

## Supplementary Material

### Scalable Synthesis of Ternary Nanocatalysts for High Efficiency Electrooxidation Catalysis by Microfluidics

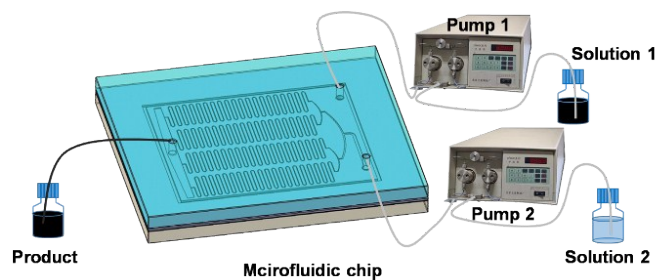
Yingyan Zhou,<sup>a</sup> Dumei Wang,<sup>a</sup> Xueming Kang,<sup>a</sup> Dongtang Zhang,<sup>\*a</sup> Xiangnan Dou,<sup>a</sup> Xiayan Wang,<sup>a</sup> Guangsheng Guo<sup>ab</sup>

<sup>a</sup> Center of Excellence for Environmental Safety and Biological Effects, Beijing Key Laboratory for Green Catalysis and Separation, Department of Chemistry and Chemical Engineering, Beijing University of Technology, Beijing 100124, P. R. China

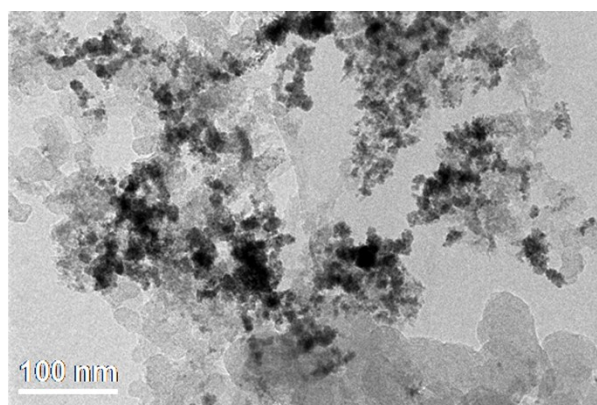
<sup>b</sup> Beijing Academy of Science and Technology, Beijing 100089, P. R. China

\*e-mail: [zhangdongtang@bjut.edu.cn](mailto:zhangdongtang@bjut.edu.cn)

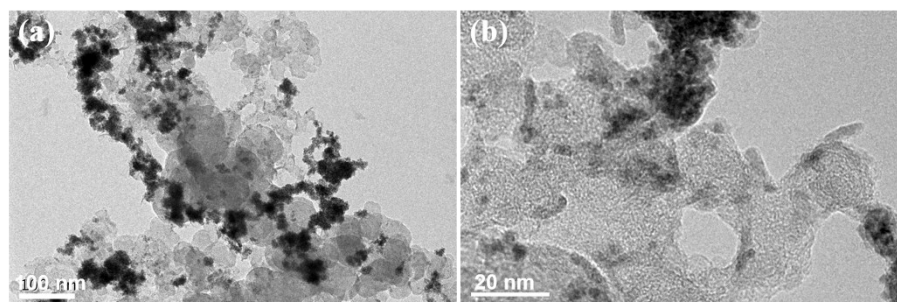
## Supplementary Figures



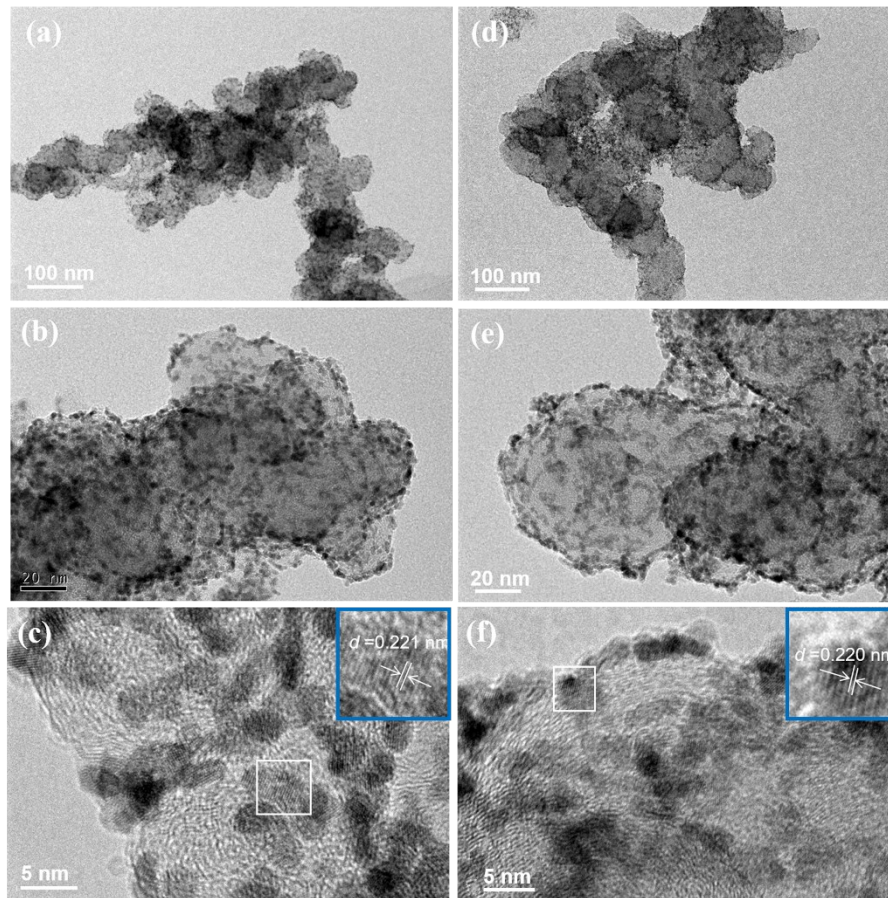
**Fig. S1.** The schematic illustration of microfluidic synthesis system.



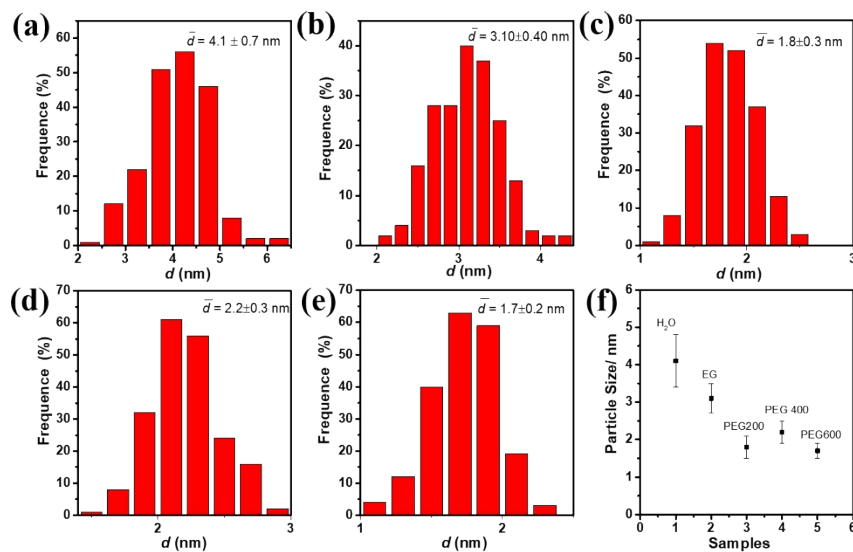
**Fig. S2.** TEM image of PtFeCu/C-H<sub>2</sub>O with the pump flow rate of 60 mL h<sup>-1</sup>.



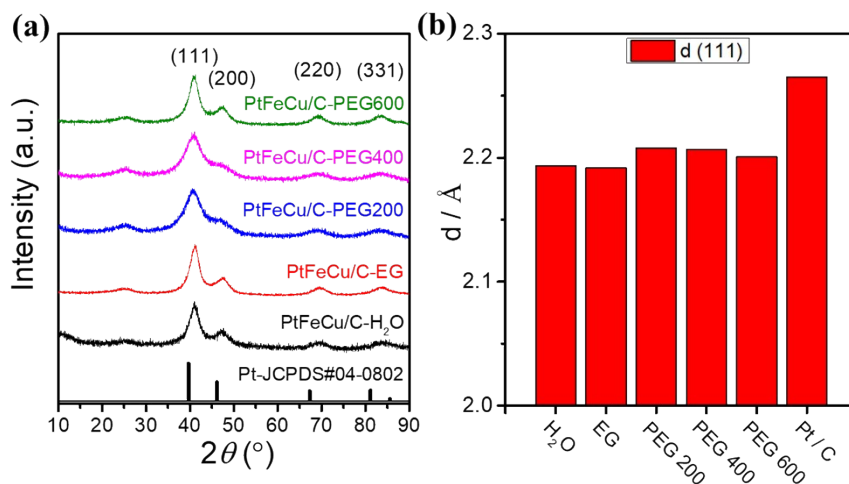
**Fig. S3.** Variously magnified TEM images of PtFeCu/C-EG with the pump flow rate of 60 mL h<sup>-1</sup>.



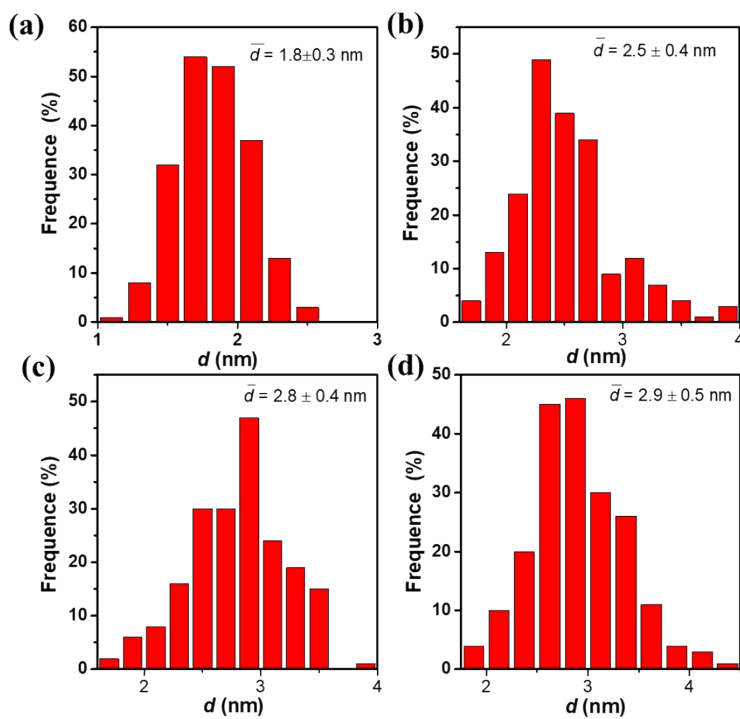
**Fig. S4.** Various magnified TEM images of PtFeCu/C prepared in PEG series solvents using flow rate of 60 mL h<sup>-1</sup>. (a-c) PEG400, (d-f) PEG600.



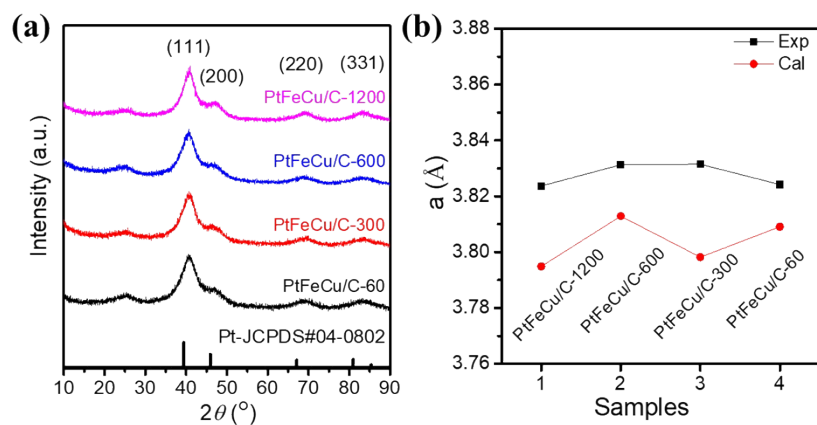
**Fig. S5.** Size histograms of the PtFeCu/C synthesized at a pump flow rate of 60 ml h<sup>-1</sup> in different solvents respectively. (a) H<sub>2</sub>O, (b) EG, (c) PEG200, (d) PEG400, (e) PEG600, (f) Particle size of different samples obtained in the above solvents.



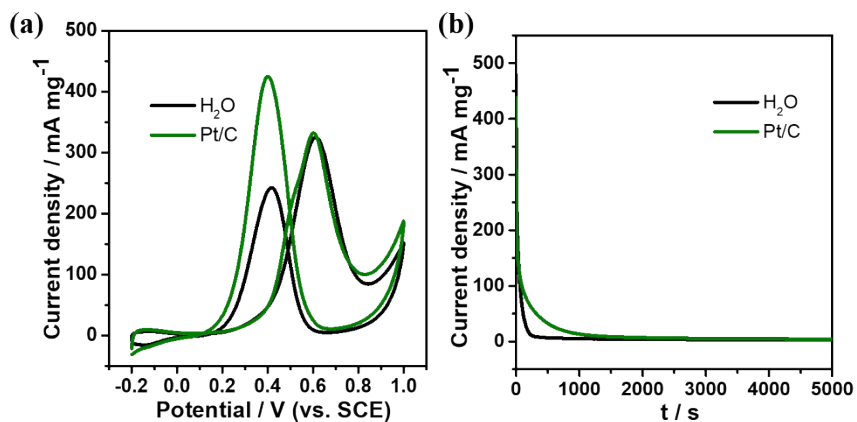
**Fig. S6.** (a) The XRD patterns of the PtFeCu/C synthesized in H<sub>2</sub>O, EG, PEG200, PEG400, and PEG600. (b) The distance of (111) plane for PtFeCu/C synthesized in different solvents.



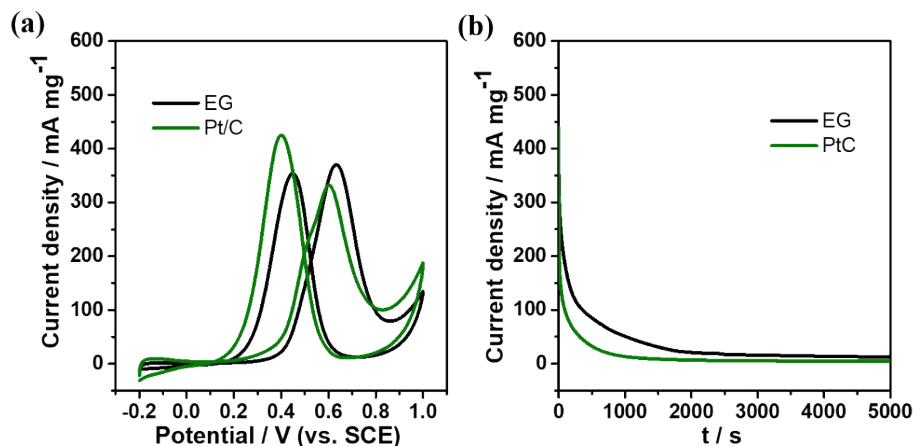
**Fig. S7.** Size histograms of the PtFeCu/C synthesized at varying pump flow speed. (a) 60 mL h<sup>-1</sup>, (b) 300 mL h<sup>-1</sup>, (c) 600 mL h<sup>-1</sup>, (d) 1200 mL h<sup>-1</sup>.



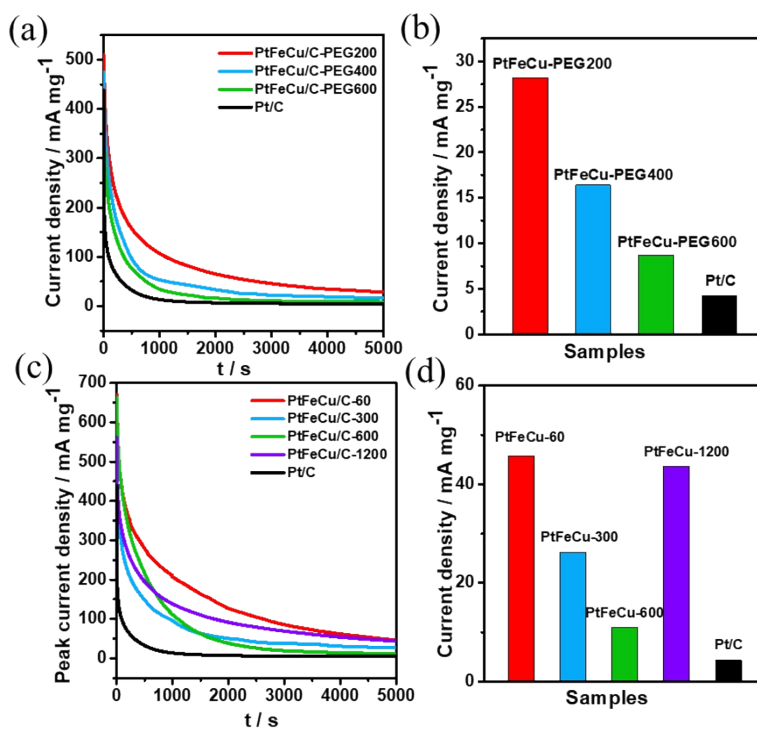
**Fig. S8.** (a) The XRD patterns of the PtFeCu/C synthesized in PEG200 with different pump flow speeds. (b) Calculated and experimental lattice parameters of samples obtained in varying flow rates.



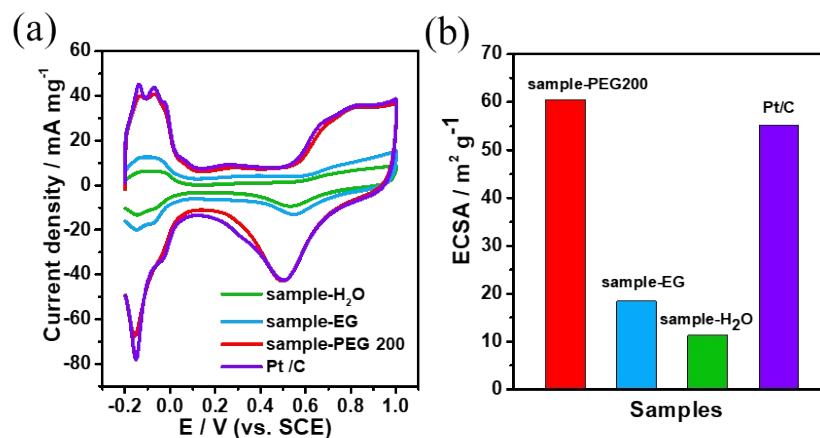
**Fig. S9.** Electrocatalytic properties of the PtFeCu/C synthesized in water solvent for methanol oxidation reaction (MOR). (a) Cyclic voltammograms of the PtFeCu/C obtained in water and commercial Pt/C in a  $N_2$ -saturated 0.5 mol/L  $H_2SO_4$  + 0.5 mol/L  $CH_3OH$  solution. Scanning rate is 50 mV/s. The currents were normalized by the mass on the GC surface. (b) Current-time curves of the catalyst and Pt/C recorded for 5000 s at 0.6V.



**Fig. S10.** Electrocatalytic properties of the PtFeCu/C synthesized in EG solvent for MOR. (a) Cyclic voltammograms of PtFeCu/C synthesized in EG and commercial Pt/C in a  $N_2$ -saturated 0.5 mol/L  $H_2SO_4$  + 0.5 mol/L  $CH_3OH$  solution. Scanning rate is 50 mV/s. The currents were normalized by the mass on the GC surface. (b) Current-time curves of the catalyst and Pt/C recorded for 5000 s at 0.6 V.



**Fig. S11.** (a) Chronoamperometric curves measured at 0.6 V for PtFeCu/C synthesized using PEG 200, 400 and 600, respectively, (b) Comparison of MOR current density at 5000 s in (a), (c) Chronoamperometric curves measured at 0.6 V for PtFeCu/C synthesized using PEG 200 at various flow rates, (d) Comparison of MOR current density at 5000 s in (c).



**Fig. S12.** Cyclic voltammetry curves and ECSA comparison of the PtFeCu/C obtained in different solvents and commercial Pt/C for MOR. (a) Cyclic voltammetry curves of the PtFeCu/C obtained in different solvents and commercial Pt/C in a N<sub>2</sub>-saturated 0.5 mol/L H<sub>2</sub>SO<sub>4</sub> solution. Scanning rate is 50 mV/s. The currents were normalized by the mass on the GC surface, (b) Comparison of ECSA of the catalysts prepared in different solvents and Pt/C.

## Supplementary Table

**Table S1.** The distance of (111) plane, lattice constant (a), diameter and full width at half maximum (FWHM) for PtFeCu/XC-72 synthesized in H<sub>2</sub>O, EG, PEG solvent series.

Samples	2 $\theta$	$d(111)/\text{\AA}$	$a/\text{\AA}$	D/nm	FWHM
PtFeCu/C-H <sub>2</sub> O	41.113	2.1937	3.7996	2.08	0.705
PtFeCu/C-EG	41.153	2.1917	3.7961	1.55	0.947
PtFeCu/C-PEG200	40.838	2.2079	3.8242	1.47	0.998
PtFeCu/C-PEG400	40.857	2.2068	3.8223	1.48	0.991
PtFeCu/C-PEG600	40.977	2.2007	3.8117	1.62	0.902
PDF#04-0802	39.76	2.265	3.9231	-	-

**Table S2.** Pt loading, atomic ratio, ICP-AES composition and lattice constant calculated according to Vegard's law of PtFeCu/C in varying solvents.

Samples	Pt Loading	Atomic ratio (Pt: Cu: Fe)	ICP Composition	a/Å
PtFeCu/C-H <sub>2</sub> O	25.76%	40.32%: 33.47%: 26.21%	Pt <sub>40</sub> Cu <sub>26</sub> Fe <sub>34</sub>	3.7329
PtFeCu/C-EG	29.94%	58.01%: 25.70%: 16.29%	Pt <sub>58</sub> Cu <sub>26</sub> Fe <sub>16</sub>	3.7898
PtFeCu/C-PEG 200	30.66%	57.50%: 20.80%: 21.70%	Pt <sub>57</sub> Cu <sub>21</sub> Fe <sub>22</sub>	3.7869
PtFeCu/C-PEG 400	33.11%	56.82%: 22.53%: 20.65%	Pt <sub>57</sub> Cu <sub>23</sub> Fe <sub>21</sub>	3.7851
PtFeCu/C-PEG 600	26.11%	54.06%: 24.39%: 21.55%	Pt <sub>54</sub> Cu <sub>24</sub> Fe <sub>22</sub>	3.7764

**Table S3.** The bulk composition and surface composition of PtFeCu/C obtained from PEG 200, EG and H<sub>2</sub>O.

Samples	The bulk atom percent of PtFeCu			The surface atom percent of PtFeCu		
	Pt/%	Fe/%	Cu/%	Pt/%	Fe/%	Cu/%
PtFeCu/C-PEG 200	53	18	29	53	26	21
PtFeCu/C-EG	59	11	30	51	31	18
PtFeCu/C-H <sub>2</sub> O	40	26	34	29	56	15



**Table S4.** XPS analysis result of Pt 4f, Cu 2p, Fe 2p spectra in PtFeCu/C-PEG200, PtFeCu/C-EG and PtFeCu/C-H<sub>2</sub>O.

Samples	Species	Binding energy	Assignment	Atomic ratio (%)
PtFeCu/C-PEG 200	Pt 4f <sub>7/2</sub>	71.35	Pt	41.7
		71.33	PtO	45.5
		70.25	PtO <sub>2</sub>	12.8
	Cu 2p <sub>3/2</sub>	933.26	Cu	58.3
		933.27	CuO <sub>x</sub>	41.7
	Fe 2p <sub>3/2</sub>	712.10	Fe	42.3
		720.06	FeO <sub>x</sub>	57.7
PtFeCu/C-EG	Pt 4f <sub>7/2</sub>	71.33	Pt	55.2
		72.05	PtO	44.8
	Cu 2p <sub>3/2</sub>	931.29	Cu	57.5
		933.39	CuO <sub>x</sub>	42.5
	Fe 2p <sub>3/2</sub>	711.83	Fe	56.7
		718.07	FeO <sub>x</sub>	43.3
PtFeCu/C-H <sub>2</sub> O	Pt 4f <sub>7/2</sub>	70.25	Pt	52.5
		72	PtO	28.5
		74.38	PtO <sub>2</sub>	19.0
	Cu 2p <sub>3/2</sub>	932.2	Cu	55.5
		933.43	CuO <sub>x</sub>	45.5
	Fe 2p <sub>3/2</sub>	711.82	Fe	54.5
		715.90	FeO <sub>x</sub>	45.5

**Table S5.** Pt loading, atomic ratio and ICP-AES composition results for PtFeCu/C in varying pump flow speed.

Pump speed (mL h <sup>-1</sup> )	Pt Loading	Atomic Ratio (Pt: Cu: Fe)	ICP Composition	a (Å)
1200	35.23 %	59.72%: 22.38%: 17.90%	Pt <sub>60</sub> Cu <sub>22</sub> Fe <sub>18</sub>	3.7947
600	33.87 %	65.08%: 24.10%: 10.82%	Pt <sub>65</sub> Cu <sub>24</sub> Fe <sub>11</sub>	3.8129
300	33.16 %	60.77%: 22.31%: 16.92%	Pt <sub>61</sub> Cu <sub>22</sub> Fe <sub>17</sub>	3.7982
60	33.20 %	64.02%: 23.06%: 12.92%	Pt <sub>64</sub> Cu <sub>23</sub> Fe <sub>13</sub>	3.8091

**Table S6.** The distance of (111) plane, lattice constant, diameter and full width at half maximum (FWHM) for PtFeCu/C synthesized in varying pump flow rate.

Samples	$2\theta$	$\theta$	$d(111)/\text{\AA}$	a	D/nm	FWHM
PtFeCu/C-1200	40.842	20.421	2.2076	3.8237	2.3559	0.621
PtFeCu/C-600	40.757	20.3785	2.2120	3.8313	1.6397	0.892
PtFeCu/C-300	40.757	20.3785	2.2121	3.8315	1.5985	0.915
PtFeCu/C-60	40.838	20.419	2.2079	3.8242	1.4660	0.998
Pt/C	39.76	19.88	2.265	3.9231	2.2685	–