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## Supporting information

Silicon-doped Iridium electrode prepared by magnetron-sputtering toward advanced electrocatalyst for overall water splitting in acidic media

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Table S1<sup>†</sup> Preparation, composition analysis and loading capacity of Si-doped Ir electrodes with different proportions

Electrode	Ir : Si Sputtering power (W)	Ir : Si by ED-XRF (mol%:mol%)	Ir : Si by XPS (mol%:mol%)	Loading capacity (mg <sub>lr</sub> cm <sup>-2</sup> )
Si-3-doped Ir	50:40	96.71:3.29	70.30:29.70	0.120
Si-5-doped Ir	40:40	95.83:4.17	72.03:27.97	0.100
Si-10-doped Ir	30:40	91.04:8.96	62.80:37.20	0.075
Si-20-doped Ir	20:40	81.07:18.93	48.76:51.24	0.050
Si-30-doped Ir	10:40	69.35:30.65	38.00:62.00	0.025
Si-50-doped Ir	5:40	47.19:52.81	25.62:74.38	0.012

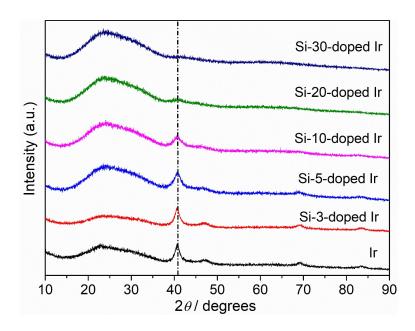


Fig. S1<sup>†</sup> X-ray diffraction curves of Si-doped Ir electrodes with different mole ratios.

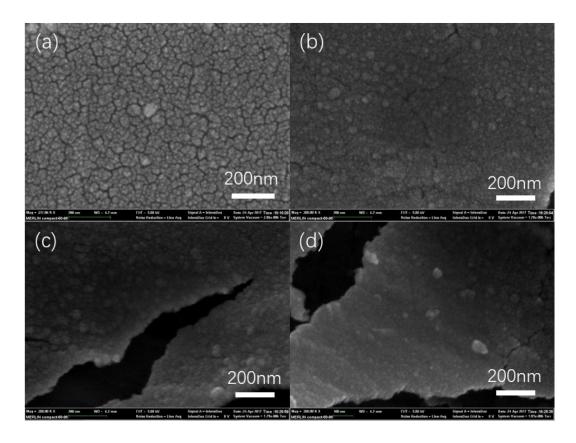


Fig. S2<sup>†</sup> SEM images of Ir (a, c) and Si-10-doped Ir (b, d) electrodes.

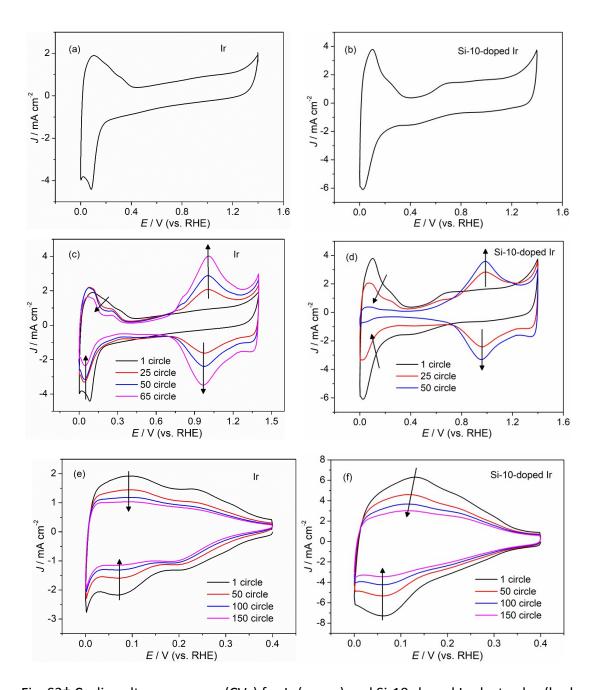


Fig. S3<sup>†</sup> Cyclic voltammograms (CVs) for Ir (a, c, e) and Si-10-doped Ir electrodes (b, d,

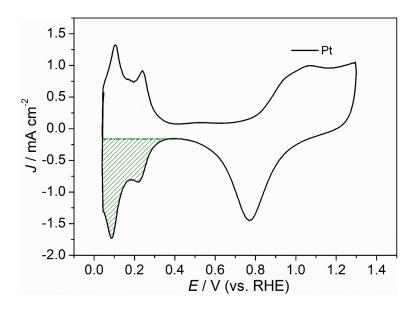


Fig. S4<sup>†</sup> Cyclic voltammogram for Pt electrode.

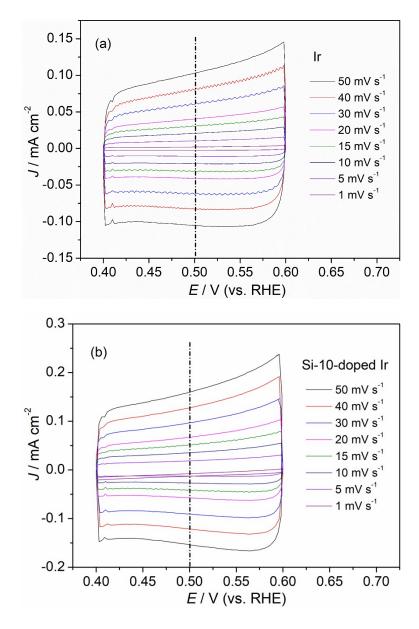


Fig. S5 $^{\dagger}$  CVs scanning with different scanning speeds at 1 - 50 mV s $^{-1}$  for Ir (a) and Si-10-doped Ir (b) electrodes.

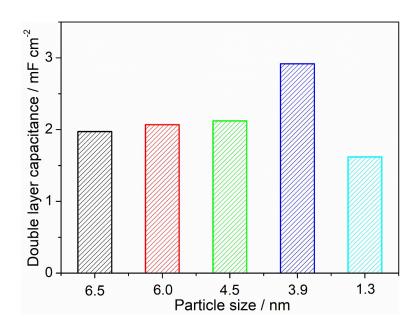


Fig. S6<sup>†</sup> Relationship between particle size and double layer capacitance.

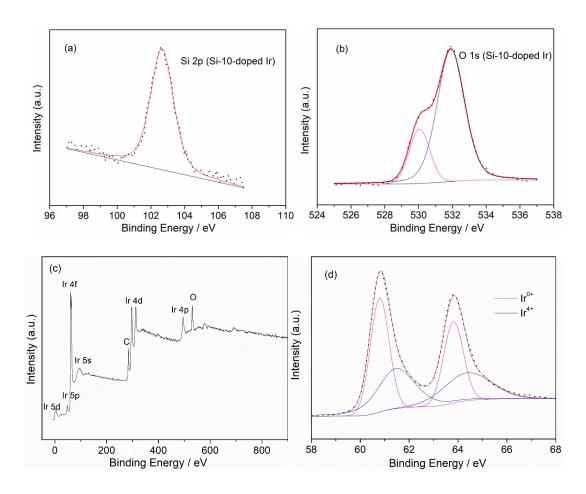


Fig. S7<sup>†</sup> XPS analysis of high-resolution spectrum of Si (a) and O (b) for Si-10-doped Ir electrodes. XPS analysis of Ir electrode for wide-range spectrum (c) and high-resolution spectrum (d).

Table S2<sup>†</sup> The Si 2p binding energy of Si-doped Ir electrodes with different proportions

Electrode	Binding energy of Si 2p (eV)	Negative shift of binding energy relative to that of Si <sup>4+</sup> (eV)
Si-3-doped Ir	102.5	0.5
Si-5-doped Ir	102.7	0.3
Si-10-doped Ir	102.6	0.4
Si-20-doped Ir	102.6	0.4
Si-30-doped Ir	102.7	0.3
Si-50-doped Ir	103.0	0
Si <sup>4+</sup>	103.0	-

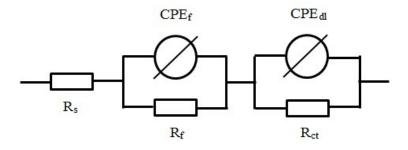


Fig. S8<sup>†</sup> The equivalent circuit of the impedance of Si-10-doped Ir and Ir electrodes

Table S3<sup>†</sup> Impedance parameters of Si-doped Ir and Ir electrodes obtained by fitting the experimental data to  $R_s(R_fQ_f)(R_{ct}Q_{dl})$  equivalent circuit

Electrocatalyst	$R_s$ $/\Omega$ cm $^2$	$ m R_f$ $/\Omega~cm^2$	C <sub>f</sub> /μF cm <sup>-2</sup>	$n_{f}$	$R_{ct}$ $/\Omega$ cm <sup>2</sup>	$C_{dl}$ / $\mu F~cm^{-2}$	n <sub>dl</sub>
lr	0.8337	0.2837	3.98	1.03	0.7702	192	0.5966
Si-10-doped Ir	0.8295	0.1931	3.50	1.08	0.5709	329	0.6588

Table S4<sup>†</sup> The content of Si-10-doped Ir after the accelerated linear potential sweeps over 1000 CV cycles and constant potential at  $\eta$  = 0.2V for 10000s

		The molar content / m	nol%
Element	Initial	After 1000	After 10000s at
	Initial	CV cycles	$\eta$ = 0.2 V
Ir	91.04	92.02	92.16
Si	8.96	7.98	7.84

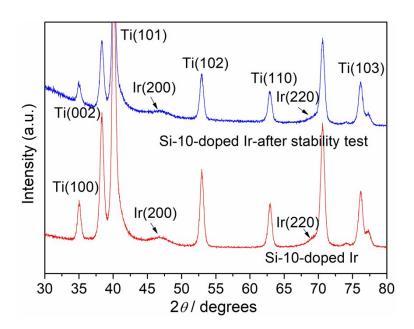


Fig. S9<sup>†</sup> X-ray diffraction curves of Si-10-doped Ir electrode before and after the long-term HER stability test at  $\eta$  = 0.2 V for 10000s.