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Electronic Supplementary Information
For

Interfacial synthesis of large-area coordination polymer membrane for rewritable nonvolatile memory devices

Zepu Zhang‡, Yijie Nie‡, Weiwei Hua, Jingxuan Xu, Chaoyi Ban, Fei Xiu*, Juqing Liu*

Key Laboratory of Flexible Electronics (KLOFE) & Institute of Advanced Materials (IAM), Nanjing Tech University (NanjingTech), 30 South Puzhu Road, Nanjing 211816, China.

*Corresponding author: iamfxiu@njtech.edu.cn;

iamjqliu@njtech.edu.cn;

‡ Z.P.Z and Y.J.N contributed equally to this work.

Experimental section

Preparation of coordination polymer membrane： The 1,2,4,5-benzene-tetramine tetrahydrochloride was purchased from Beijing Huaweiruike Chemical. The Cobalt acetate tetrahydrate and Triethylamine was purchased from Guoyao. The 1,2,4,5-benzene-tetramine tetrahydrochloride solution was obtained by dissolving 40 mg 1,2,4,5-benzene-tetramine tetrahydrochloride into 20mL DI water (Solution A). Similarly, Solution B was obtained by adding 13 mg cobalt acetate tetrahydrate ($Co(CH_3COO)_2•4H_2O$) in 20 mL water. First, 560 μL of triethylymine was added to the solution B, followed by mixing solution B with solution A. The mixture was sonicated for 5 minutes and then maintained still at 60℃ for 3 hours, leading to the formation of large-area brown film at the gas-liquid interface.

Device Fabrication: First, an ITO substrate (20mm×20mm) was pre-cleaned by ultrasonication for 15 min in deionized water, ethanol, and isopropanol, subsequently. Then the prepared coordination polymer membrane was coated onto the ITO substrate by lifting the ITO support up directly from reacted solution and then dried at 60 ℃ for 30 minutes. Finally, 150 nm top Al electrodes were thermally evaporated on membrane/ITO. Importantly, thickness of the polymer active layer was modulated by controlling the concentration of polymer suspension as well as the reaction duration time.

Characterization: The thickness size of thin film membranes were measured by scanning electron microscopy (SEM, JSM 7800F). X-ray diffraction patterns of out-of-plane and in-plane were obtained by using Smartlab (3KW) and Smartlab Ⅲ, respectively. X-ray photoelectron spectroscopy (XPS, Thermo escalab 250Xi) is used to confirm the elements and chemical composition of membranes. A Fourier transform infrared spectroscopy (FTIR, DT-40) spectrum is measured neat on a KBr plate. The electrical characteristics (I-V and I-t) of the devices were conducted by using a Keithley 4200 under ambient conditions.

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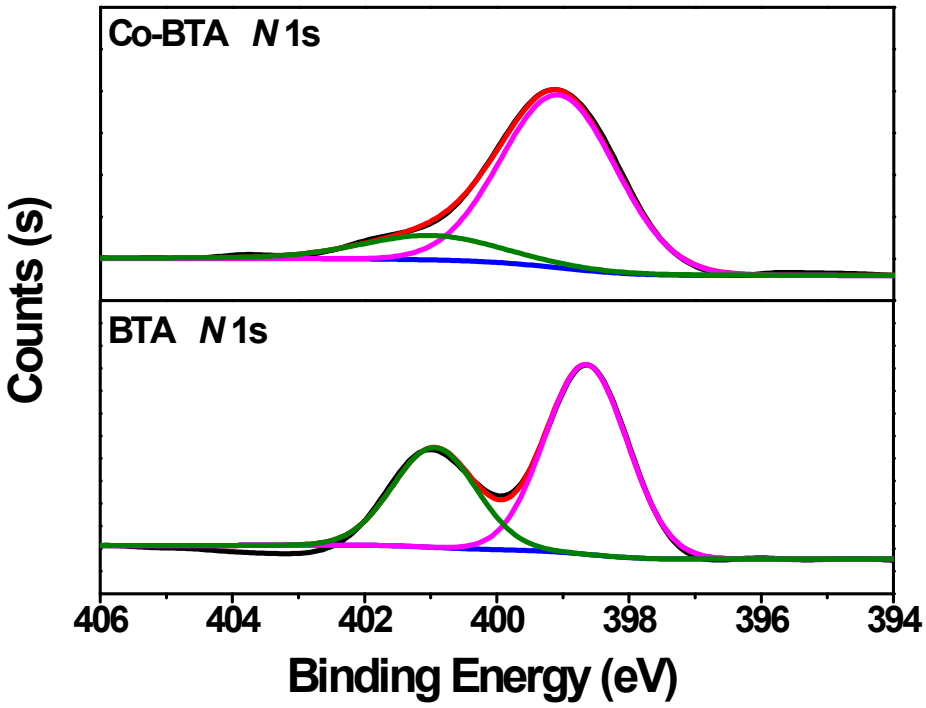

Figure S1. Comparison of N 1s core lever between film and ligand.

Table S1. Element quantities analysis of C, O, N, Co of the coordination polymer membrane based on the XPS analysis.

Element	C	O	N	Co
At%	56.06	23.64	15.80	4.50