## Modulating Catalytic Activity and Durability of PtFe Alloy Catalysts for Oxygen Reduction Reaction Through Controlled Carbon Shell Formation

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**Figure S1.** TGA data to estimate the metal loading of (**a**) PtFe\_ASP, (**b**) PtFe 700\_Ar, (**c**) PtFe 700\_H<sub>2</sub>, (**d**) PtFe 900\_Ar, and (**e**) PtFe 900\_H<sub>2</sub> catalysts.



**Figure S2.** Low-resolution TEM images of (**a**) PtFe 700\_Ar, (**b**) PtFe 700\_H<sub>2</sub>, (**c**) PtFe 900\_Ar and (**d**) PtFe 900\_H<sub>2</sub>. Magnified TEM images of (**e**) PtFe 700\_Ar, (**f**) PtFe 700\_H<sub>2</sub>, (**g**) PtFe 900\_Ar and (**h**) PtFe 900\_H<sub>2</sub>.



**Figure S3.** TEM images and particle size distribution of (**a**) commercial Pt/C, (**b**) PtFe\_ASP, (**c**) PtFe 700\_Ar, (**d**) PtFe 700\_H2, (**e**) PtFe 900\_Ar, and (**f**) PtFe 900\_H2.



**Figure S4.** N 1s core level XPS spectra of (**a**) PtFe\_ASP, (**b**) PtFe 700 Ar, (**c**) PtFe 700 H<sub>2</sub>, (**d**) PtFe 900 Ar, and (**e**) PtFe 900 H<sub>2</sub>.

**Table S1.** Comparison of ECSA values of commercial Pt/C and different heat-treated PtFe samples (before and after 5000 potential cycles).

	Before ADT	After ADT
Pt/C	94.18	38.63
PtFe 700_Ar	22.45	-
PtFe 900_Ar	13.72	-
<b>PtFe 700_H</b> <sub>2</sub>	34.49	32.96
<b>PtFe 900_H</b> 2	22.69	23.05



**Figure S5.** TEM image and elemental maps of Pt, Fe, and C for (**a**) PtFe\_ASP, (**b**) PtFe 700\_H<sub>2</sub>, and (**c**) PtFe 900\_H<sub>2</sub>. The corresponding EDX spectra of (**d**) PtFe\_ASP, (**e**) PtFe 700\_H<sub>2</sub>, and (**f**) PtFe 900\_H<sub>2</sub>, repectively.

Energy-dispersive X-ray spectrocopy (EDX) analysis were performed to identify elemental distribution of PtFe\_ASP and different heat-treated PtFe catalysts. The elemental map of PtFe alloy catalyst revealed uniform distribution of elements, Pt and Fe. Furthermore, in the corresponding EDX spectra, it was confirmed that the prepared PtFe nanoparticles have near equivalent atomic ratios of Pt to Fe (Pt:Fe = 1:1).



**Figure S6.** TEM image and elemental maps of Pt, Fe, and C for (**a**) PtFe 700\_H<sub>2</sub> and (**b**) PtFe 900\_H<sub>2</sub> after the ADTs. The corresponding EDX spectra of (**c**) PtFe 700\_H<sub>2</sub> and (**d**) PtFe 900\_H<sub>2</sub>, respectively.

After the ADTs of PtFe 700\_H<sub>2</sub> and PtFe 900\_H<sub>2</sub> catalysts, their EDX mapping images showed no obvious change in the density of elemental distribution (Figure S6a and b). Notably, the corresponding EDX spectra (Figure S6c and d) revealed that the atomic ratios of Pt:Fe (1:1) were hardly changed even after the ADTs, indicating no significant loss of Pt or Fe atoms by dissolution.