## **Electronic Supplementary Material**



Fig. S1 Morphology and elemental distribution of annealed<sub>Ar</sub> PtNi<sub>3.5</sub> NPs.

(a) TEM image; (b) and (c) HR-TEM images; (d) HAADF-STEM image; (e) STEM EDX-mapping of Ni; (f) STEM EDX-mapping of Pt; (g) STEM EDX-mapping of composite Ni versus Pt.

**Table S1** Fitting results of XPS from the Ni 2p core level regions of as-prepared PtNi<sub>3.5</sub>, acid-treated Pt<sub>3</sub>Ni, the annealed<sub>5% H<sub>2</sub></sub> PtNi<sub>3.5</sub>, acid-treated annealed<sub>5% H<sub>2</sub></sub> Pt<sub>2.1</sub>Ni, and annealed<sub>5% H<sub>2</sub></sub> acid-treated Pt<sub>3</sub>Ni

Sample		$NI^{2+}_{1}/NI^{0}_{1}$			
	Ni <sup>0</sup>	Ni <sup>2+</sup>	Ni <sup>0</sup> , sat	Ni <sup>2+</sup> , sat	INI /INI
As-prepared PtNi <sub>3.5</sub> /C	852.41	855.87	857.32	860.61	2.0
Acid-treated Pt <sub>3</sub> Ni/C	853.17	856.54	859.53	863.39	1.1
Annealed <sub>5% H2</sub> PtNi <sub>3.5</sub> /C	852.93	855.67	857.75	861.09	1.8
Acid-treated annealed <sub>5% H2</sub> Pt <sub>2.1</sub> Ni/C	853.09	856.38	859.54	863.21	0.5
Annealed5%H2 acid-treated Pt3Ni/C	853.02	856.30	860.64	864.82	1.5

**Table S2** Fitting results of XPS from Pt 4f core level regions of as-prepared PtNi<sub>3.5</sub>, acid-treated Pt<sub>3</sub>Ni, annealed<sub>5% H<sub>2</sub></sub> PtNi<sub>3.5</sub>, acid-treated annealed<sub>5% H<sub>2</sub></sub> Pt<sub>2.1</sub>Ni, and annealed<sub>5% H<sub>2</sub></sub> acid-treated Pt<sub>3</sub>Ni

Sample	Pos			
	$Pt^0$	$Pt^{2+}$	Pt <sup>4+</sup>	$Pt/PtO_x$
As-prepared PtNi <sub>3.5</sub> /C	71.10	71.98	74.63	1.1
Acid-treated Pt <sub>3</sub> Ni/C	71.57	72.57	74.88	1.5
Annealed <sub>5% H2</sub> PtNi <sub>3.5</sub> /C	71.72	72.79	75.02	1.5
Acid-treated annealed <sub>5% H2</sub> Pt <sub>2.1</sub> Ni/C	71.71	72.79	75.04	1.5
Annealed $_{5\% H_2}$ acid-treated Pt <sub>3</sub> Ni/C	71.76	72.77	75.02	1.0