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

Supporting Information.

GO was synthesized via acid oxidation of natural graphite flakes using a modified Hummers' method. Natural graphite (4 g) and $NaNO_3$ (4 g) were placed into cold (0 °C) H_2SO_4 (150 mL). $KMnO_4$ (15 g) was gradually added with stirring. The temperature was held below 5 °C and the mixture was stirred for a period of 1 h. Then, the mixture was stirred at 35 °C (± 3 °C) for a further 16 h. Subsequently, distilled water (200 mL) was added and the temperature was increased to more than 90 °C. This temperature was maintained for 1 h, and the reaction was terminated by adding distilled water (200 mL) and 30% H_2O_2 solution (20 mL). The mixture was then washed with HCl aqueous solution and distilled water. Solid GO specimens were obtained via freeze drying. Figure S1 shows the AFM surface morphology of GO. Chemically derived graphene oxide (GO) is an atomically thin sheet of graphite that has traditionally served as a precursor for graphene, but is now attracting increasing attention from chemists based on its own characteristics. After a chemical reaction may occur between the $-NH_2$ in the KH550 and the $-COOH$ in the GO, such thin sheets are wrapped by KH550 and lead to the formation of porous structures.

To test the proton conductivities and the specific capacitance of KH550-GO solid electrolyte, an ITO/KH550-GO/IZO sandwich structure was prepared, as shown in Figure S2. The patterned 150-nm-thick IZO electrode was deposited on the KH550-GO-coated ITO PET substrate via radio-frequency (RF) magnetron sputtering with an IZO ceramic target (In_2O_3:ZnO = 90:10 wt.%). The deposition pressure, working power, and Ar flow rate were 0.5 Pa, 100 W, and 14 sccm, respectively. The length and width of the patterned IZO electrode are 150 μm and 1000 μm, respectively.

The conductivity was calculated using the following equation.

$$\sigma = \frac{d}{R_0 \times L \times W}$$

where d is the solid electrolyte thickness. The solid electrolyte thickness of 17.6 μm, 16.5 μm and 17.1 μm of KH550-GO solid electrolyte, KH550 solid electrolyte and GO solid electrolyte are obtained by SEM, respectively. L (1000 μm) and W (150 μm) are the length and width of the electrode area, respectively. R_0 is the measured resistance.

From the perspective of the group, the deprotonated ability in –COOH is stronger than that in Si-OH, while the proton conductivity of the different materials is different. Firstly, due to the strong hydrophilic property, although KH550 is alkaline (~9), experimentally the proton conductivity of pure KH550 solid electrolyte is larger than that of pure GO slightly. The devices gated by pure KH550 solid electrolyte were fabricated, the electrical properties is shown as the followed Figure. S3. Secondly, although pure GO solid electrolyte-gate synaptic transistors show good electrical properties, the power dissipation is a little larger and it is not easy to fabricate GO solid electrolyte. [Proton-Conducting Graphene Oxide-Coupled Neuron Transistors for Brain-Inspired Cognitive Systems, Adv. Mater. 2016, 28, 3557–3563] Finally, when KH550 solution and GO solution is mixed, the chemical reaction may occur between $-NH_2$ in the KH550 and $-COOH$ in the GO and the new solution (KH550-GO) is obtained as shown in Figure. S4(a). Although the number of protons would be reduced in the KH550-GO solution, it can not affect the proton conductivity of KH550-GO solid electrolyte. For KH550-GO solid electrolyte, KH550-GO solid electrolyte keeps the good hydrophilic of KH550 solid electrolyte. Water molecules in the air can be adsorbed and ionized under the effect of electric field. Therefore, the proton conductivity of KH550-GO solid electrolyte could be improved. If the chemical reaction can be not carried out, the solution would be layered after several days as shown in Figure. S4(d). Therefore, after the chemical reaction, KH550-GO solid electrolyte is a new kind of composite materials, but not kind of mixed materials. Therefore, the proton conductivity of KH550-GO solid electrolyte is greater than that of pure KH550 solid electrolyte or pure GO solid electrolyte.

The protons of KH550-GO solid electrolyte are mainly from the ionization of water molecules which are adsorbed from the air. After the chemical reaction between $-NH_2$ in the KH550 and $-COOH$ in the GO, KH550-GO solid electrolyte keeps the good hydrophilic of KH550 solid electrolyte and dispersion of GO solid electrolyte. Therefore, the proton conductivity of KH550-GO solid electrolyte is not depended on the group of the deprotonated ability of Si-OH or –COOH.

The SRDP responses to the pulse frequency are illustrated in Figure S3. Relevant pulses with 1-V amplitude and 5-ms width were applied to the device, as shown in Figure S3 (a)–(c). The channel current used here was either the EPSC or the inhibitory postsynaptic current (IPSC), which is related to the release probability of a neurotransmitter in a bio-synapse, as shown in Figure S3 (d). Firstly, two specific pluses with 0.2-V amplitude and 5-ms width were applied to the drain electrode. Then, two groups of pulses with the same f (25 Hz) were applied to the drain and source electrodes, within the interval between the two specific pluses. In addition, a group of pulses with 5-Hz f were applied to the gate electrode. Because the gate-pulse f were decreased, no protons could accumulate and the channel current induced by the second specific pulse would be inhibited.

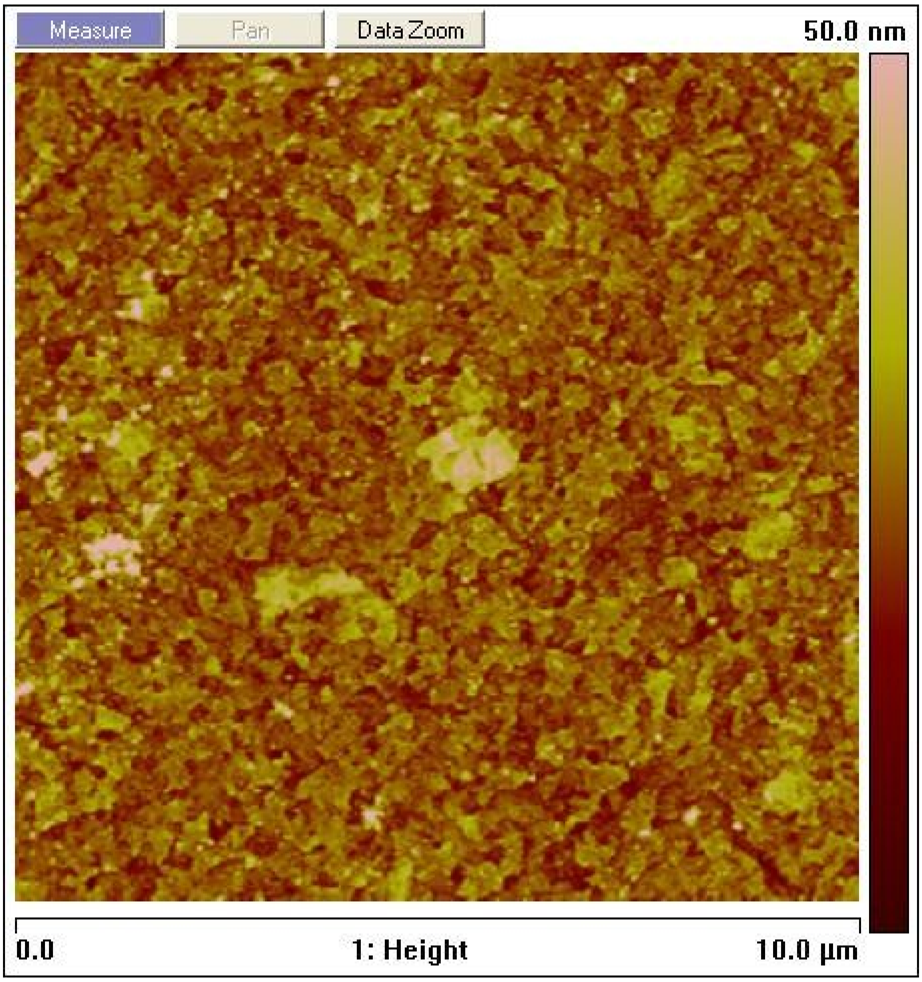

Figure S1. The AFM surface morphology of GO

Figure S2. An ITO/KH550-GO/IZO sandwich structure for testing the proton conductivities and the specific capacitance

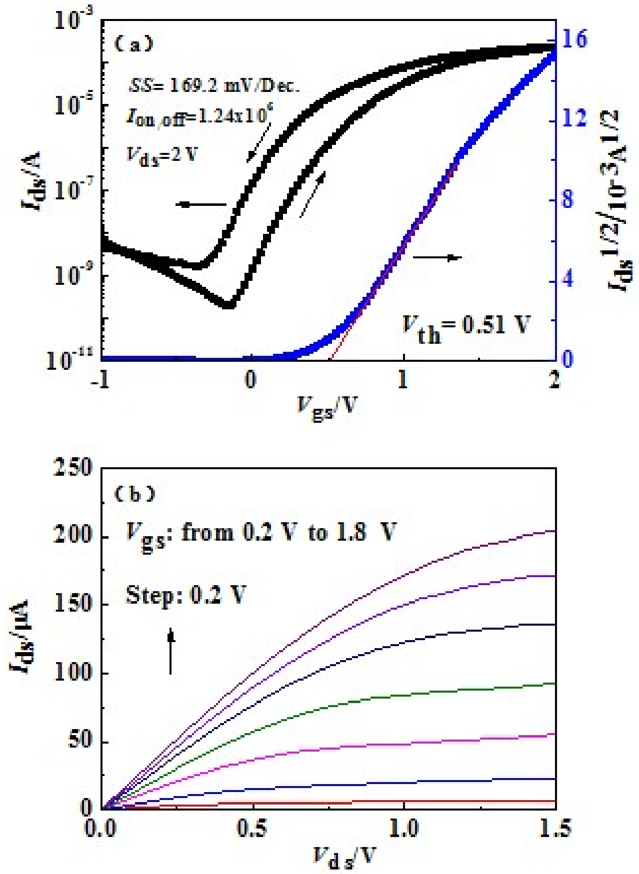

Figure. S3 (a) Transfer characteristics (I_{ds}-V_{gs}) of the IZO thin film transistor gated by KH550 solid electrolyte at V_{ds}=2 V. The curves are displayed in logarithmic scales and square root scales (b) Output characteristics (I_{ds}-V_{ds} curves) of of the IZO thin film transistor gated by KH550 solid electrolyte with V_{gs}=0.2 V to 1.8 V in 0.2 V steps from bottom to top.

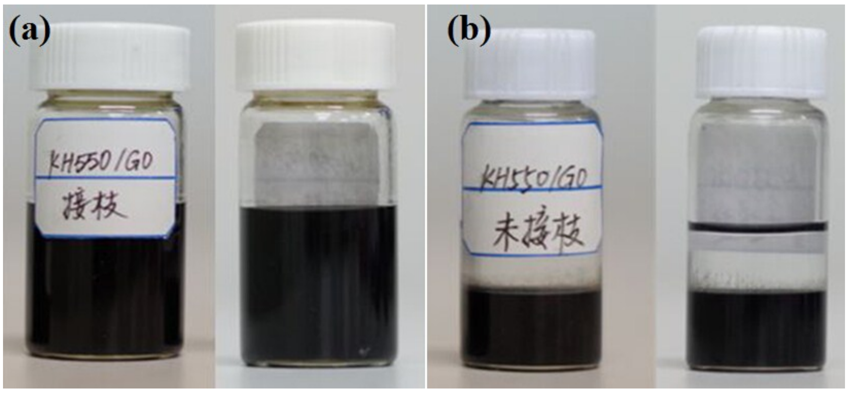

Figure. S4 (a) KH550-GO solution after the chemical reaction which occurs between $-NH_2$ in the KH550 and $-COOH$ in the GO. (b) the mixed solution of KH550 and GO.

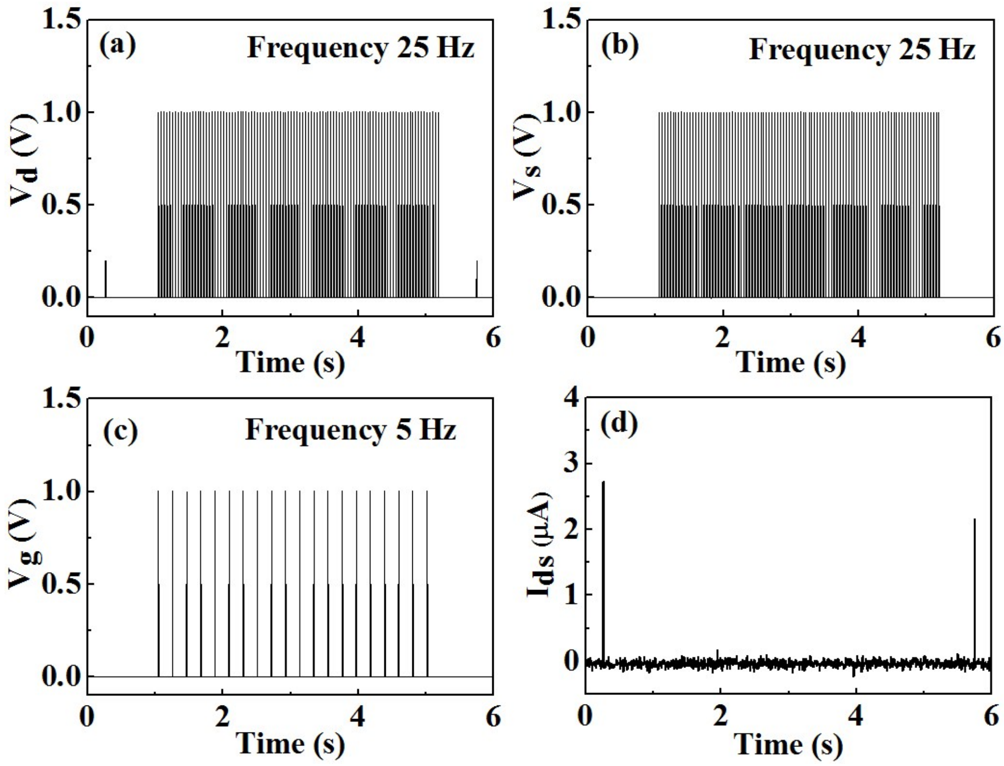

Figure S5. (a) Pulse stimulations with the frequency of 25 Hz applied in drain electrode of the flexible protonic/electronic hybrid synaptic transistor. (b) Pulse stimulations with the frequency of 25 Hz applied in source electrode of the flexible protonic/electronic hybrid synaptic transistor. (c) Pulse stimulations with the frequency of 5 Hz applied in gate electrode of the flexible protonic/electronic hybrid synaptic transistor. (d) The channel current related to pulse stimulations.