

## Supporting Information

for

# Efficiency of Oxygen Evolution on Iridium Oxide Determined from the pH Dependence of Charge Accumulation

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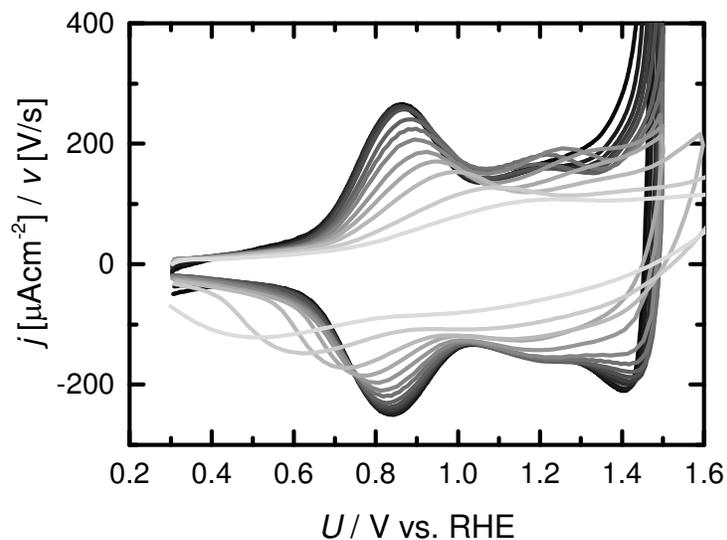
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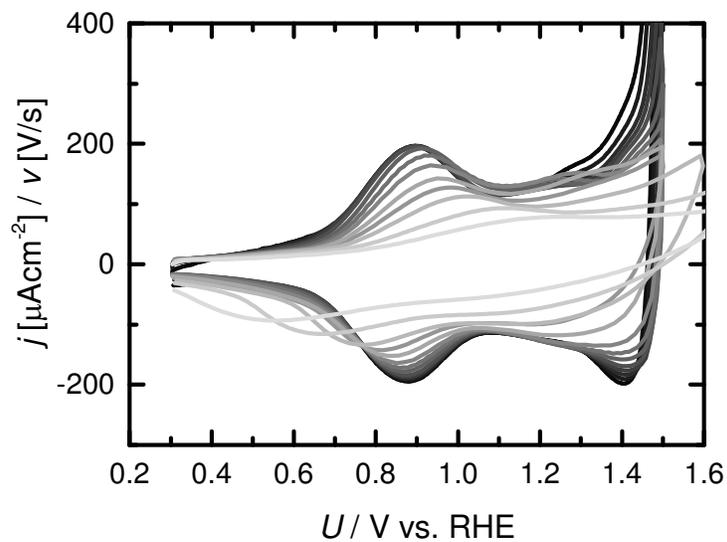
**Table S1. Temperature dependence of the pH and solution color during hydrolysis of  $K_2IrCl_6$**

Time /min	Temperature / °C	pH	Color
0	25.2	12.12	Brown
1	28.0	11.97	
2	31.2	11.89	
3	34.2	11.63	
4	37.0	11.76	
5	39.3	11.72	
6	42.5	11.66	
7	46.0	11.59	
8	49.1	11.53	Golden yellow
9	53.0	11.49	
10	54.0	11.41	
11	58.6	10.99	Lemon yellow
12	60.5	10.51	
13	64.0	10.02	Cloudy yellow
14	67.0	9.32	
15	70.0	8.81	Cloudy blue
16	16.0	9.50	Cloudy blue

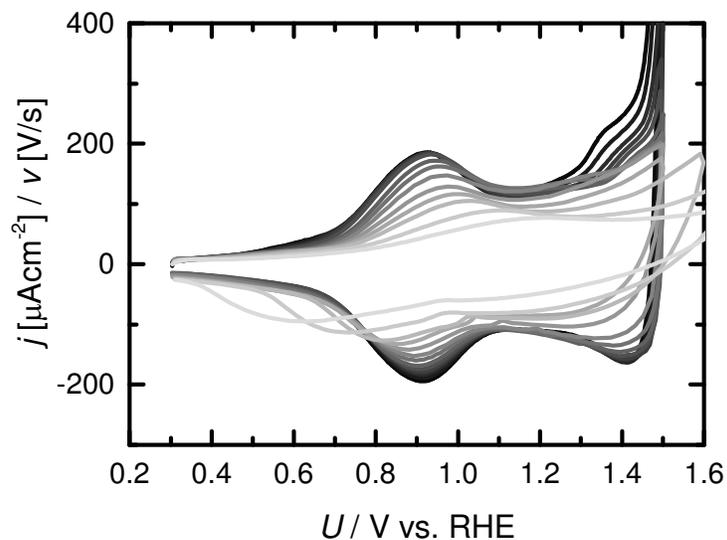
The solution was cooled in an ice bath immediately after the temperature reached 70.0 °C at 15 minutes.



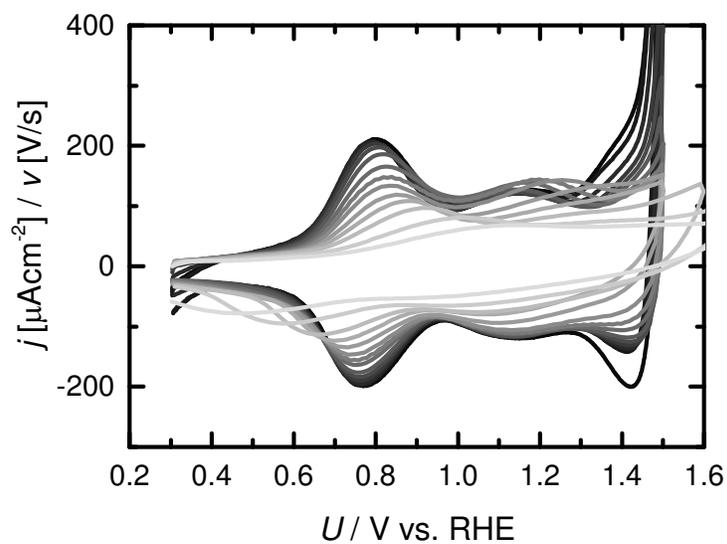
**Figure S1.** Cyclic voltammograms used for the trumpet plot analysis (pH 2) shown in a gradient from black (10 mV/s) to white (100 V/s). Current densities were normalized by the scan rate. Scan rates: 10, 20, 50, 100, 200, 500 [mV/s], 1, 2, 5, 10, 20, 50, 100 [V/s].



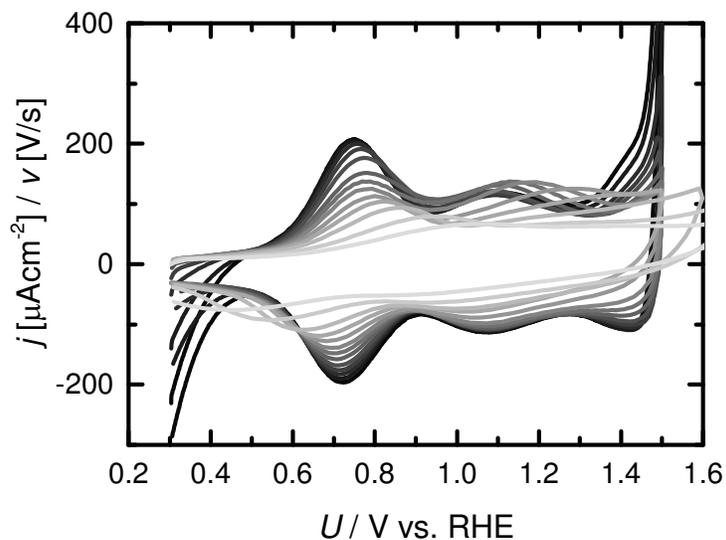
**Figure S2.** Cyclic voltammograms used for the trumpet plot analysis (pH 4) shown in a gradient from black (10 mV/s) to white (100 V/s). Current densities were normalized by the scan rate. Scan rates: 10, 20, 50, 100, 200, 500 [mV/s], 1, 2, 5, 10, 20, 50, 100 [V/s].



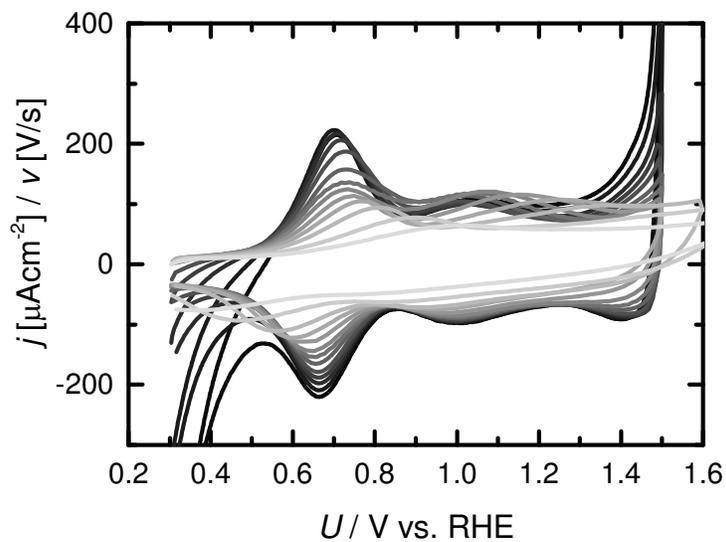
**Figure S3.** Cyclic voltammograms used for the trumpet plot analysis (pH 6) shown in a gradient from black (10 mV/s) to white (100 V/s). Current densities were normalized by the scan rate. Scan rates: 10, 20, 50, 100, 200, 500 [mV/s], 1, 2, 5, 10, 20, 50, 100 [V/s].



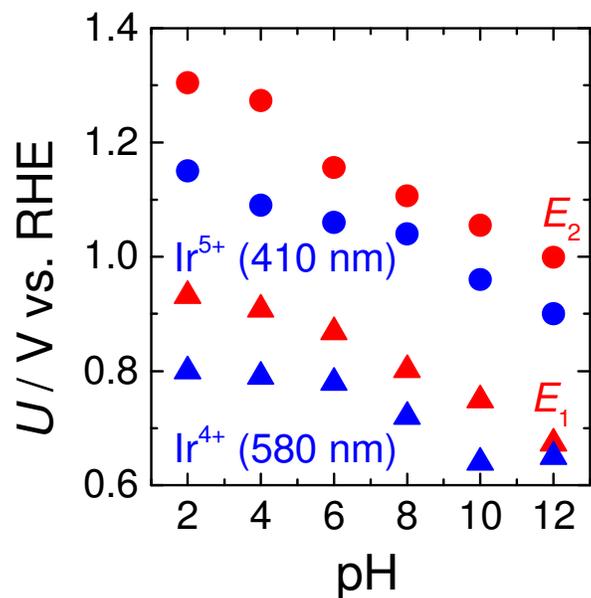
**Figure S4.** Cyclic voltammograms used for the trumpet plot analysis (pH 8) shown in a gradient from black (10 mV/s) to white (100 V/s). Current densities were normalized by the scan rate. Scan rates: 10, 20, 50, 100, 200, 500 [mV/s], 1, 2, 5, 10, 20, 50, 100 [V/s].



**Figure S5.** Cyclic voltammograms used for the trumpet plot analysis (pH 10) shown in a gradient from black (10 mV/s) to white (100 V/s). Current densities were normalized by the scan rate. Scan rates: 10, 20, 50, 100, 200, 500 [mV/s], 1, 2, 5, 10, 20, 50, 100 [V/s].



**Figure S6.** Cyclic voltammograms used for the trumpet plot analysis (pH 12) shown in a gradient from black (10 mV/s) to white (100 V/s). Current densities were normalized by the scan rate. Scan rates: 10, 20, 50, 100, 200, 500 [mV/s], 1, 2, 5, 10, 20, 50, 100 [V/s].



**Figure S7.** pH dependence of the formal potential of valence change ( $E_1$ ,  $E_2$ ) and the onset potential corresponding to  $Ir^{4+}$  (580 nm) and  $Ir^{5+}$  (410 nm) absorption. Red symbols: redox potentials, blue symbols: absorption. Triangles:  $Ir^{4+}$ , circles:  $Ir^{5+}$ . The onset potential was defined as the potential where the differential absorption at the specified wavelength exceeded 0.005. Differential absorption spectra were obtained by subtracting the spectra at the onset potential and that 100 mV negative. The difference in pH dependence with the onset of OER (Figure 2) indicates the iridium species generated at  $E_1$  and  $E_2$  are not responsible for initiating OER.