

[4] a) S. M. Yoon, S. C. Warren, B. A. Grzybowski, *Angew. Chem. Int. Ed.* **2014**, *53*, 4437–4441; *Angew. Chem.* **2014**, *126*, 4526–4530; b) A. A. Talin, A. Centrone, A. C. Ford, M. E. Foster, V. Stavila, P. Haney, R. A. Kinney, V. Szalai, F. E. Gabaly, H. P. Yoon, F. Léonard, M. D. Allendorf, *Science* **2014**, *343*, 66–69.

[5] a) L. Pan, G. Liu, H. Li, S. Meng, L. Han, J. Shang, B. Chen, A. E. Platero-Prats, W. Lu, X. Zou, R. W. Li, *J. Am. Chem. Soc.* **2014**, *136*, 17477–17483; b) L. Pan, Z. Ji, X. Yi, X. Zhu, X. Chen, J. Shang, G. Liu, R.-W. Li, *Adv. Funct. Mater.* **2015**, *25*, 2677–2685.

[6] C. Zhang, P. Chen, W. Hu, *Chem. Soc. Rev.* **2015**, *44*, 2087–2107.

[7] a) D. B. Strukov, G. S. Snider, D. R. Stewart, R. S. Williams, *Nature* **2008**, *453*, 80 – 83; b) D. H. Kwon, K. M. Kim, J. H. Jang, J. M. Jeon, M. H. Lee, G. H. Kim, X. S. Li, G. S. Park, B. Lee, S. Han, M. Kim, C. S. Hwang, *Nat. Nanotechnol.* **2010**, *5*, 148 – 153; c) M. J. Lee, C. B. Lee, D. Lee, S. R. Lee, M. Chang, J. H. Hur, Y. B. Kim, C. J. Kim, D. H. Seo, S. Seo, U. I. Chung, I. K. Yoo, K. Kim, *Nat. Mater.* **2011**, *10*, 625 – 630; d) J. Kim, D. Son, M. Lee, C. Song, J.-K. Song, J. H. Koo, D. J. Lee, H. J. Shim, J. H. Kim, M. Lee, T. Hyeon, D.-H. Kim, *Sci. Adv.* **2016**, *2*, e1501101.

[8] R. Waser, R. Dittmann, G. Staikov, K. Szot, *Adv. Mater.* **2009**, *21*, 2632 – 2663.

[9] A. Asamitsu, Y. Tomioka, H. Kuwahara, Y. Tokura, *Nature* **1997**, *388*, 50 – 52.

[10] U. Russo, D. Ielmini, C. Cagli, A. L. Lacaita, *IEEE Trans. Electron Devices* **2009**, *56*, 193 – 200.

[11] a) M. Ungureanu, R. Zazpe, F. Golmar, P. Stoliar, R. Llopis, F. Casanova, L. E. Hueso, *Adv. Mater.* **2012**, *24*, 2496 – 2500; b) A. Bera, H. Peng, J. Lourembam, Y. Shen, X. W. Sun, T. Wu, *Adv. Funct. Mater.* **2013**, *23*, 4977 – 4984; c) H. Tan, G. Liu, X. Zhu, H. Yang, B. Chen, X. Chen, J. Shang, W. D. Lu, Y. Wu, R. W. Li, *Adv. Mater.* **2015**, *27*, 2797 – 2803.

[12] a) E. R. Kandel, *Science* **2001**, *294*, 1030 – 1038; b) A. Citri, R. C. Malenka, *Neuropsychopharmacology* **2008**, *33*, 18 – 41; c) S. F. Cooke, T. V. Bliss, *Brain* **2006**, *129*, 1659 – 1673.

[13] a) K. S. Park, Z. Ni, A. P. Cote, J. Y. Choi, R. Huang, F. J. Uribe-Romo, H. K. Chae, M. O'Keeffe, O. M. Yaghi, *Proc. Natl. Acad. Sci. USA* **2006**, *103*, 10186 – 10191; b) A. Phan, C. J. Doonan, F. J. Uribe-Romo, C. B. Knobler, M. O'Keeffe, O. M. Yaghi, *Acc. Chem. Res.* **2010**, *43*, 58 – 67.

[14] C. T. Poon, D. Wu, W. H. Lam, V. W. Yam, *Angew. Chem. Int. Ed.* **2015**, *54*, 10569 – 10573; *Angew. Chem.* **2015**, *127*, 10715 – 10719.

[15] a) R. C. Webb, A. P. Bonifas, A. Behnaz, Y. Zhang, K. J. Yu, H. Cheng, M. Shi, Z. Bian, Z. Liu, Y. S. Kim, W. H. Yeo, J. S. Park, J. Song, Y. Li, Y. Huang, A. M. Gorbach, J. A. Rogers, *Nat. Mater.* **2013**, *12*, 938 – 944; b) S. T. Han, Y. Zhou, V. A. Roy, *Adv. Mater.* **2013**, *25*, 5425 – 5449.

[16] a) S. H. Jo, T. Chang, I. Ebong, B. B. Bhadviya, P. Mazumder, W. Lu, *Nano Lett.* **2010**, *10*, 1297 – 1301; b) T. Ohno, T. Hasegawa, T. Tsuruoka, K. Terabe, J. K. Gimzewski, M. Aono, *Nat. Mater.* **2011**, *10*, 591 – 595; c) M. Prezioso, F. Merrikh-Bayat, B. D. Hoskins, G. C. Adam, K. K. Likharev, D. B. Strukov, *Nature* **2015**, *521*, 61 – 64.

[17] G. Lu, J. T. Hupp, *J. Am. Chem. Soc.* **2010**, *132*, 7832 – 7833.

[18] a) R. Waser, M. Aono, *Nat. Mater.* **2007**, *6*, 833 – 840; b) X. Tian, S. Yang, M. Zeng, L. Wang, J. Wei, Z. Xu, W. Wang, X. Bai, *Adv. Mater.* **2014**, *26*, 3649 – 3654; c) Y. Yang, P. Gao, S. Gaba, T. Chang, X. Pan, W. Lu, *Nat. Commun.* **2012**, *3*, 732; d) H. Wang, Y. Du, Y. Li, B. Zhu, W. R. Leow, Y. Li, J. Pan, T. Wu, X. Chen, *Adv. Funct. Mater.* **2015**, *25*, 3825 – 3831.

[19] J. A. Gee, J. Chung, S. Nair, D. S. Sholl, *J. Phys. Chem. C* **2013**, *117*, 3169 – 3176.

[20] K. Zhang, R. P. Lively, M. E. Dose, A. J. Brown, C. Zhang, J. Chung, S. Nair, W. J. Koros, R. R. Chance, *Chem. Commun.* **2013**, *49*, 3245 – 3247.

[21] Y. Zhou, S. T. Han, X. Chen, F. Wang, Y. B. Tang, V. A. Roy, *Nat. Commun.* **2014**, *5*, 4720.

[22] K. Zhang, L. Zhang, J. Jiang, *J. Phys. Chem. C* **2013**, *117*, 25628 – 25635.

[23] a) W. Sun, Z. Chen, S.-Y. Huang, *Fluid Phase Equilib.* **2005**, *238*, 20 – 25; b) S. J. Suresh, A. L. Prabhu, A. Arora, *J. Chem. Phys.* **2007**, *126*, 134502.

[24] J. C. Saint Remi, T. Remy, V. Van Hunskerken, S. van de Perre, T. Duerinck, M. Maes, D. De Vos, E. Gobechiya, C. E. Kirschhock, G. V. Baron, J. F. Denayer, *ChemSusChem* **2011**, *4*, 1074 – 1077.

[25] H. Amrouche, B. Creton, F. Siperstein, C. Nieto-Draghi, *RSC Adv.* **2012**, *2*, 6028.

[26] a) T. Sekitani, U. Zschieschang, H. Klauk, T. Someya, *Nat. Mater.* **2010**, *9*, 1015 – 1022; b) S. C. Mannsfeld, B. C. Tee, R. M. Stoltenberg, C. V. Chen, S. Barman, B. V. Muir, A. N. Sokolov, C. Reese, Z. Bao, *Nat. Mater.* **2010**, *9*, 859 – 864; c) G. Schwartz, B. C. Tee, J. Mei, A. L. Appleton, H. Kim do, H. Wang, Z. Bao, *Nat. Commun.* **2013**, *4*, 1859; d) D. Son, J. Lee, S. Qiao, R. Ghaffari, J. Kim, J. E. Lee, C. Song, S. J. Kim, D. J. Lee, S. W. Jun, S. Yang, M. Park, J. Shin, K. Do, M. Lee, K. Kang, C. S. Hwang, N. Lu, T. Hyeon, D. H. Kim, *Nat. Nanotechnol.* **2014**, *9*, 397 – 404; e) G. Sun, J. Liu, L. Zheng, W. Huang, H. Zhang, *Angew. Chem. Int. Ed.* **2013**, *52*, 13351 – 13355; *Angew. Chem.* **2013**, *125*, 13593 – 13597.

[27] a) T. Sekitani, T. Yokota, U. Zschieschang, H. Klauk, S. Bauer, K. Takeuchi, M. Takamiya, T. Sakurai, T. Someya, *Science* **2009**, *326*, 1516 – 1519; b) J. Park, S. Lee, J. Lee, K. Yong, *Adv. Mater.* **2013**, *25*, 6423 – 6429; c) S. Ambrogio, S. Balatti, S. Choi, D. Ielmini, *Adv. Mater.* **2014**, *26*, 3885 – 3892.

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