Supporting Information

Insight into the Ordering Process and Ethanol Oxidation Performance of Au-Pt-Cu Ternary Alloys

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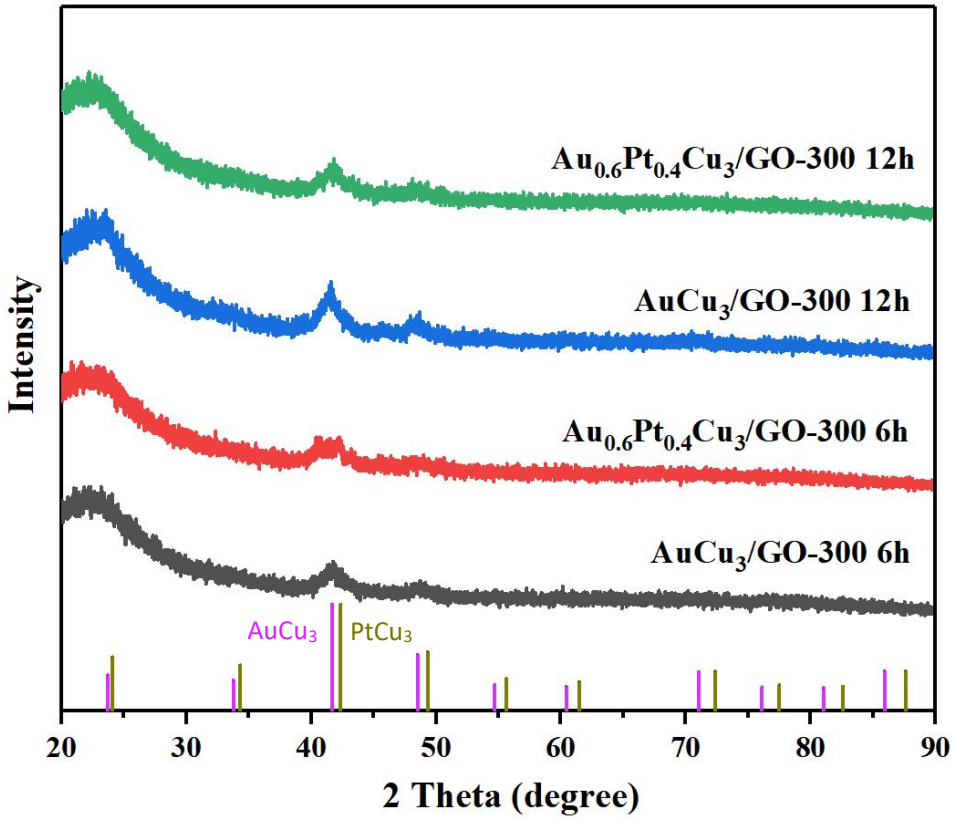


Fig. S1 The synthesis of $Au_xPt_{1-x}Cu_3$/rGO under 300 degrees.

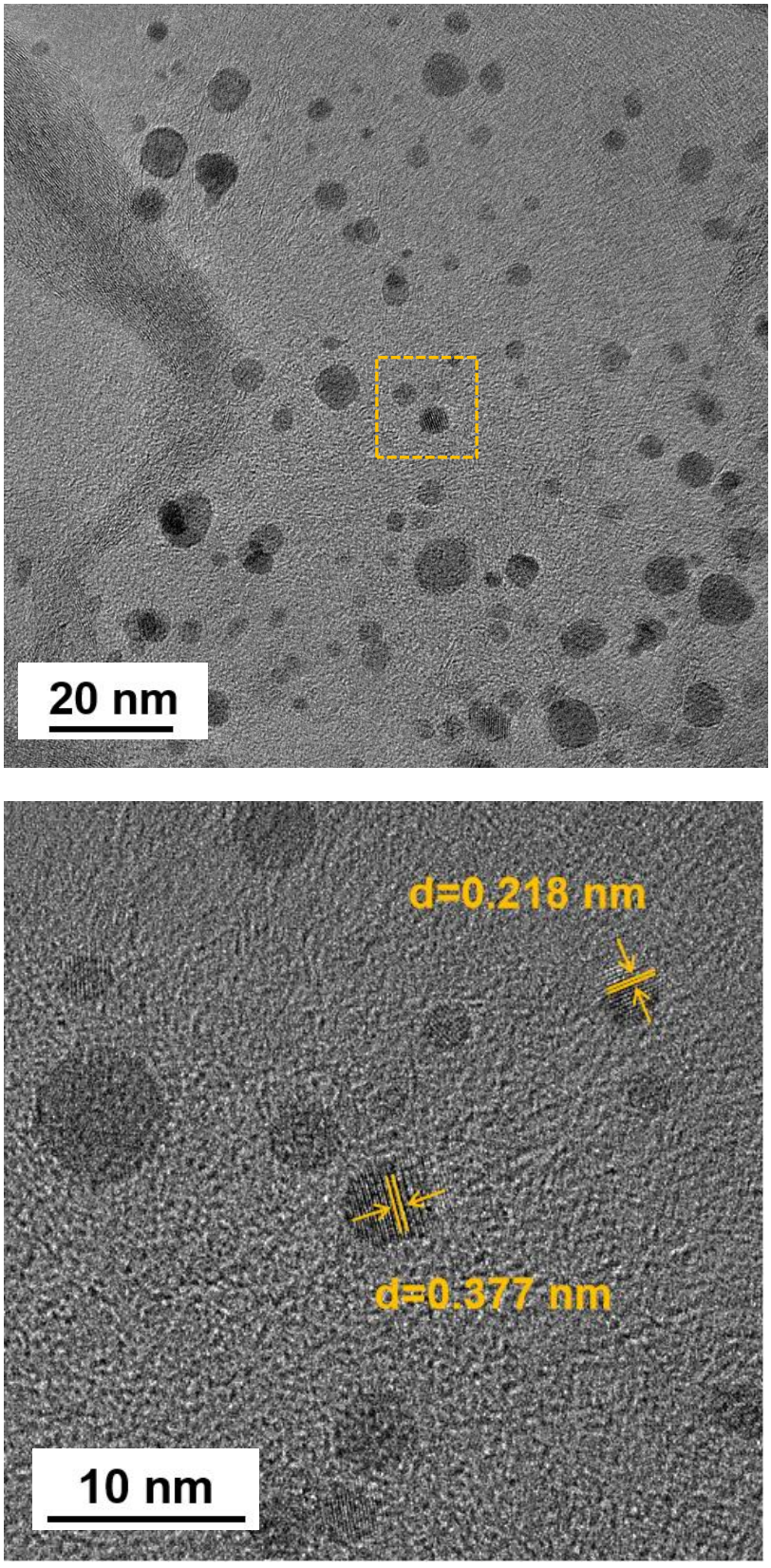
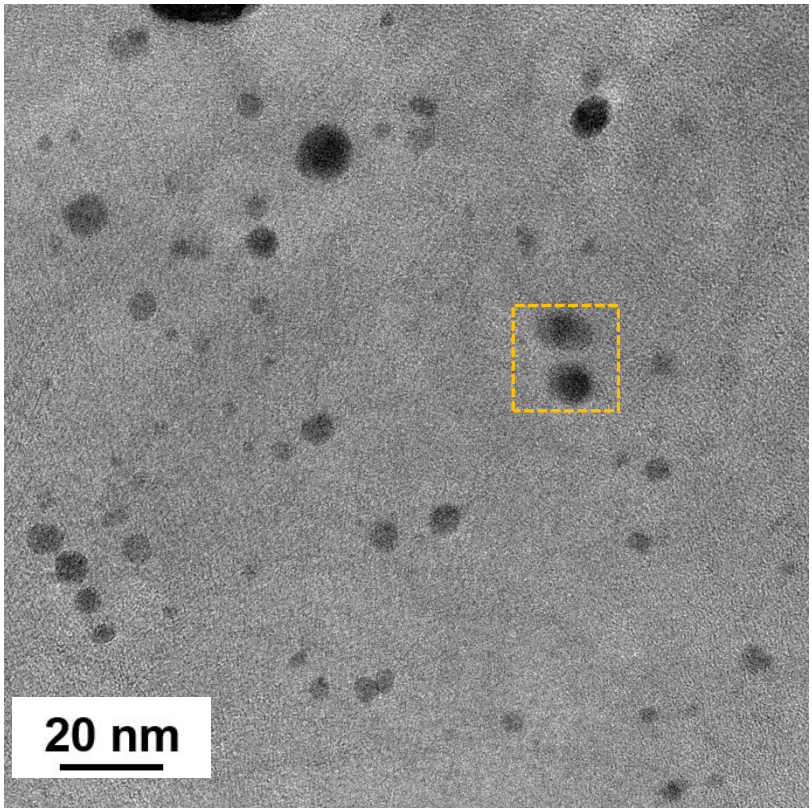


Fig. S2 High-resolution transmission electron microscopy images of $Au_{0.2}Pt_{0.8}Cu_3$/rGO.



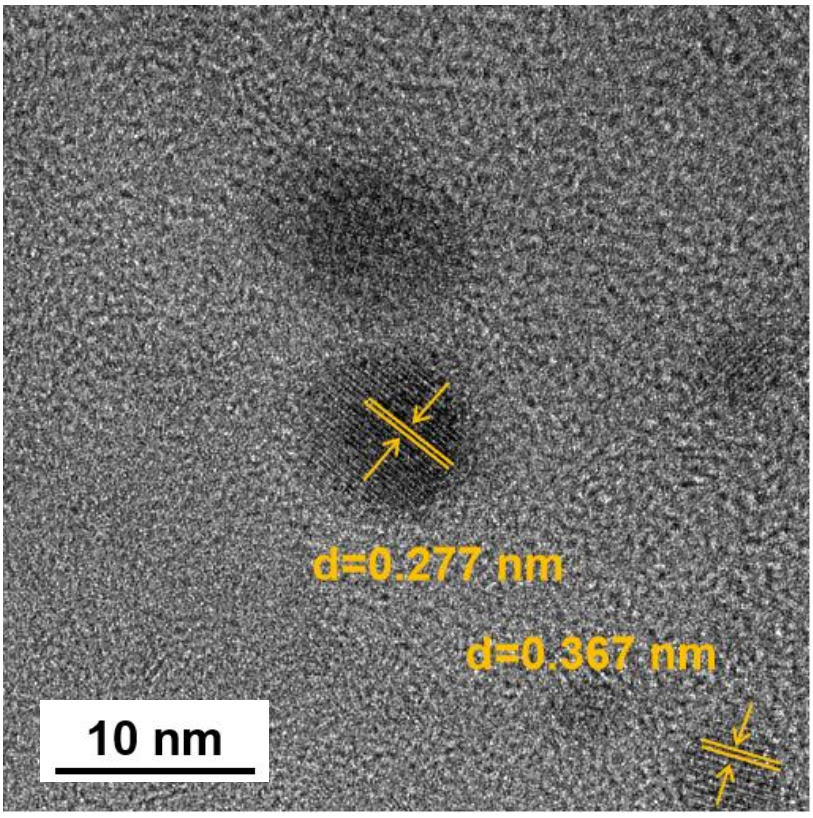


Fig. S3 High-resolution transmission electron microscopy images of $Au_{0.6}Pt_{0.4}Cu_3$/rGO.

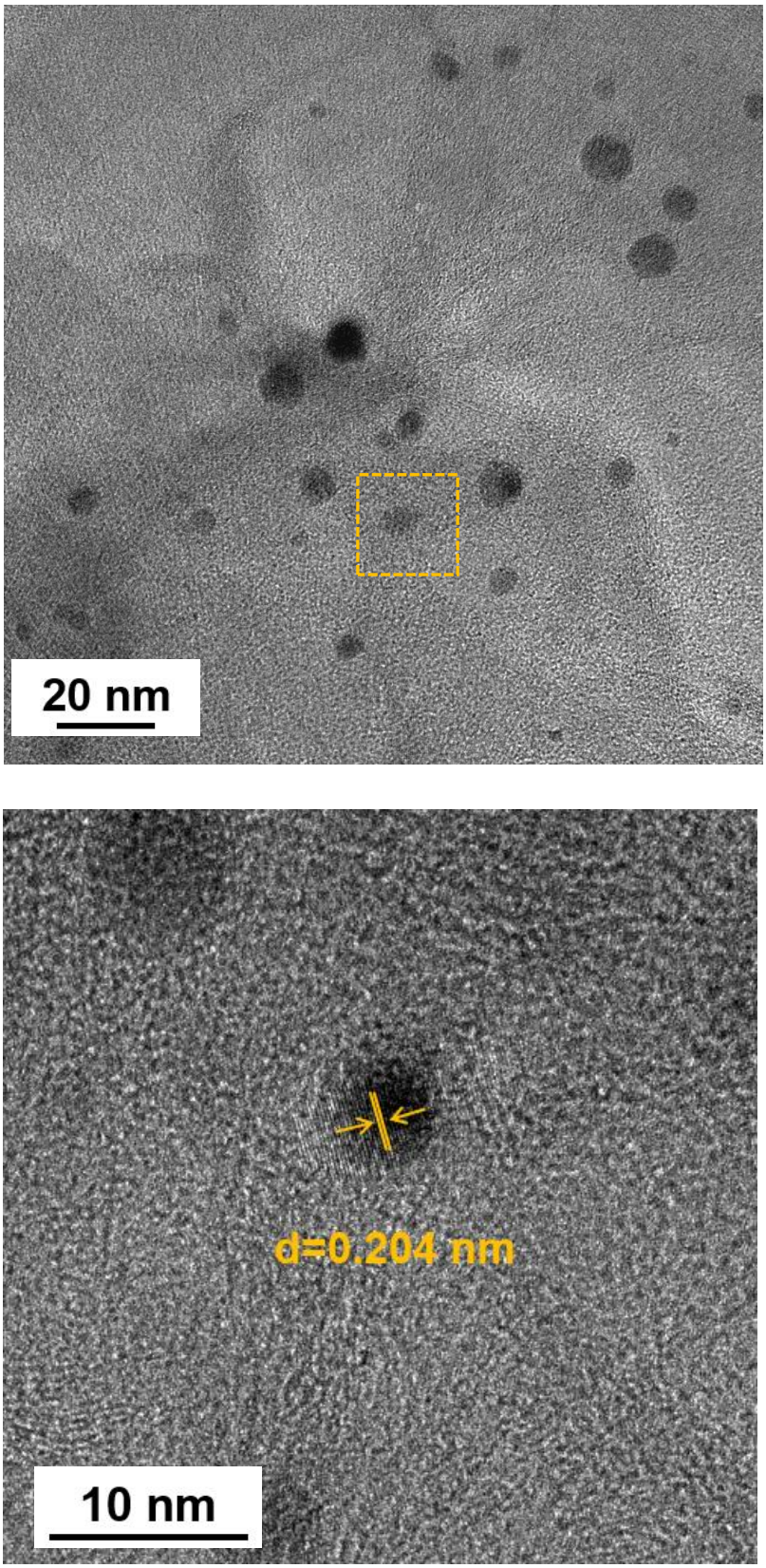


Fig. S4 High-resolution transmission electron microscopy images of $Au_{0.8}Pt_{0.2}Cu_3$/rGO.

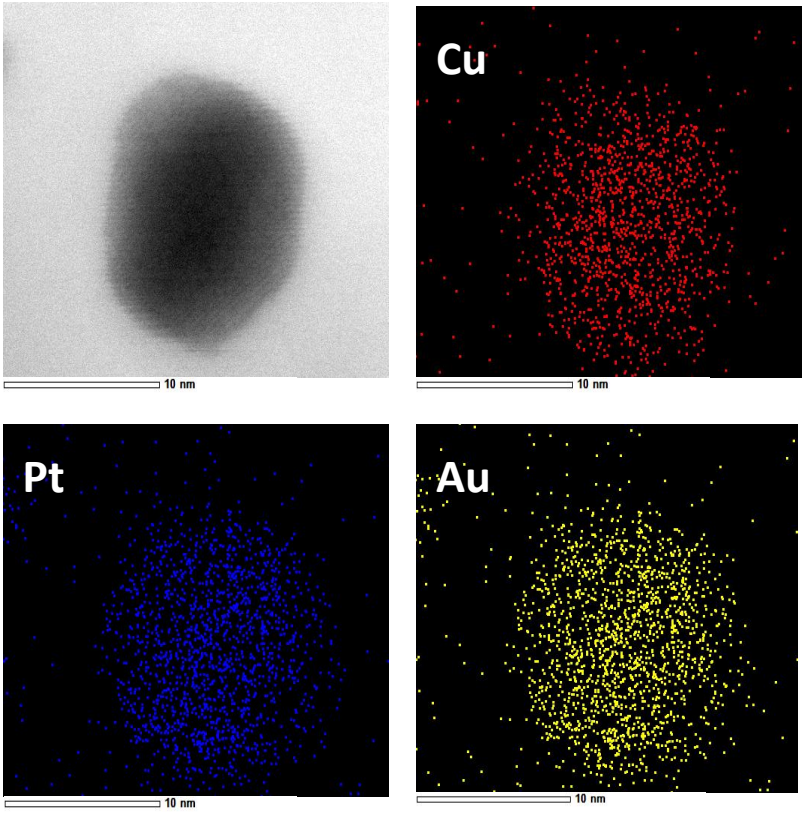


Fig. S5 Energy Dispersive Spectroscopy (EDS) mapping of $Au_{0.2}Pt_{0.8}Cu_3$/rGO.

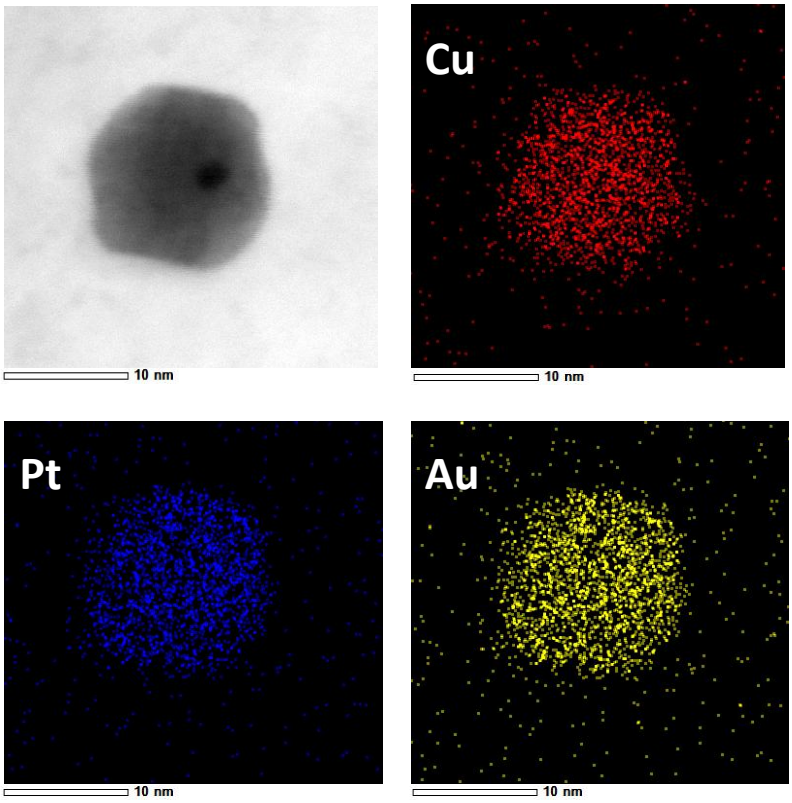


Fig. S6 Energy Dispersive Spectroscopy (EDS) mapping of $Au_{0.6}Pt_{0.4}Cu_3$/rGO.

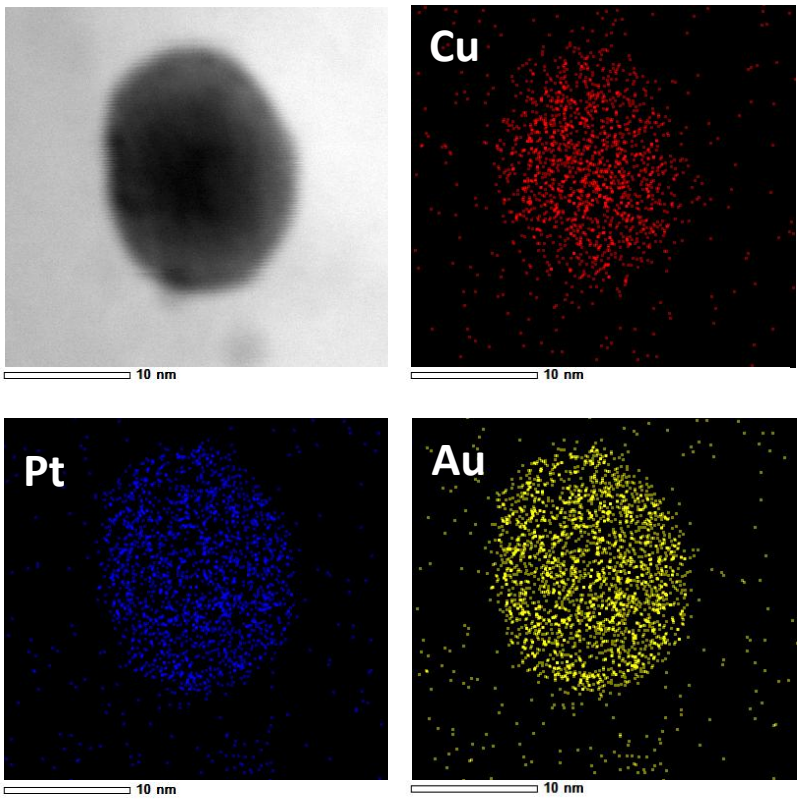


Fig. S7 Energy Dispersive Spectroscopy (EDS) mapping of $Au_{0.8}Pt_{0.2}Cu_3$/rGO.

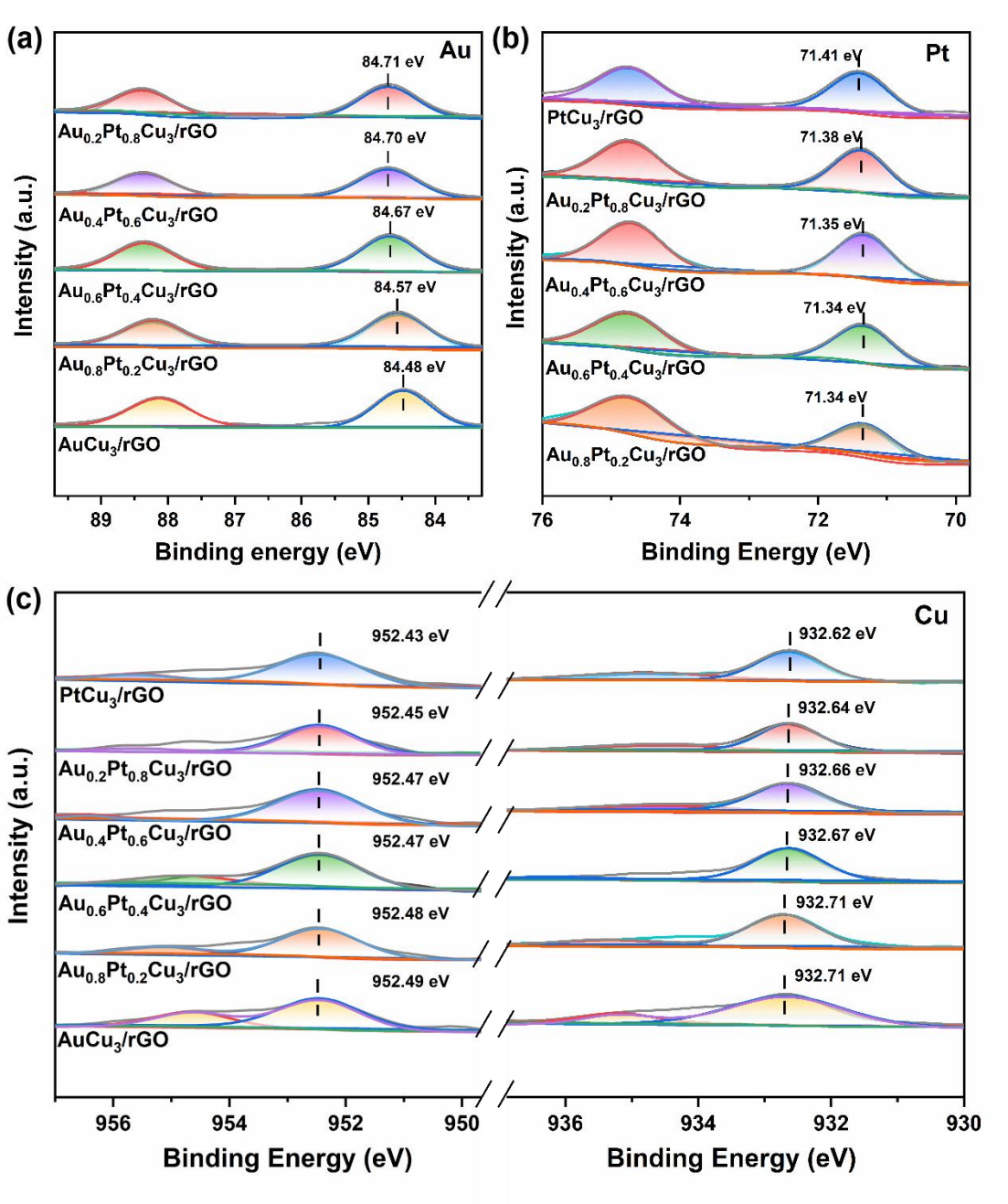


Fig. S8 X-ray photoelectron spectroscopy patterns of (a) Au 4f, (b) Pt 4f, (c) Cu $2p_{1/2}$ and Cu $2p_{3/2}$ in $Au_xPt_{1-x}Cu_3$/rGO ordered alloys.

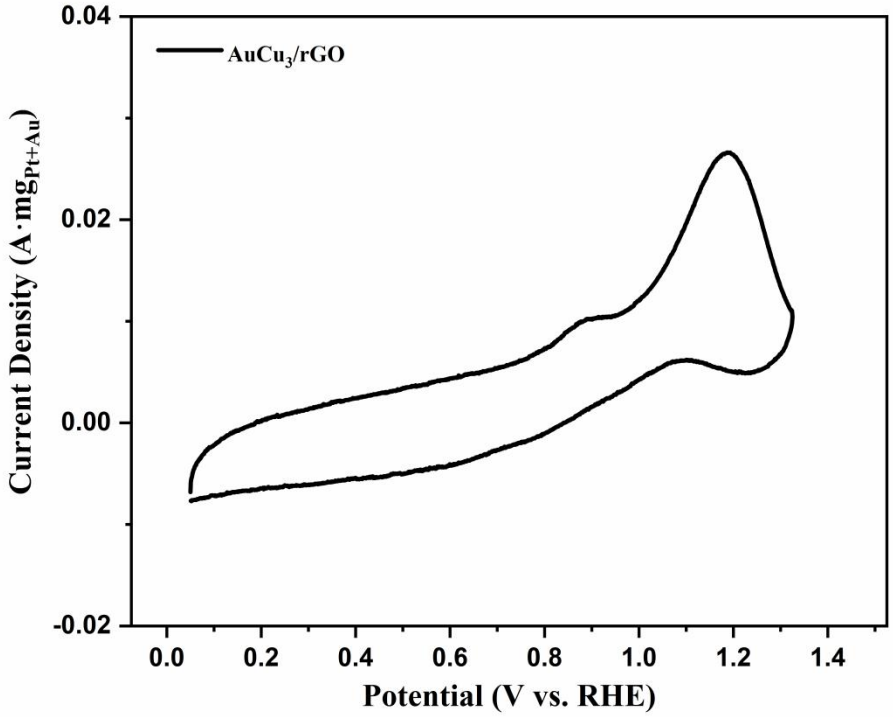


Fig. S9 Ethanol oxidation reaction in 1.0 M KOH and 1.0M EtOH with $AuCu_3$/rGO.

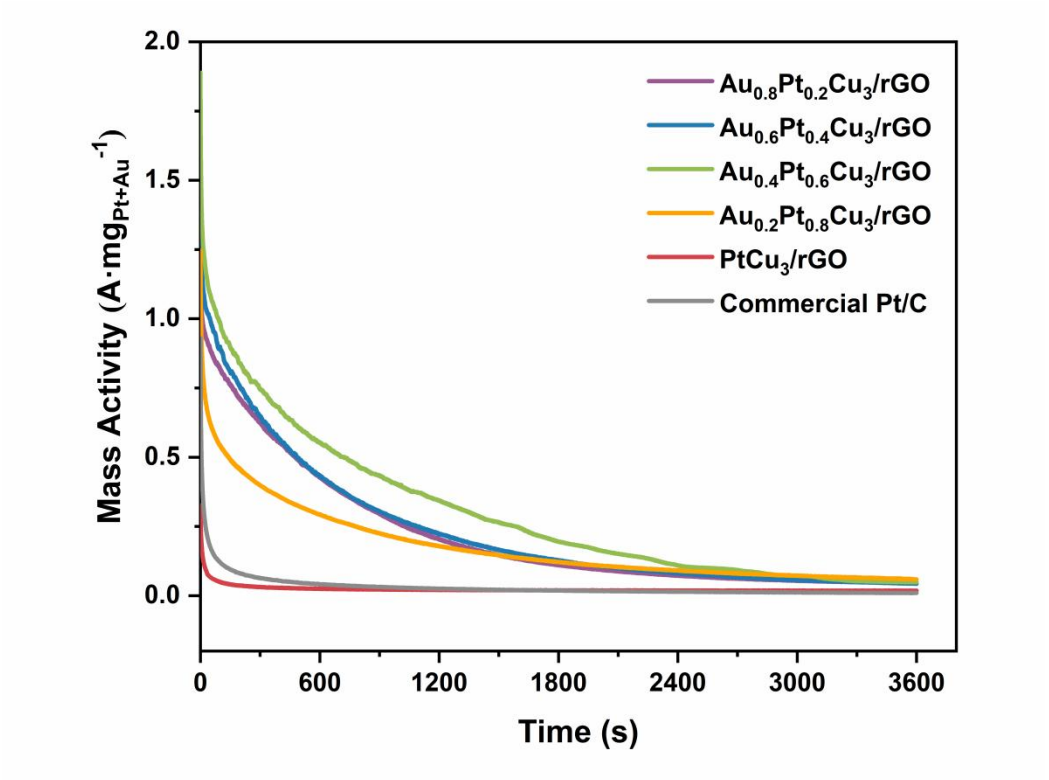


Fig. S10 Chronoamperometry testing at 0.77 V_{RHE} for 3600 seconds in N_2-saturated 1.0 M KOH and 1.0 M EtOH mixture solution.

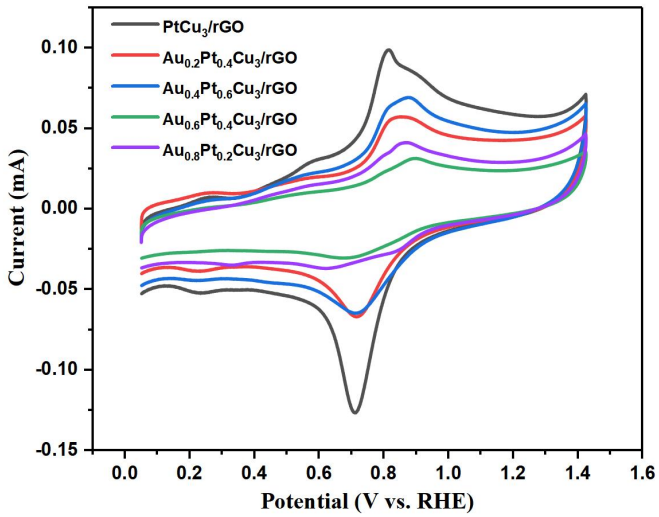


Fig. S11 Cyclic Voltammetry of $Au_xPt_{1-x}Cu_3$/rGO in 1.0M KOH with a scan rate of 50 mV • s^{-1}..

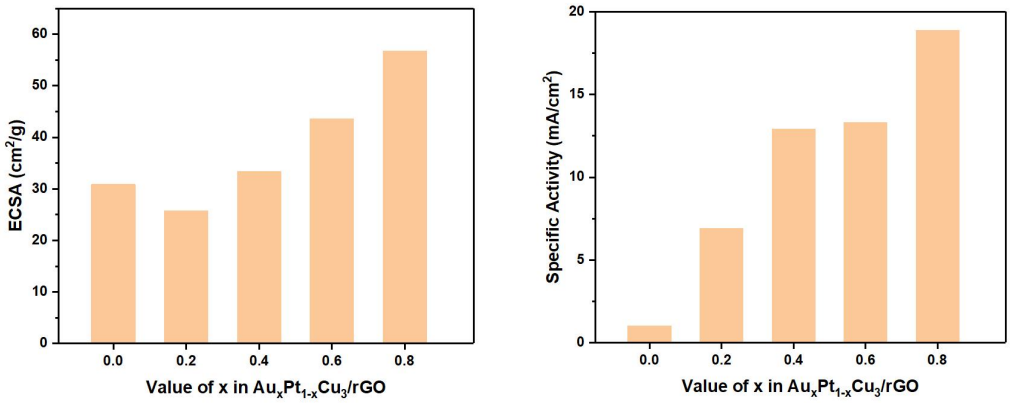


Fig. S12 Electrochemical active surface area (ECSA) and specific activity of $Au_xPt_{1-x}Cu_3$/rGO in 1.0 M KOH and 1.0M EtOH. The ECSA is calculated with the area of the Pt oxide reduction peak.[1-2]

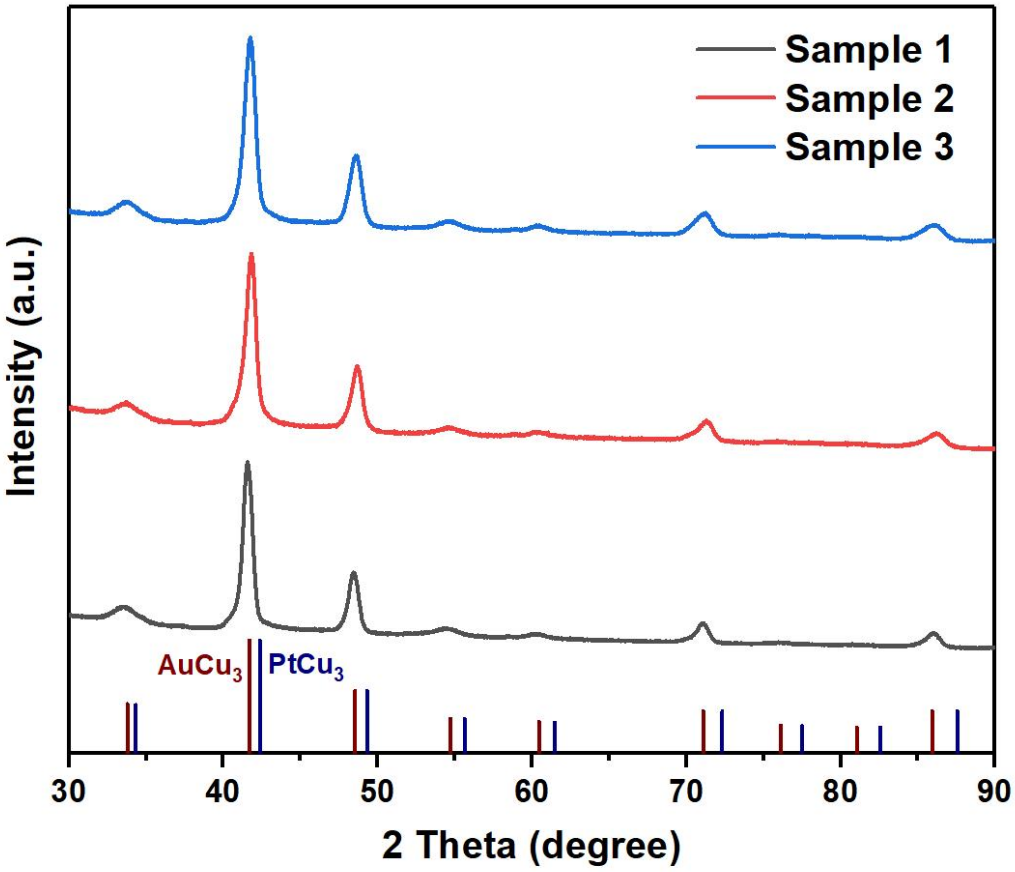


Fig. S13 XRD pattern of $AuCu_3$/rGO parallel samples.

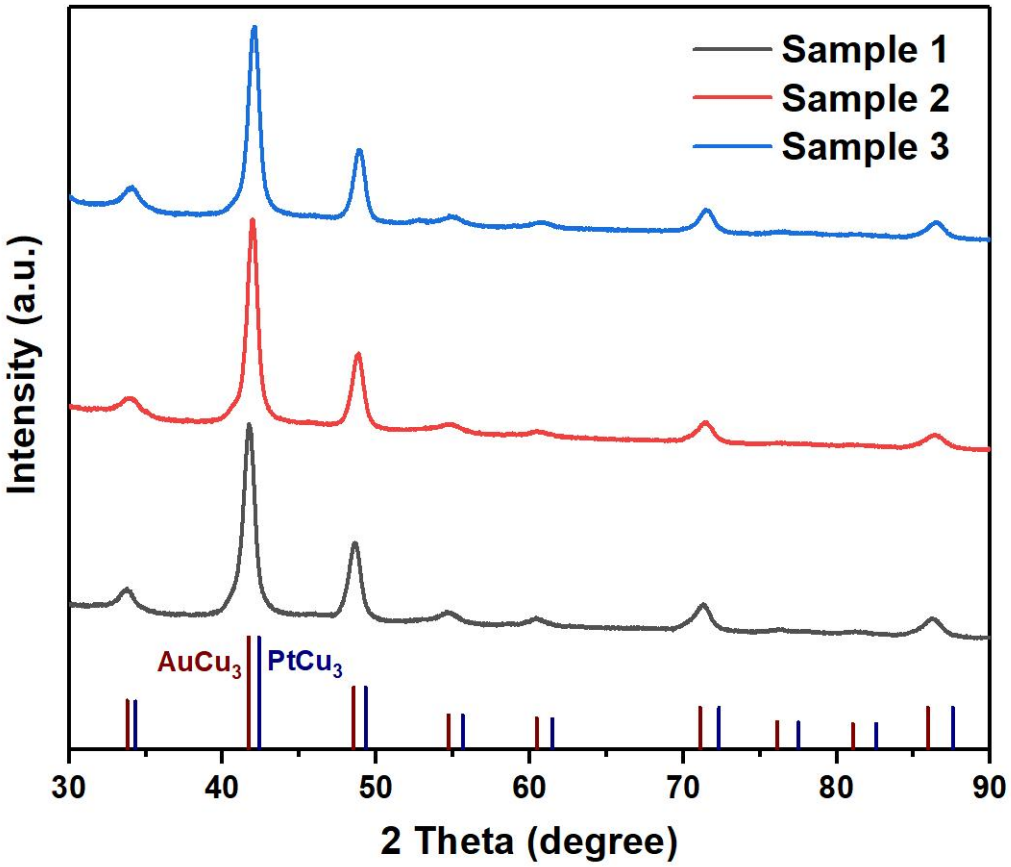


Fig. S14 XRD pattern of $Au_{0.8}Pt_{0.2}Cu_3$/rGO parallel samples.

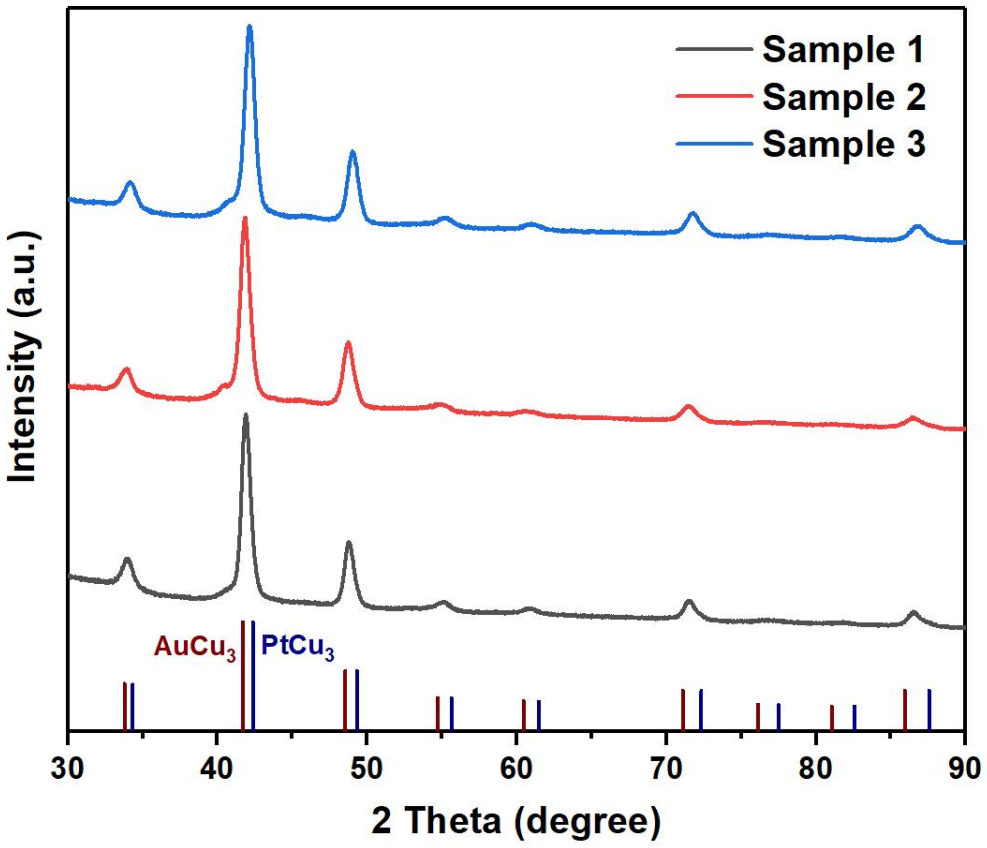


Fig. S15 XRD pattern of $Au_{0.6}Pt_{0.4}Cu_3$/rGO parallel samples.

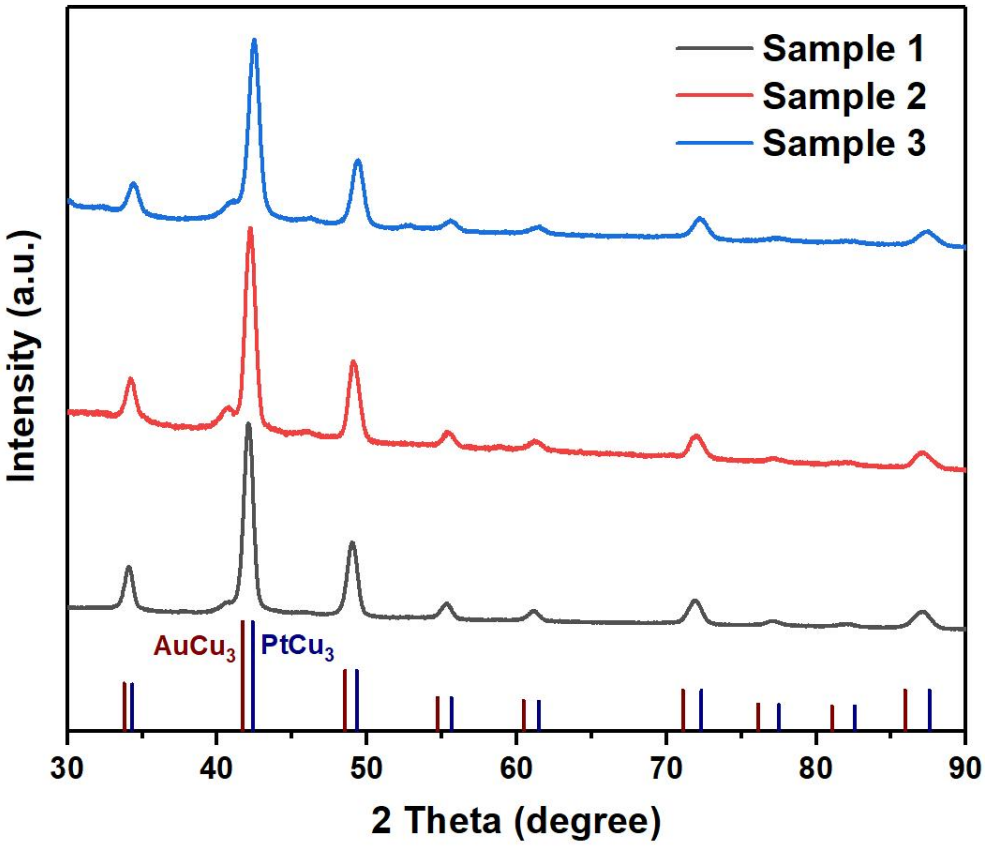


Fig. S16 XRD pattern of $Au_{0.4}Pt_{0.6}Cu_3$/rGO parallel samples.

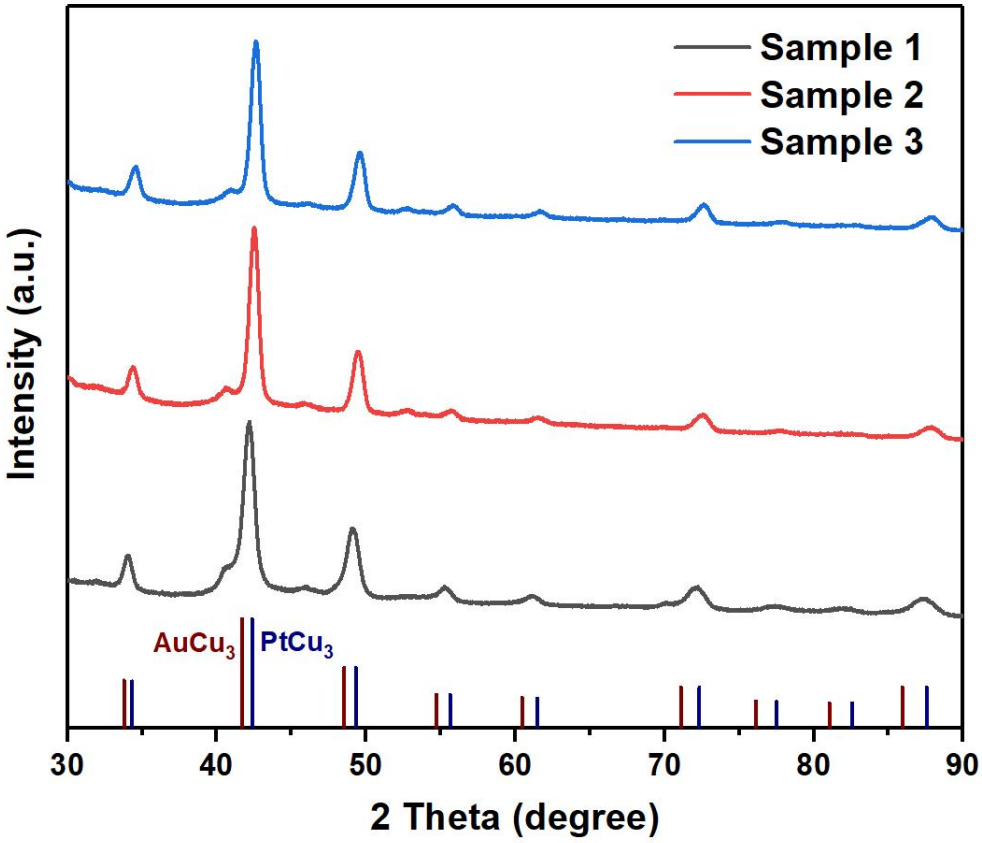


Fig. S17 XRD pattern of $Au_{0.2}Pt_{0.8}Cu_3$/rGO parallel samples.

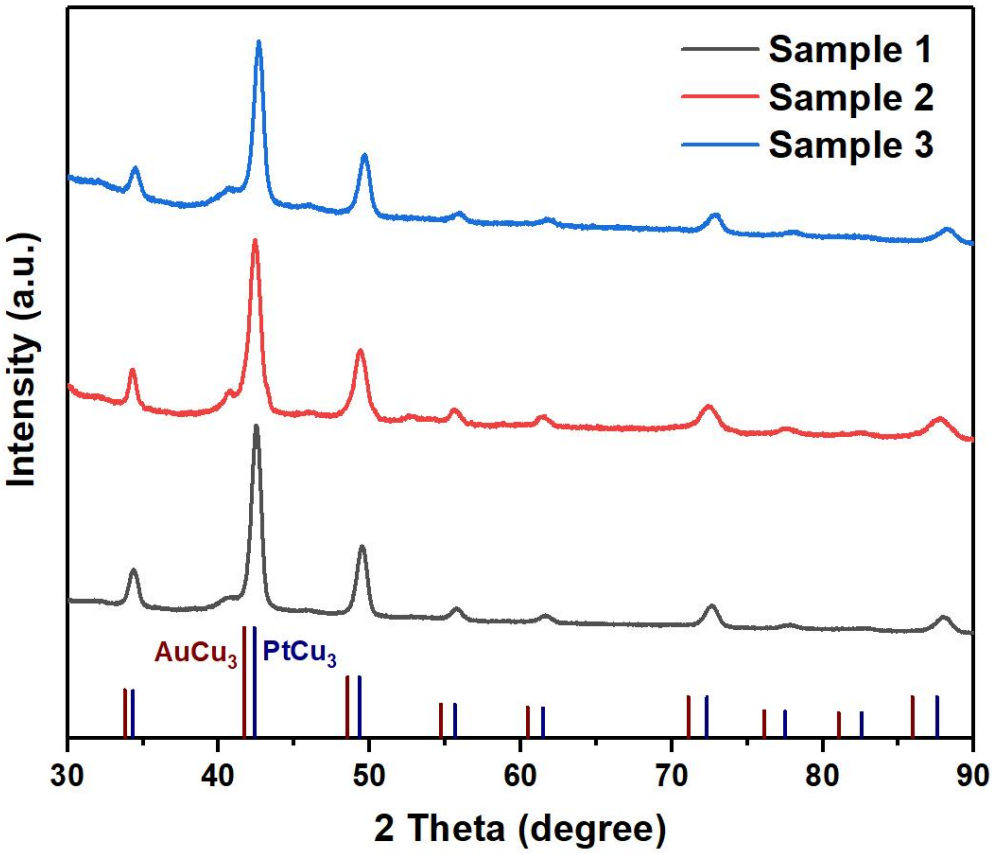


Fig. S18 XRD pattern of $PtCu_3$/rGO parallel samples.

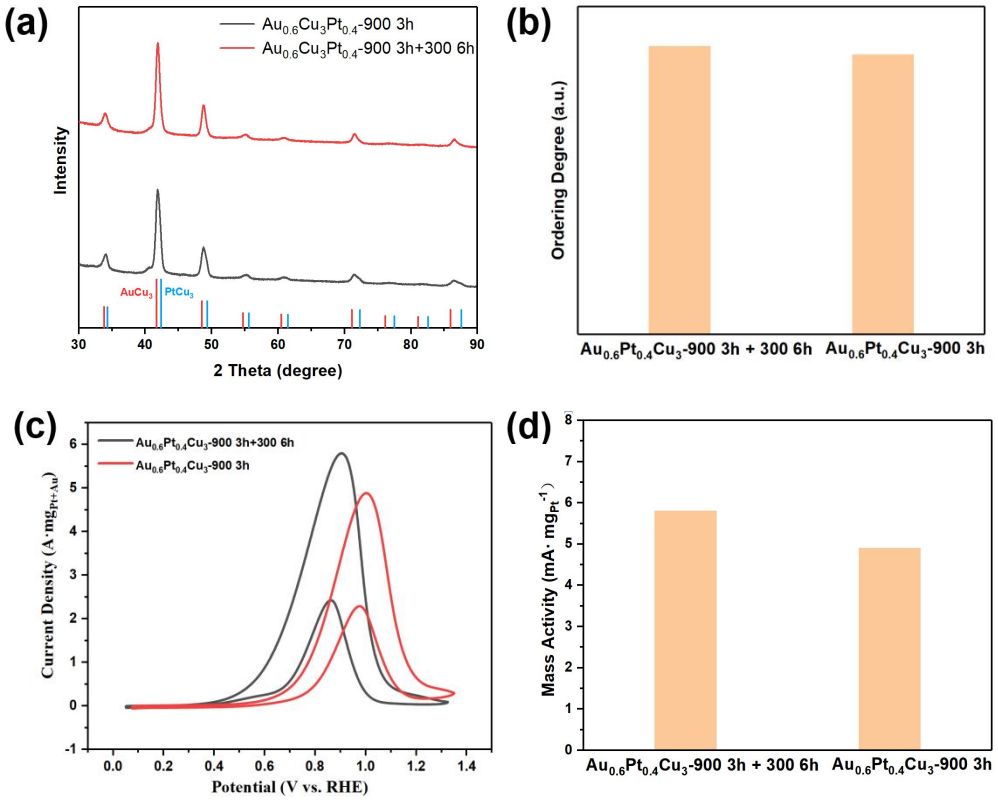


Fig. S19 (a) XRD pattern of $Au_{0.6}Pt_{0.4}Cu_3$/rGO samples annealed at 900℃ for 3h, annealed at 900 ℃ for 3h and 300 ℃ for 6h and (b) their ordering degree. (c) Ethanol oxidation performance of $Au_{0.6}Pt_{0.4}Cu_3$/rGO samples annealed at 900℃ for 3h, annealed at 900℃ for 3h and 300℃ for 6h and (d) activity comparison.

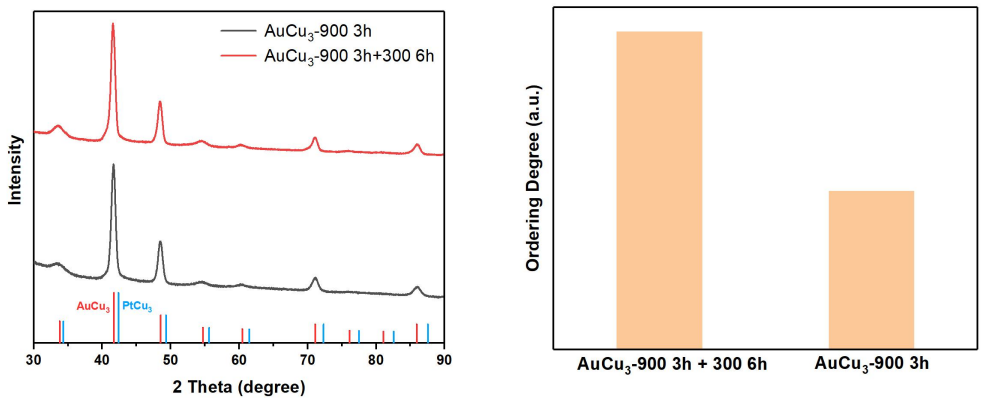


Fig. S20 XRD pattern of $AuCu_3$/rGO samples annealed at 900℃ for 3h, annealed at 900℃ for 3h and 300℃ for 6h and their ordering degree.

Table S1 ICP results of $Au_xPt_{1-x}Cu_3$/rGO samples with various values of x.

Sample	Atomic Ratio		
	Au	Cu	Pt
$PtCu_3$/rGO	0	0.818607955	0.181392045
$Au_{0.2}Cu_3Pt_{0.8}$/rGO	0.028962411	0.829645331	0.141392258
$Au_{0.4}Cu_3Pt_{0.6}$/rGO	0.073480344	0.823508243	0.103011414
$Au_{0.6}Cu_3Pt_{0.4}$/rGO	0.115593784	0.81551273	0.068893487
$Au_{0.8}Cu_3Pt_{0.2}$/rGO	0.138738842	0.826128833	0.035132325
$AuCu_3$/rGO	0.217636877	0.782363123	0

Table S2 Ratios of precursors in $Au_xPt_{1-x}Cu_3$/rGO samples with various values of x.

Sample	$HAuCl_4 \cdot 4H_2O$ (mmol)	$CuCl_2 \cdot 2H_2O$ (mmol)	$H_2PtCl_6 \cdot 6H_2O$ (mmol)
$PtCu_3$/rGO	0	0.075	0.025
$Au_{0.2}Cu_3Pt_{0.8}$/rGO	0.005	0.075	0.020
$Au_{0.4}Cu_3Pt_{0.6}$/rGO	0.010	0.075	0.015
$Au_{0.6}Cu_3Pt_{0.4}$/rGO	0.015	0.075	0.010
$Au_{0.8}Cu_3Pt_{0.2}$/rGO	0.020	0.075	0.005
$AuCu_3$/rGO	0.025	0.075	0

Table S3 d-band center of Pt in the $Pt_{18}Cu_{54}$.

Sites	d-band center (eV）
Pt1:	-1.9396
Pt2:	-1.9422
Pt3:	-1.9428
Pt4:	-1.9423
Pt5:	-1.9409
Pt6:	-1.9421
Pt7:	-1.9436
Pt8:	-1.941
Pt9:	-1.9411
Pt10:	-1.9395
Pt11:	-1.9425
Pt12:	-1.9427
Pt13:	-1.9421
Pt14:	-1.9409
Pt15:	-1.9421
Pt16:	-1.9433
Pt17:	-1.9411
Pt18:	-1.9403
Average	-1.9417

Table S4 d-band center of Pt in the $Au_6Pt_{12}Cu_{54}$.

Sites	d-band center (eV）
Pt1:	-1.9841
Pt2:	-2.0016
Pt3:	-2.003
Pt4:	-1.9831
Pt5:	-1.9941
Pt6:	-1.9945
Pt7:	-1.9814
Pt8:	-1.9871
Pt9:	-1.987
Pt10:	-1.9813
Pt11:	-1.977
Pt12:	-1.9776
Average	-1.98765

Table S5 d-band center of Pt in the $Au_{12}Pt_6Cu_{54}$.

Sites	d-band center (eV）
Pt1:	-2.1428
Pt2:	-2.1434
Pt3:	-2.1096
Pt4:	-2.1625
Pt5:	-2.1505
Pt6:	-2.1428
Average	-2.1419

Reference

(1) F. P. Lohmann-Richters, B. Abel and Á. Varga, *J. Mater. Chem. A*, 2018, 6, 2700-2707.

(2) M. Łukaszewski, M. Soszko and A. Czerwiński, *Int. J. Electrochem. Sci.*, 2016, 11, 4442-4469.