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

Supplementary Information

Tea Helps Neuromorphic Computing: Flexible Memristors from Tea Polyphenols

Wei-Ning Zhang,[a] Xiang-Yang Li,[a] Zhe Zhang,[a] Wen-Jing Sun,[a] Hao-Li Zhang [a]*

[a] State Key Laboratory of Applied Organic Chemistry, College of Chemistry and Chemical Engineering, Lanzhou University, Lanzhou, 730000, P. R. China

* Corresponding author. Email: haoli.zhang@lzu.edu.cn

Experimental Section

Device fabrication.

Tea Polyphenols (TPs) was purchased from Macklin with 99% purity and used without further purification. 5 mg of TPs was dissolved in 1 mL of deionized water and stirred until dissolved, after which 50 mg of silver nitrate was added to the solution and stirred magnetically in a condition protected from light. The ITO conductive glass (ITO, thickness 150 ± 50 nm) substrate was cleaned twice after 10 minutes of sonication using soapy water, ultrapure deionized water, acetone, and isopropanol, and then dried using N_2 gas. After washing, the ITO conductive glass was dried in a vacuum oven at a temperature of 120 °C for half an hour. The flexible memristor device used polyethylene terephthalate (PET) as the substrate, PET substrate with ITO conductive film was cleaned in the same way and sequence as the glass substrate, without high-temperature drying. ITO bottom electrode was treated in an oxygen vacuum plasma processor for five minutes before device fabrication. The prepared solution was filtered by a syringe filter with a pore size of 0.22 μm. For a 1 cm × 1 cm substrate, 40 μL of the solution was drop-coated on the ITO bottom electrode. The TPs-Ag^+ film was dried at ambient temperature and next vacuum annealed at 50 °C overnight to get a uniform layer. The Ag top electrode with a thickness of 150 nm was deposited using thermal evaporation.

Materials characterization.

Nuclear magnetic resonance (NMR) spectrum was recorded in CH_3OD solution at 25 ℃ employing Bruker Avance III 400 MHz liquid state NMR spectrometer. Fourier transform infrared spectroscopy (FT-IR) was performed using a Nicolet iS50 spectrometer (Thermo Fisher Scientific, USA). Ultraviolet-visible (UV–Vis) absorption spectrum was recorded using Hitachi U-3900 UV-vis spectrophotometer.

Ultra-high performance liquid chromatography-mass spectrometry (UPLC-MS): The LC analysis of TPs materials was carried out using Vanquish ultra-high performance liquid chromatography system (Thermo Scientific) consisting of a Hypersil GOLD C_{18} column (100 mm × 2.1 mm, 1.9 μm; Thermo Scientific). The

column temperature was 35 ℃, and the injection volume was 5.0 μL. The mobile phases composed of water containing 0.1% formic acid (phase A: 85%) and acetonitrile (phase B: 15%) were eluted in isocratic mode at a flow rate of 0.3 mL/min. The MS analysis of TPs materials was conducted on the Orbitrap Exploris 120 mass spectrometer with electrospray ion source (ESI) in negative ionization mode.

Device characterization.

SEM analysis was carried out using Hitachi S-4800 system. XPS data was obtained using Kratos Axis Supra System. The electrical characterizations of the fabricated devices were conducted in a nitrogen atmosphere under ambient temperature using a semiconductor parameter analyzer (Keithley 4200-SCS, USA). In a standard test setup, the device was positioned within a probe station, with bias voltages applied to the Ag top electrode, while the ITO bottom electrode remained grounded.

Materials characterization.

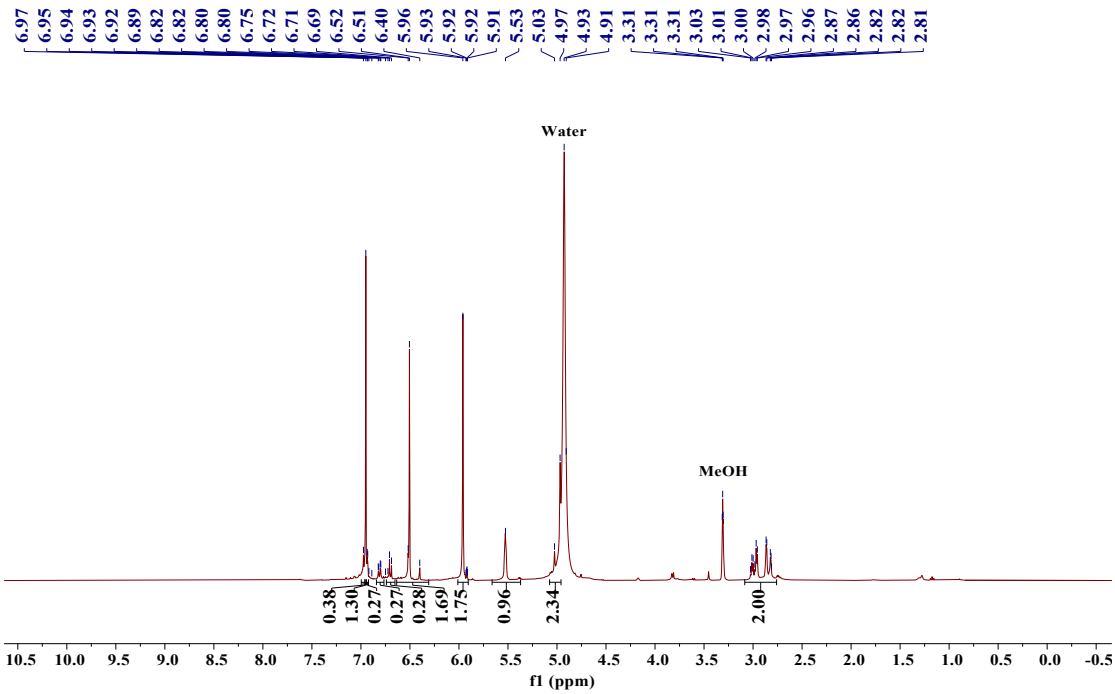

Fig S1. ^{1}H NMR spectrum of the TPs materials used in this work, recorded in deuterated methanol.

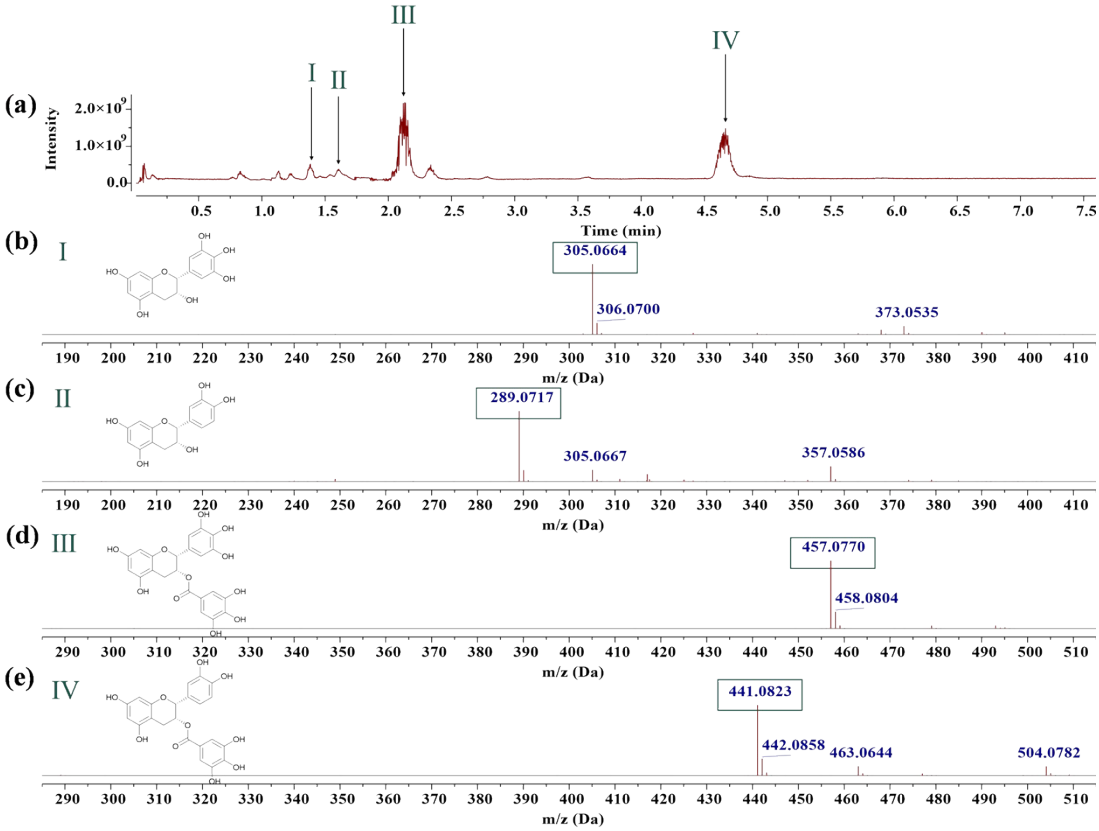

Fig S2. UPLC-MS spectrum of the TPs materials used in this work. (a) The UPLC spectra indicate that III and IV are main components of TPs materials. (b-e) The MS spectra reveal that the main components III and IV are epigallocatechin gallate (EGCG) and epicatechin gallate (ECG), respectively.

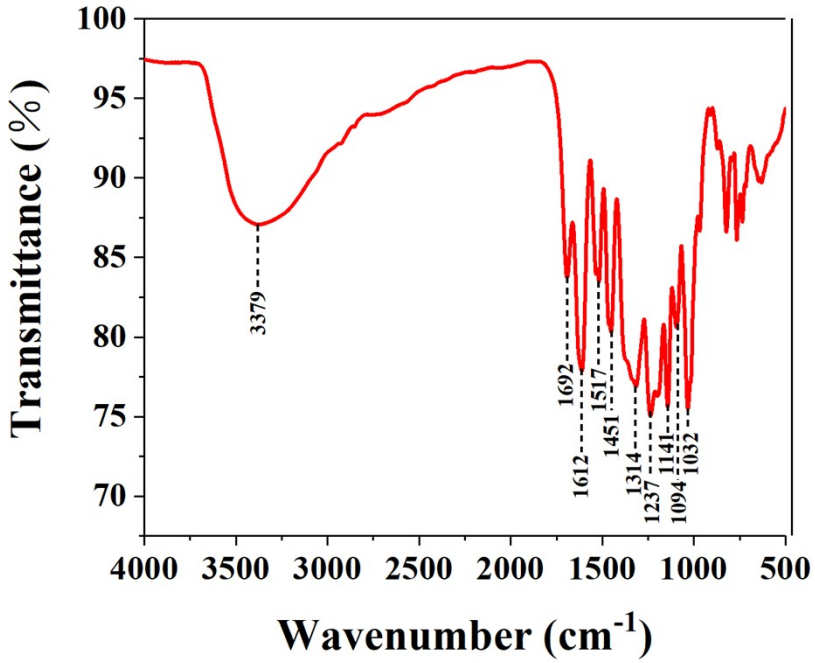

Fig S3. Fourier transform infrared spectroscopy (FTIR) of the TPs materials used in this work.

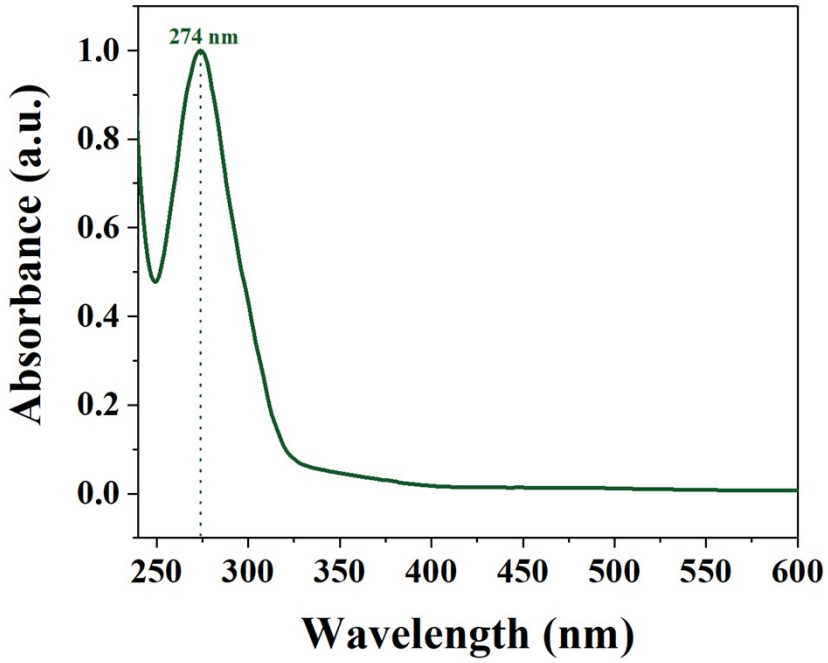

Fig S4. UV–Vis absorption spectrum of the TPs materials used in this work.

Device characterization.

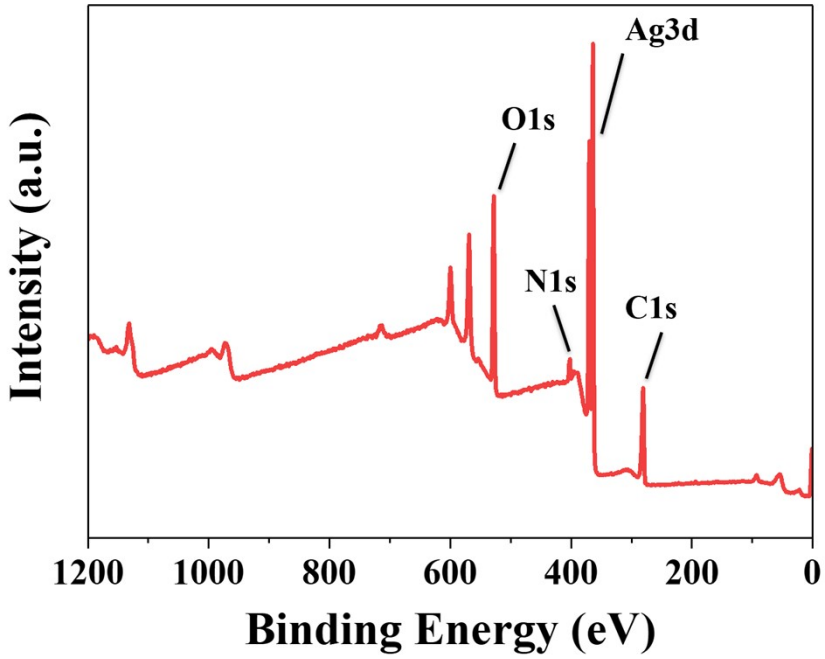

Fig S5. XPS survey spectrum of the TPs-Ag^{+} film.

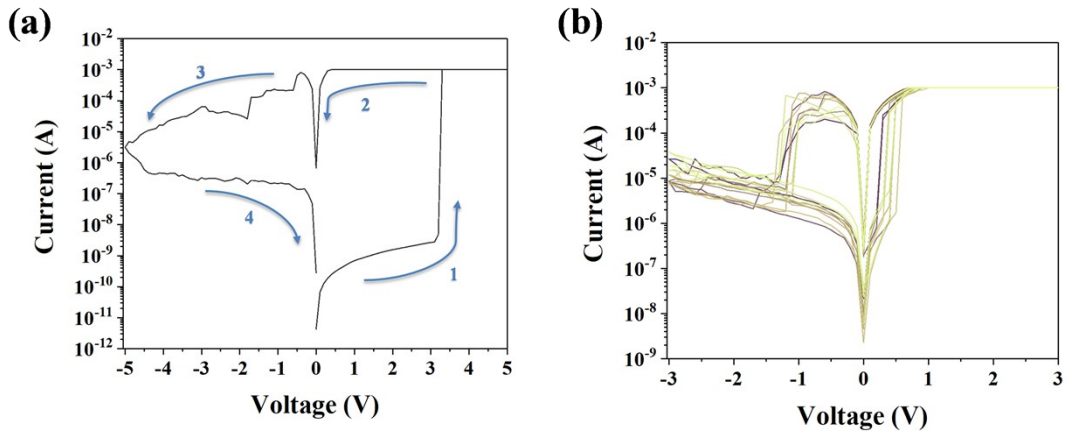

Fig S6. I−V characteristics of the TPs device with I_{CC} = 10^{-3} A. (a) Illustration of the memristor electroforming process and nonvolatile properties, indicating that the device reverts to the initial state only when the opposite voltage is applied (as indicated by the arrows). (b) Representative I–V curves after the first voltage sweep.

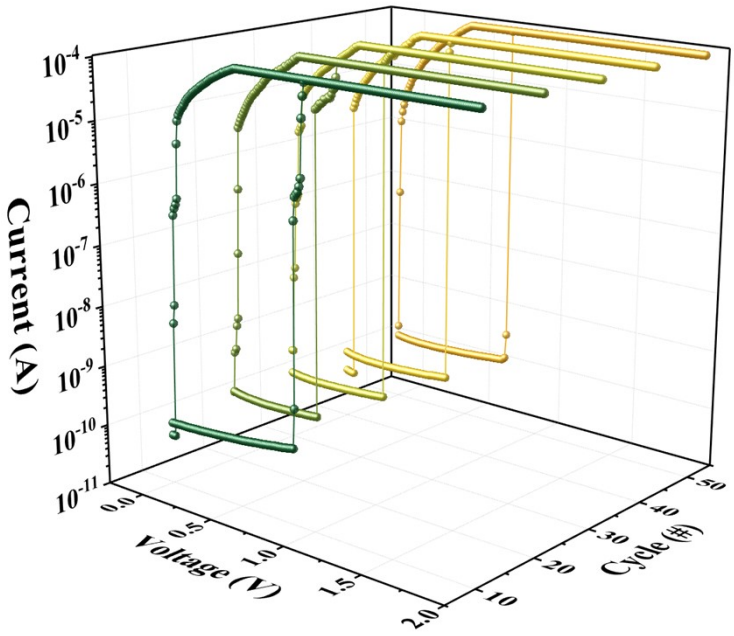

Fig S7. The continuous 50 I–V sweeps of a memristor device at I_{CC} = 10^{-4} A.

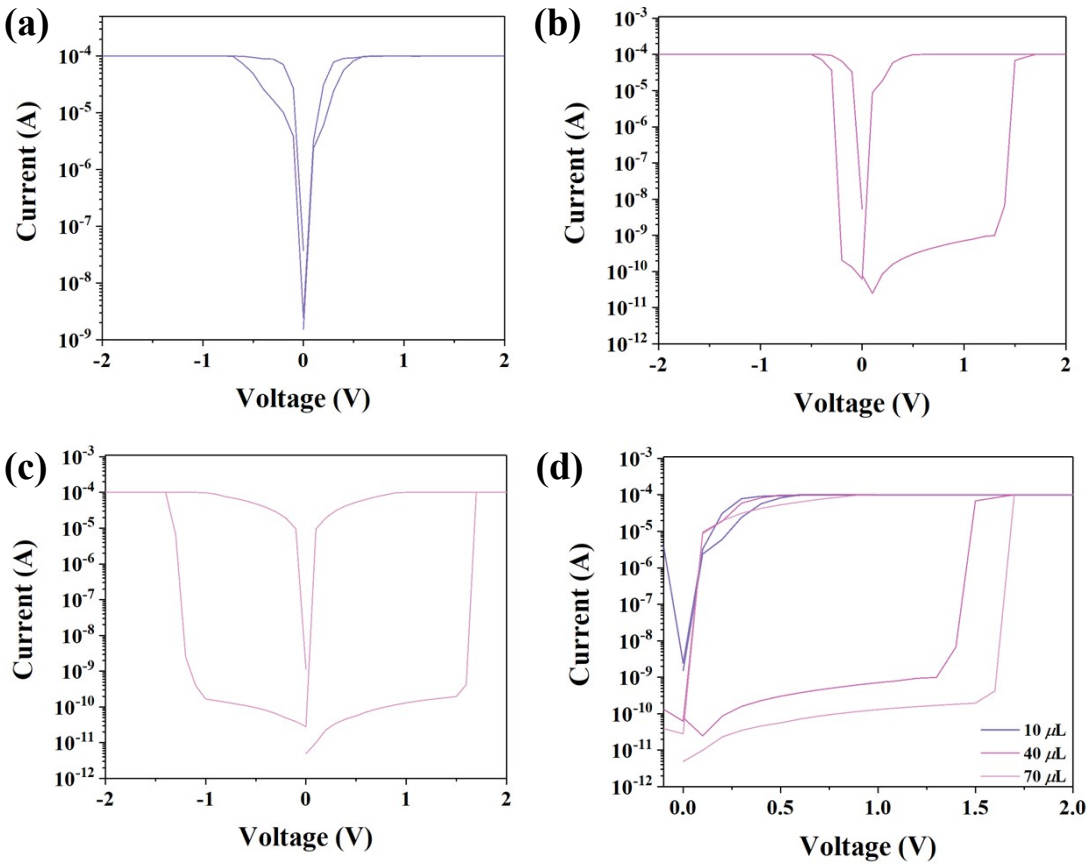

Fig S8. I−V characteristics of the device during voltage sweeps with different dosages of tea polyphenol solution. (a) 10 μL; (b) 40 μL; (c) 70 μL

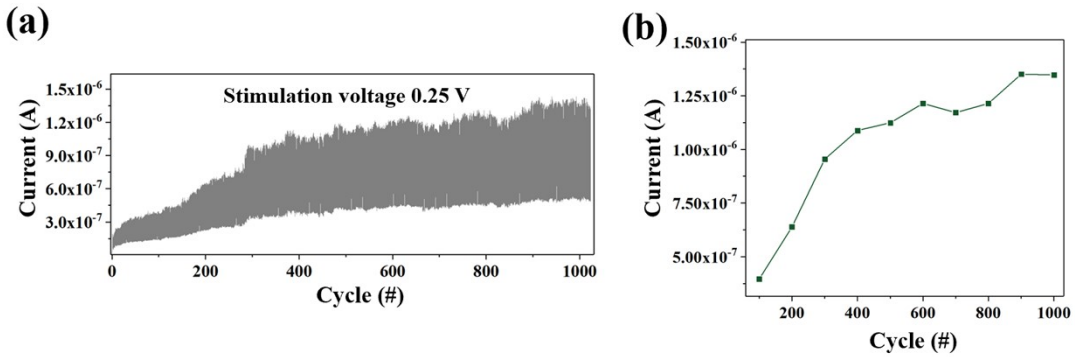

Fig S9. (a) Schematic indicating EPSC as a function of stimulation voltage cycle number. (b) SNDP simulation under the repeated stimulation voltage of 0.25 V.

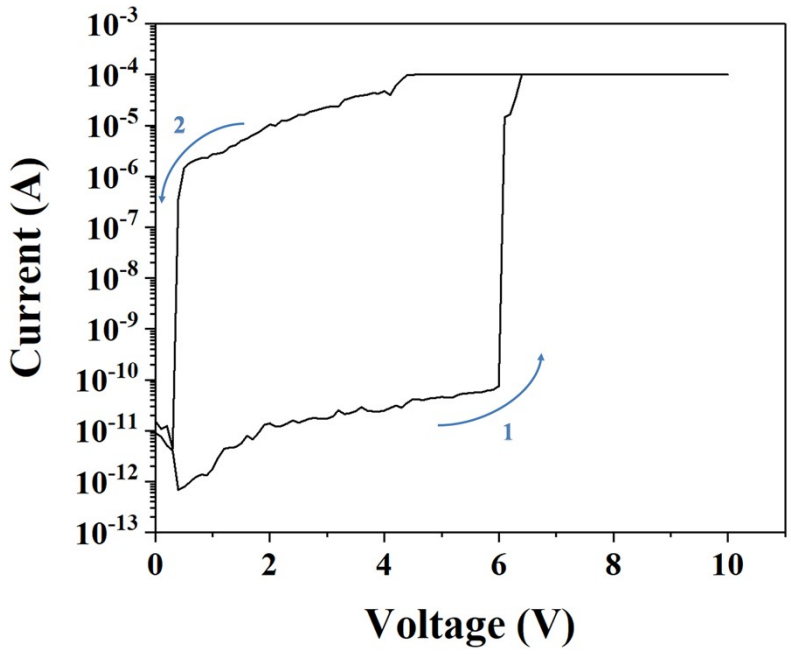

Fig S10. An electroforming process to initiate the Ag CFs of the TP-based flexible memristor. The sweep direction is indicated by arrows.