Supporting information

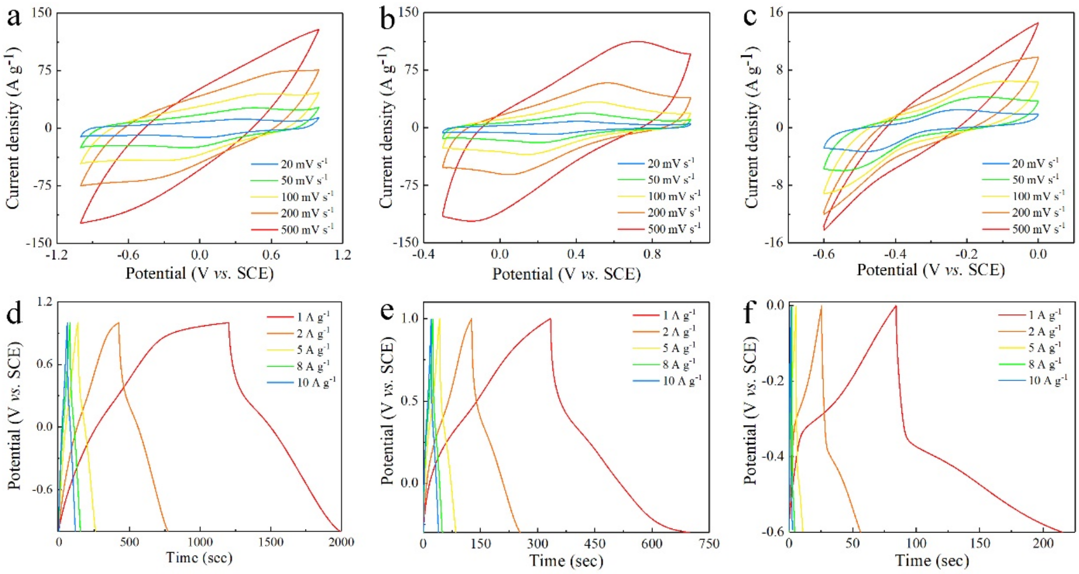

figure S1. the CV curves of HTO in 1 M Na_2SO_4 (a), 1 M H_2SO_4 (b) and 1 M KOH (c) at different scan rates. GCD curves of HTO in 1 M Na_2SO_4 (d), 1 M H_2SO_4 (e) and 1 M KOH (f) at different current density.

Table S1. The detailed Electrochemical properties of HTO in Na_2SO_4, H_2SO_4 and KOH

	KOH	H_2SO_4	Na_2SO_4
potential window (V)	0.6	1.3	2
specific capacitance (F g^{-1})	174.7	280.8	394.6
rate capability (%)	12.2	53.1	72.6
Rs (Ω)	6.146	1.881	2.719
Rct (Ω)	6.451	4.916	1.574

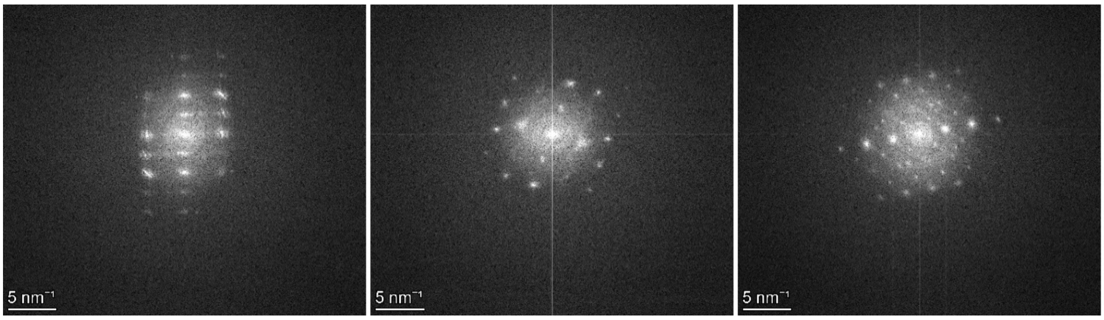

figure S2. The electron diffraction patterns of HTO-air, HTO-Ar and HTO-Ar/C

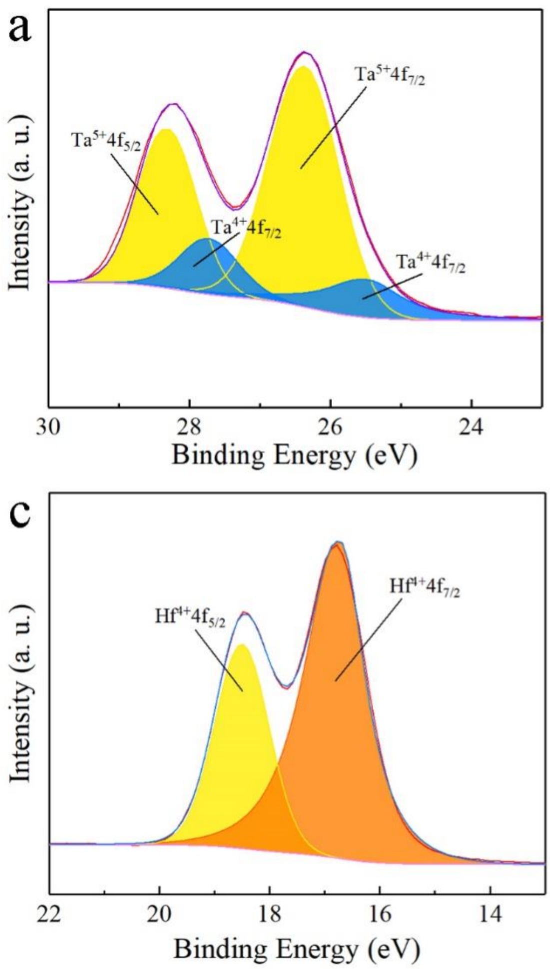

Figure S3 Ta and Hf XPS spectra of the HTO-Ar (a, b) and HTO-Ar/C (c, d).

Capacitance Contribution Calculation Methods

Trasatti Method

The Trasatti method was used to differentiate the capacitance contribution from pseudocapacitance (C_p) and electrical double layer capacitance (C_{dl}) reactions. CV measurements of COFs were first obtained with the scan rates ranging from 1 to 1000 mV s^{-1}. Then, corresponding gravimetric capacitances were evaluated based on The integral area of the CV curve.

Plotting the reciprocal of gravimetric capacitances (C^{-1}) against the square root of scan rates ($v^{0.5}$) should yield a linear correlation between them. Specifically, the correlation can be described by the following Equation S1:

$$C^{-1} = constant\ v^{0.5} + C_T^{-1} \quad \text{(S1)}$$

Where C was experimental gravimetric capacitance, v was the scan rate and C_T was the total capacitance, respectively. The "total capacitance" equals the sum of electrical double layer capacitance and pseudocapacitance.

Plotting the gravimetric capacitances (C) against the reciprocal of square root of scan rates ($v^{-0.5}$) should also give a linear correlation described by the following Equation S2 (if assuming a semi-infinite diffusion of ions):

$$C = constant\, v^{-0.5} + C_{dl} \tag{S2}$$

Linear fit the plot and extrapolate the fitting line to y-axis gived the maximum electrical double layer capacitance (C_{dl}). Subtraction of C_{dl} from C_T got the maximum C_p.

Dunn Method

The Dunn method was applied to quantitatively differentiate the capacitance contribution from surface capacitive effects (herein we regard them as electrical double layer capacitance effects) and diffusion-controlled processes (herein we regard them as pseudocapacitance reactions) capacitive effects. At a fixed potential, the current density [$I_{(V)}$] read from a CV can be expressed as a combination of two terms as follow:

$$I_{(V)} = k_1 v + k_2 v^{0.5} \tag{S3}$$

Where the term $k_1 v$ accounted for the current density contributed from electrical double layer capacitance effects while the term $k_2 v^{0.5}$ was the current density associated with pseudocapacitance reactions. Dividing $v^{0.5}$ on both sides of the Equation S3 yields:

$$I_{(V)}/v^{0.5} = k_1 v^{0.5} + k_2 \tag{S4}$$

Therefore, Image of $I_{(V)}/v^{0.5}$ vs. $v^{0.5}$ was plotted by fetching the $I_{(V)}$ from CVs obtained under a series of scan rates, where a linear fitting line was obtained with slope equals k_1 and y-intercept of k_2. Plugging the obtained k_1 and k_2 into Equation S3 allowed one to differentiate the capacitance contribution from electrical double layer capacitance and pseudocapacitance at the specific potential V and a selected scan rate v. The capacitance ratio of pseudocapacitance to electrical double layer capacitance equals the ratio of the area of the purple or pink region to the area of the white region.

Repeating the aforementioned steps for other scan rates results in the pseudocapacitance and electrical double layer capacitance contributions.

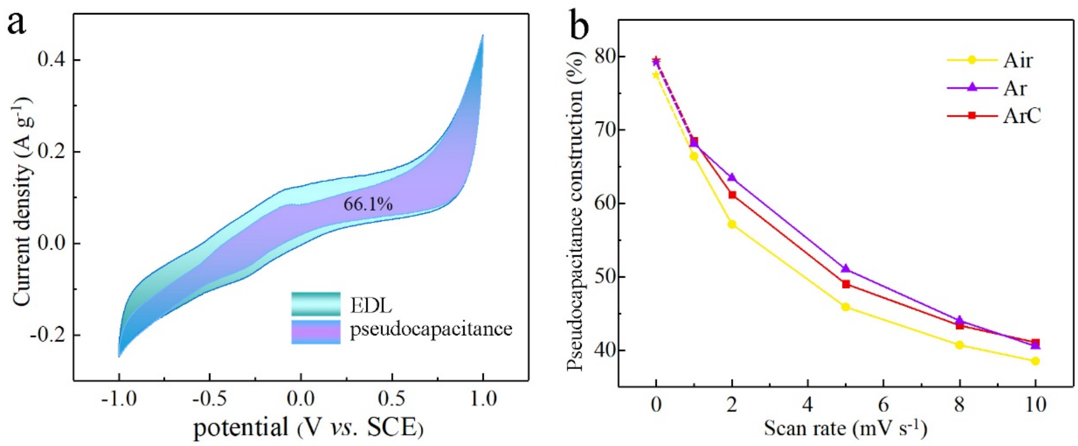

Figure S4 (a) CV curves and diffusion contributions of HTO samples at a scanning rate of 1 mV s^{-1}. (b) Diffusion contribution of three samples at different scanning rates.