Supplementary Information

Stabilising Gallium-based Liquid Metal Alloy Nanoparticles by Carbon Encapsulation

Imtisal Zahid[a], Karma Zuraiqi[a]*, Caiden J. Parker[a], Muhammad Hamza Nazir[a], Pierre H. A. Vaillant[b], Edwin L. H. Mayes[b], Ali Zavabeti[a], Susanne Wintzheimer[d,e], Vaishnavi Krishnamurthi[a], Dan Yang[c], Aaron Elbourne[b], Ken Chiang[a]*, Torben Daeneke[a]*

[a]Department of Chemical and Environmental Engineering, School of Engineering RMIT University, Melbourne, VIC 3001, Australia

[b]School of Science RMIT University Melbourne, VIC 3001, Australia

[c]School of Engineering, Chemical Engineering, The University of Western Australia (M017), 6009 Perth, Australia

[d]Department of Chemistry and Pharmacy, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), Egerlandstraße 1, 91058 Erlangen, Germany

[e]Fraunhofer-Institute for Silicate Research ISC, Neunerplatz 2, 97082 Würzburg, Germany

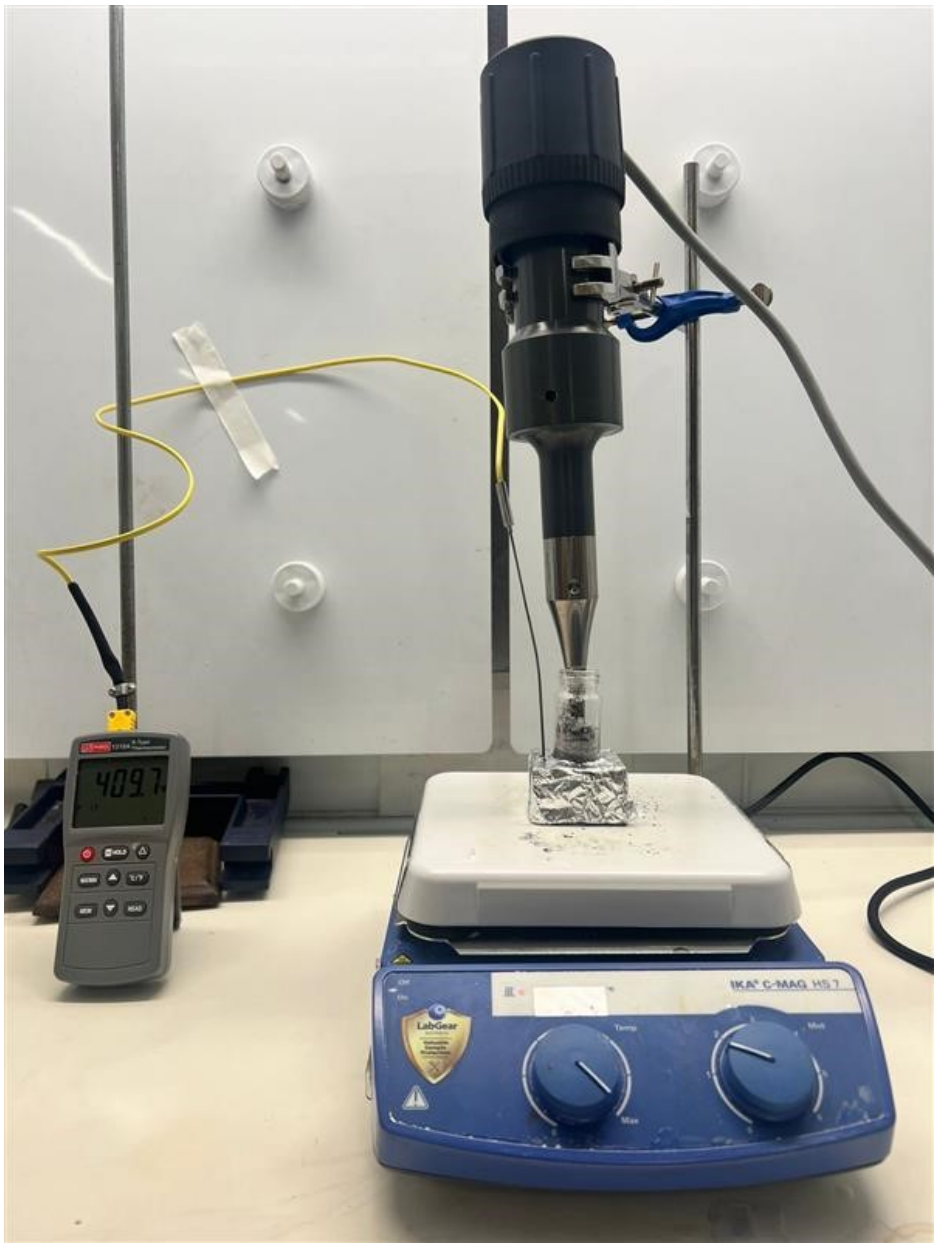

Figure S1. Synthesis setup for bimetallic and trimetallic LM NPs.

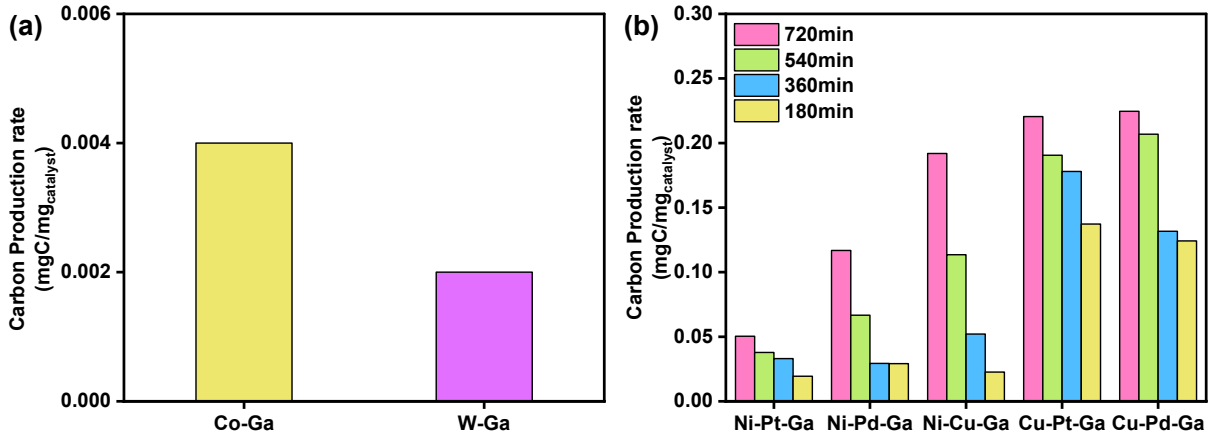

Figure S2. Methane pyrolysis at the following reaction conditions: 600 °C, 0.1L min^{-1} gas flow of 4% methane/argon over various binary and ternary catalysts (a) Carbon production rate over Co-Ga and W-Ga, (b) Carbon production at different time intervals in the presence of ternary alloy nanoparticles (NPs).

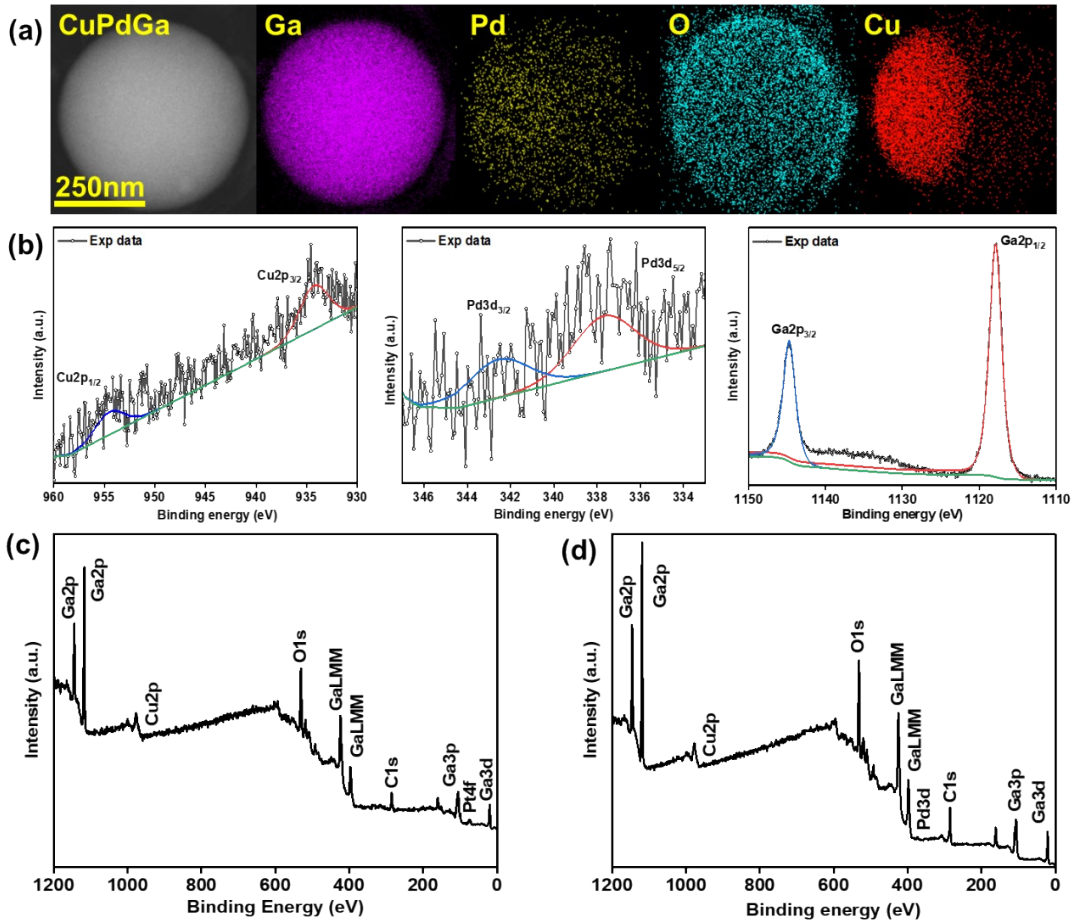

Figure S3. Surface and elemental analysis of the pristine Cu-Pd-Ga (3.0:0.5:96.5) nanoparticle (NP), (a) Dark-field Transmission electron microscopy (TEM-EDS) image of Cu-Pd-Ga NP with an individual elemental map of Ga, Pt, O, and Cu, (b) XPS spectrum of the Ga*2p* region, Cu*2p* region and Pd*3d* region in Cu-Pd-Ga (3.0:0.5:96.5) alloy NPs, (c,d) XPS survey scan of Cu-Pt-Ga and Cu-Pd-Ga.

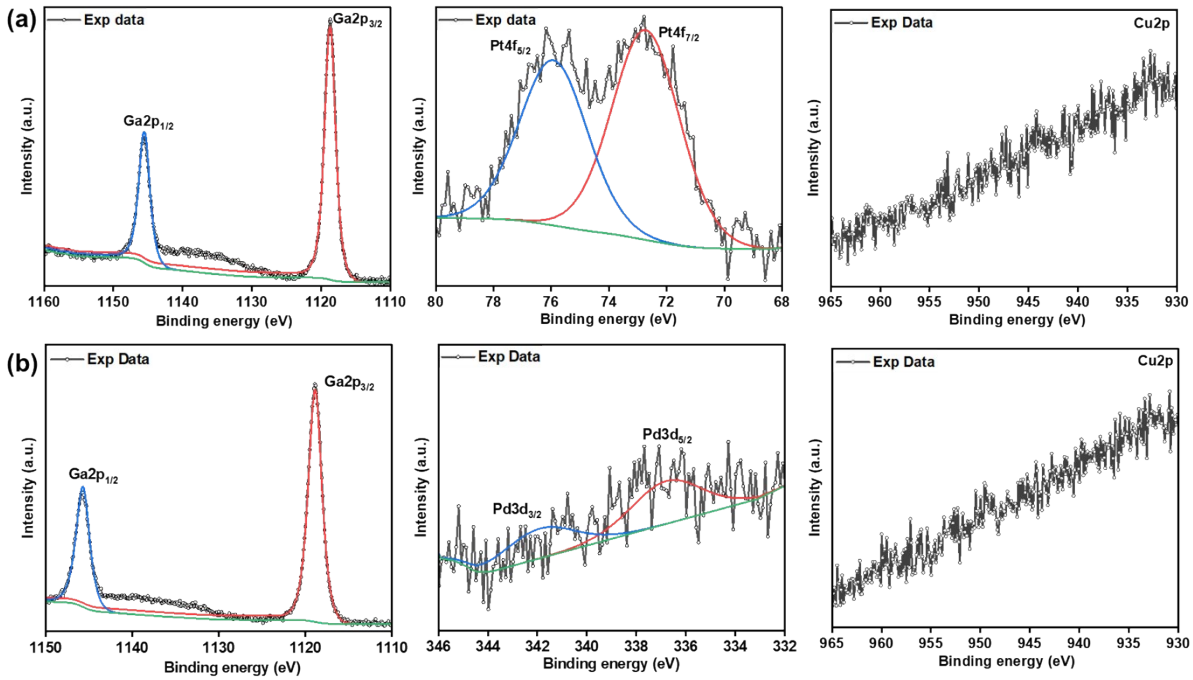

Figure S4. Surface and elemental analysis of CC-Cu-Pt-Ga and CC-Cu-Pd-Ga, (a) Ga2p region and Pt *4f* region over CC-Cu-Pt-Ga catalyst, (b) CC-Cu-Pd-Ga XPS spectrum of Ga*2p* region and Pd*3d* region.

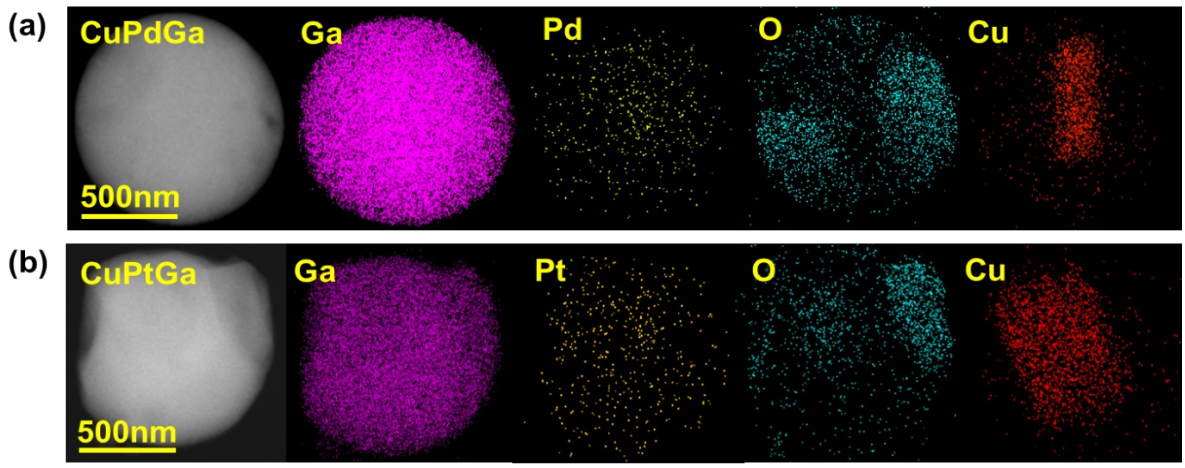

Figure S5. Dark field transmission electron microscopy (DF-TEM) images of fresh and spent ternary alloy NPs after methane decomposition, (a) CC-Cu-Pd-Ga with elemental mapping images of Ga, Pd, O and Cu, (b) CC-Cu-Pt-Ga catalyst TEM images along an elemental map of Ga, O, Pt and Cu.

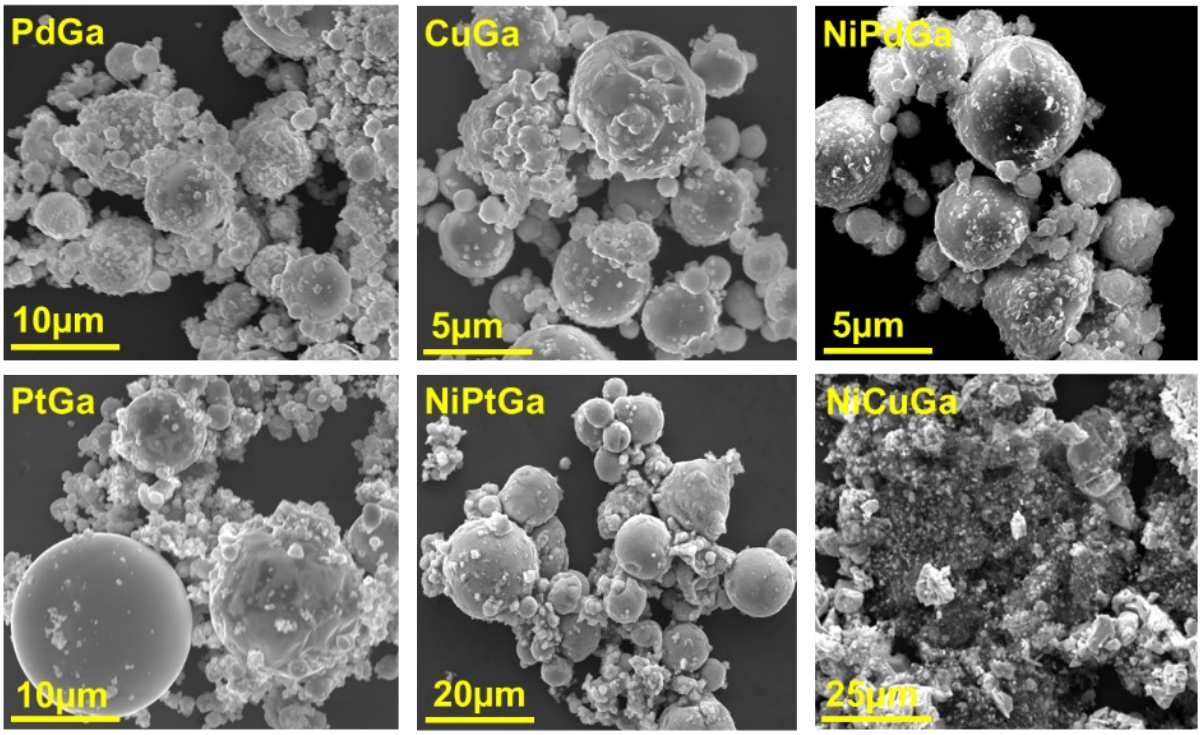

Figure S6. SEM imaging of binary and ternary NPs (Pd-Ga, Cu-Ga, Pt-Ga, Ni-Pt-Ga, Ni-Pd-Ga, and Ni-Cu-Ga) after methane pyrolysis and carbon formation.

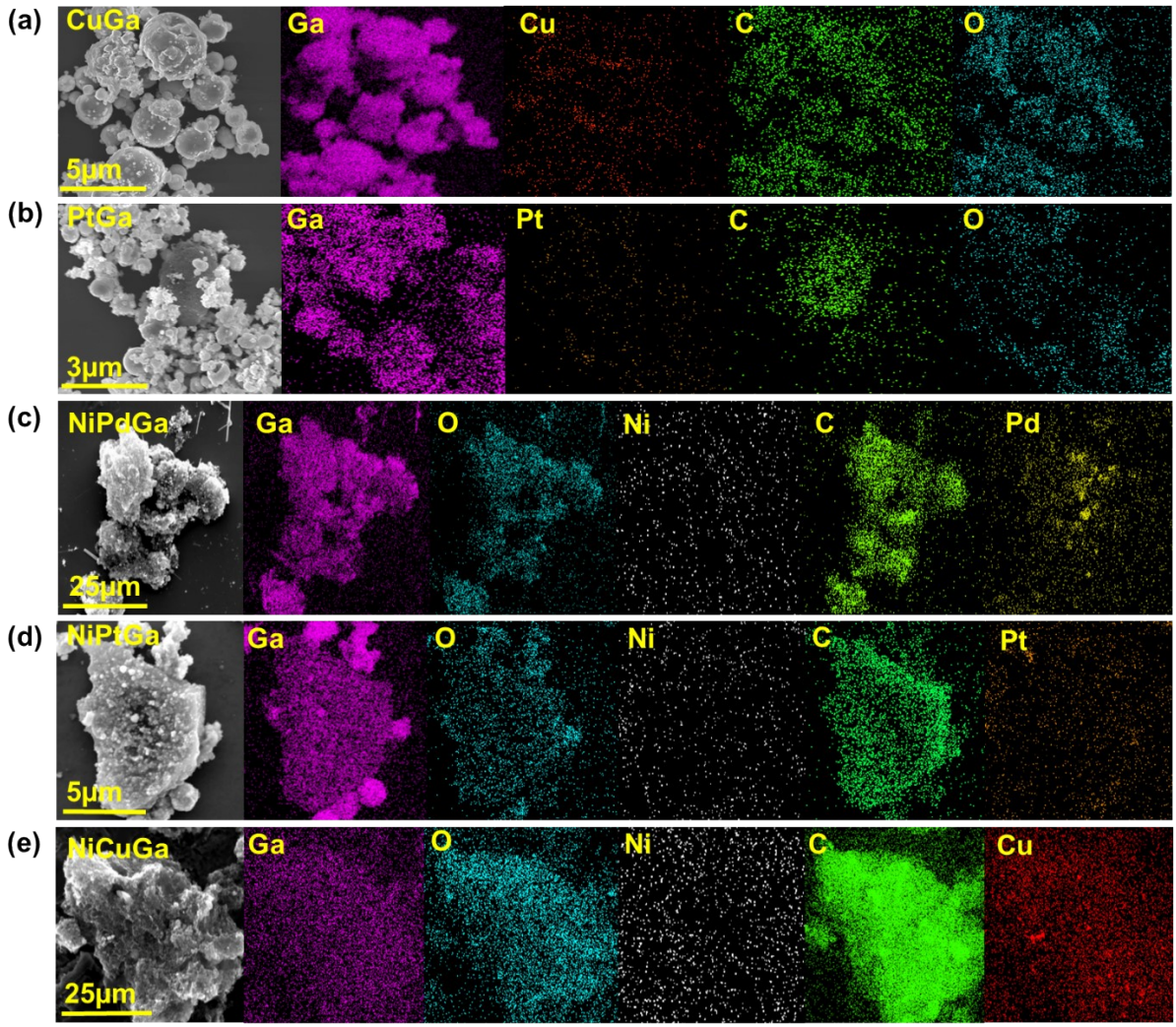

Figure S7. SEM-EDS of binary and ternary alloy NPs after methane decomposition at 600°C, (a) Cu-Ga (03:97), (b) Pt-Ga (0.5:99.5), (c) Ni-Pd-Ga (3.0:0.5:96.5), (d) Ni-Pt-Ga (3.0:0.5:96.5), (e) Ni-Cu-Ga (3.0:03:94).

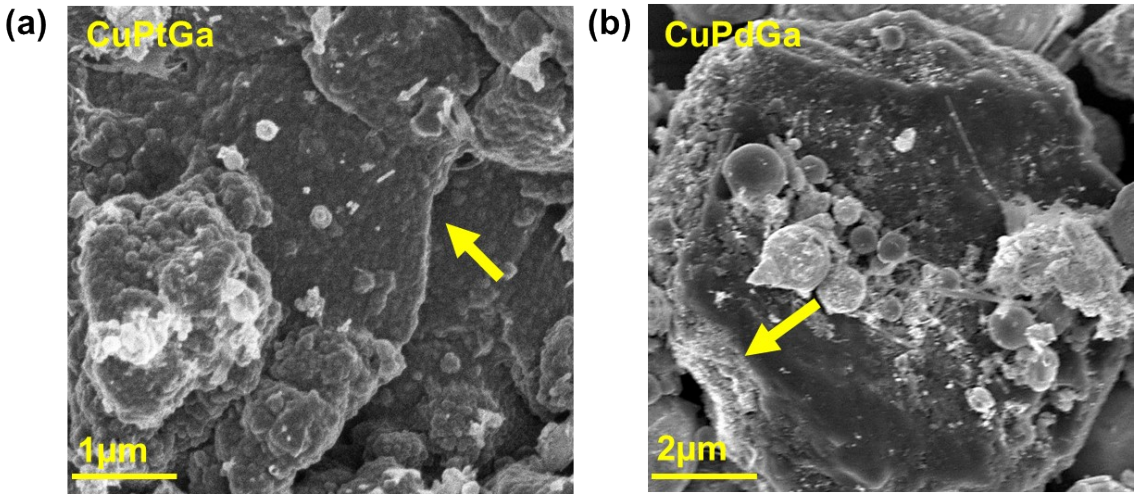

Figure S8. SEM imaging of rod-like structures formed during methane pyrolysis over the following catalysts: (a) Cu-Pt-Ga, (b) Cu-Pd-Ga.

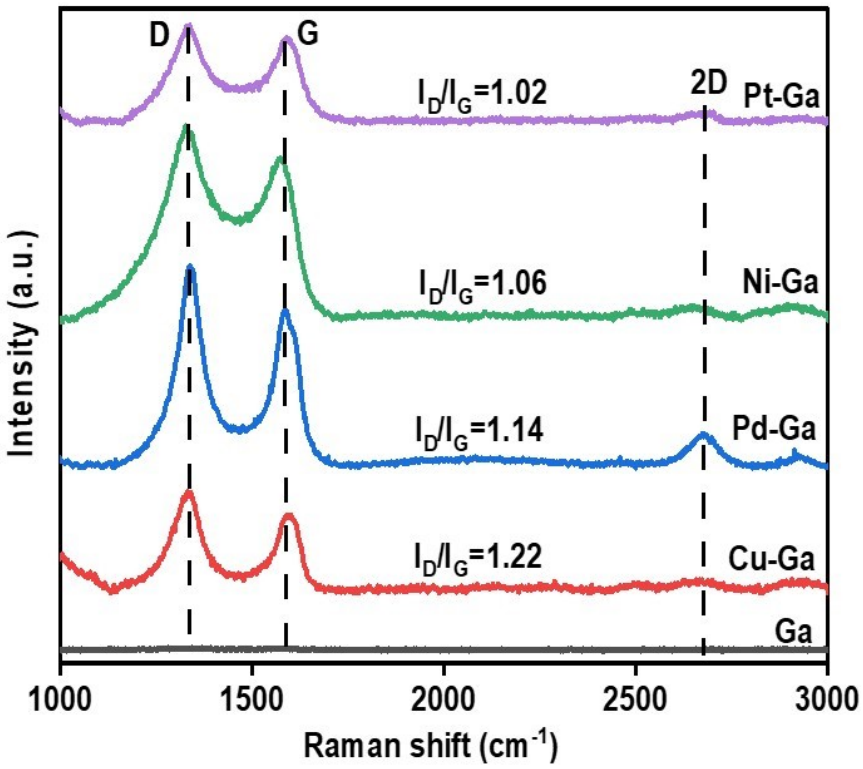

Figure S9. RAMAN spectra of binary alloy NPs after methane decomposition at 600 °C for 180 min.

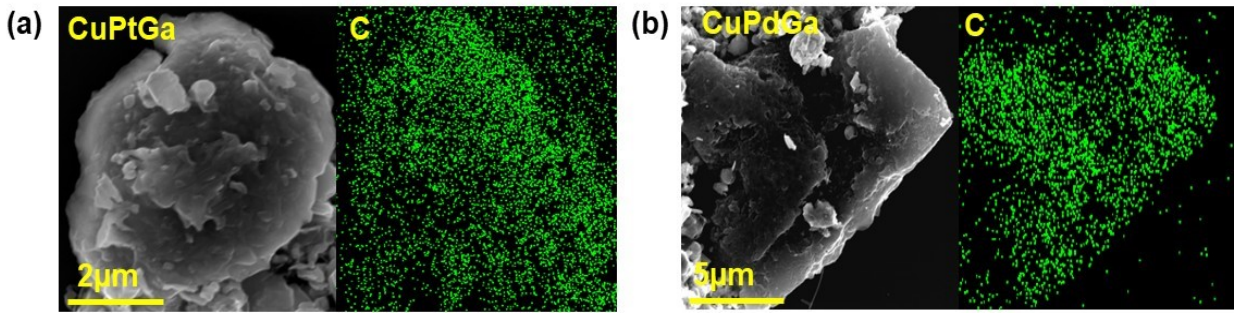

Figure S10. (a) SEM-EDS of graphitic carbon product formed after methane pyrolysis over Cu-Pd-Ga (3.0:0.5:96.5) NPs, (b) Product graphitic carbon by Cu-Pt-Ga (3.0:0.5:96.5) NPs.

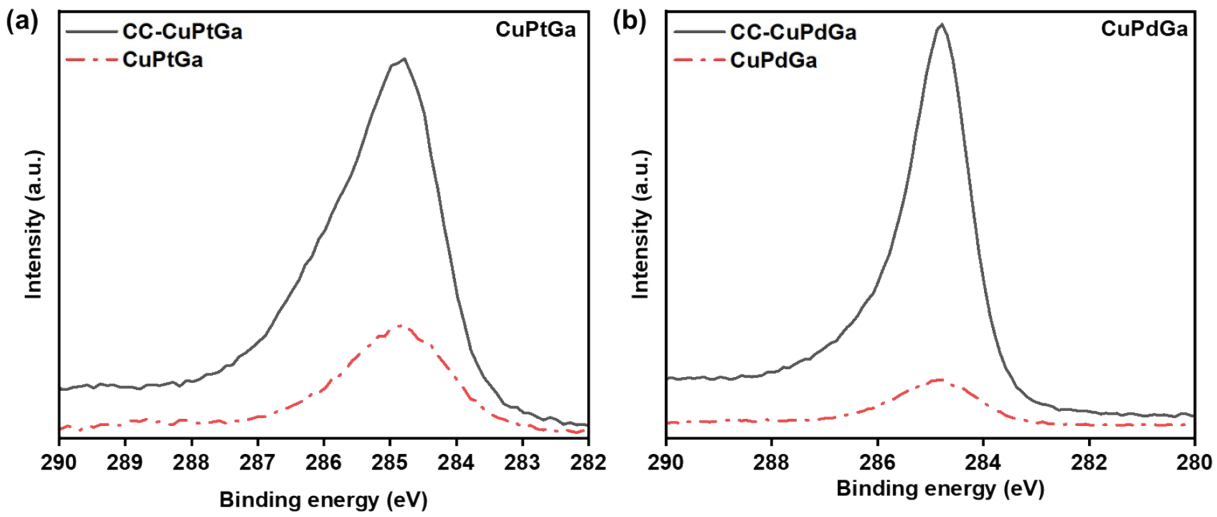

Figure S11. XPS spectrum of carbon region (C1s) detected before and after carbon coating by the following catalysts: (a) Cu-Pt-Ga and (b) Cu-Pd-Ga.

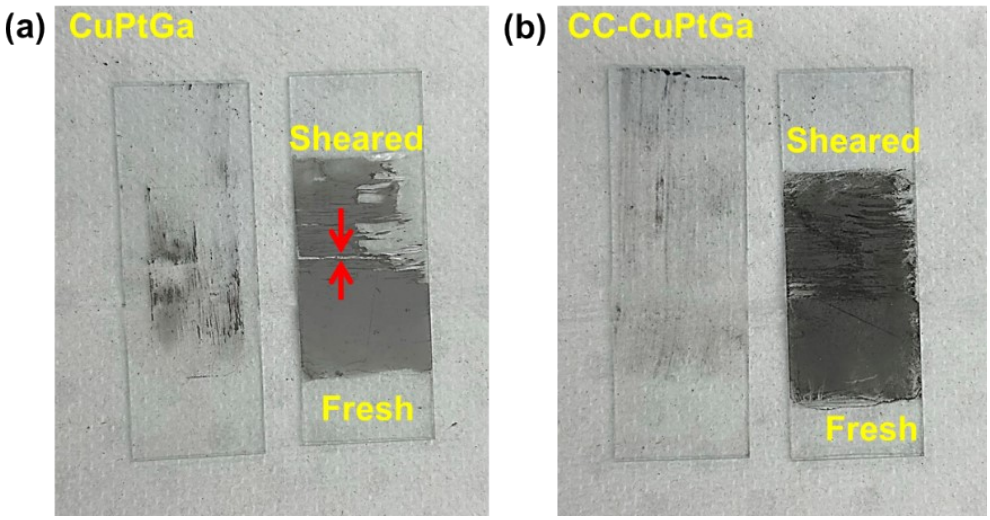

Figure S12. (a, b) Comparison of Cu-Pt-Ga and CC-Cu-Pt-Ga before and after mechanical stress testing (MS).

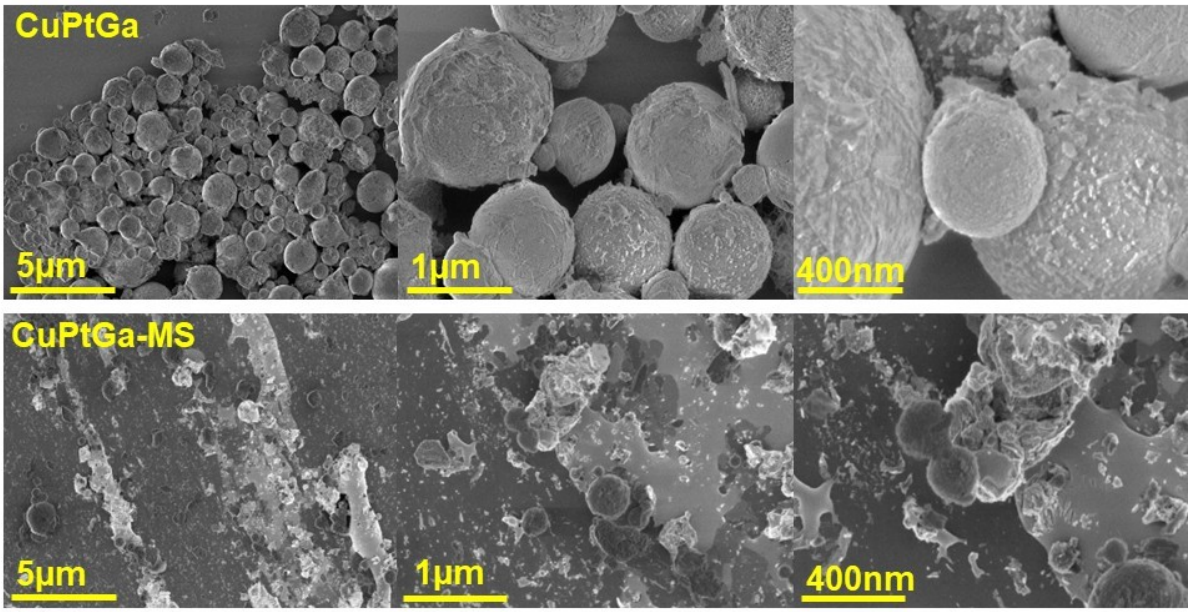

Figure S13. SEM imaging of Cu-Pt-Ga nanoparticles before and after mechanical stress testing (MS).

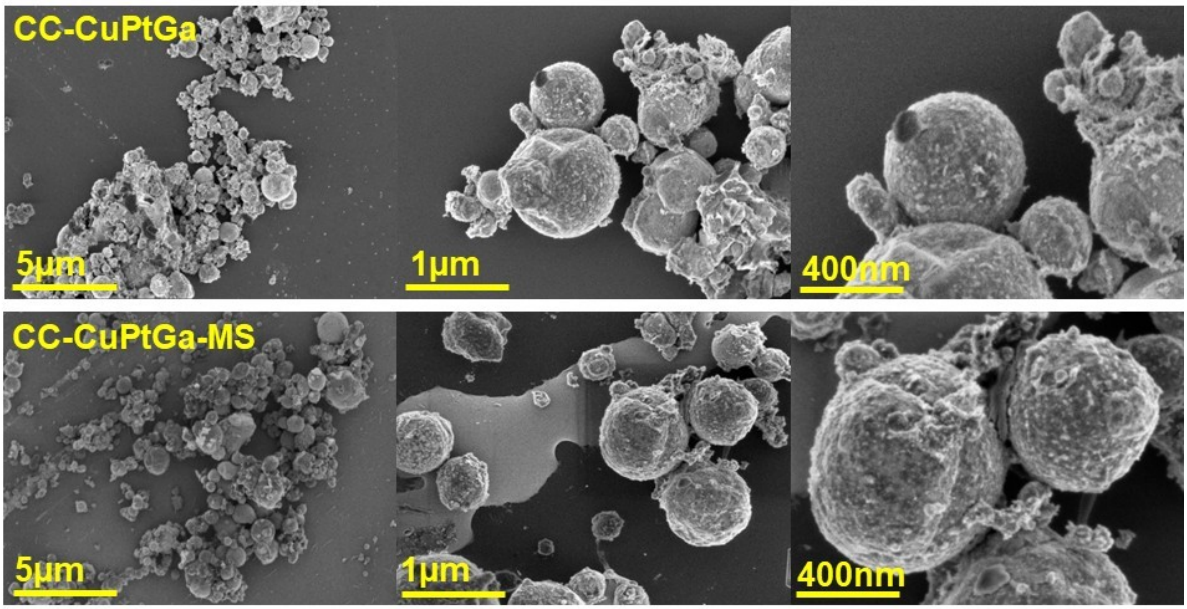

Figure S14. SEM imaging of CC-Cu-Pt-Ga nanoparticles before and after mechanical stress testing (MS).

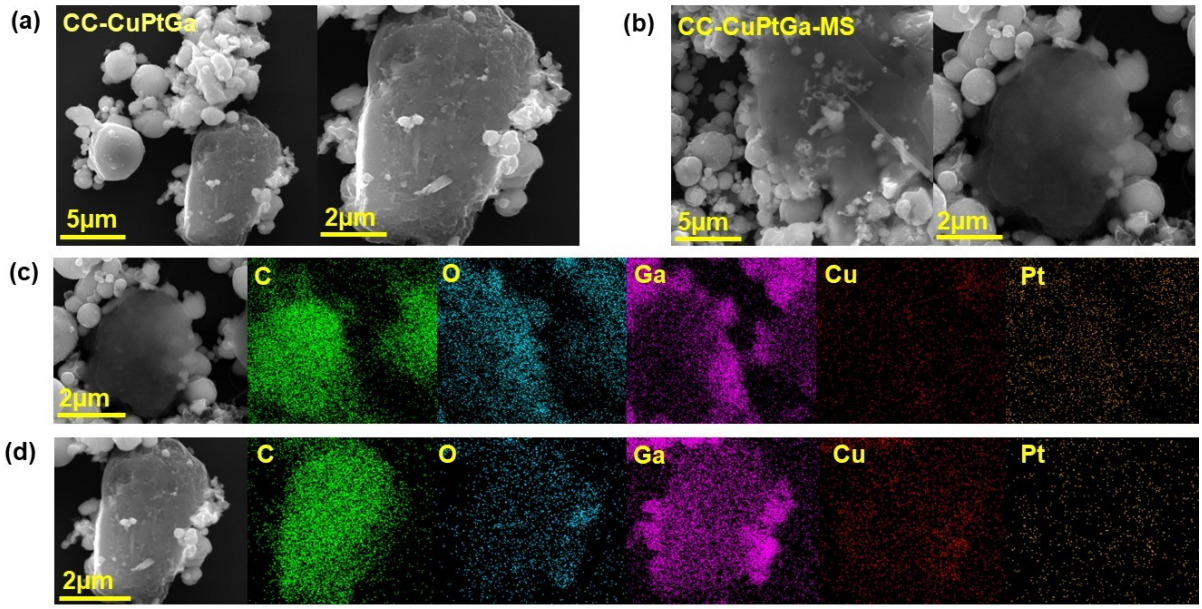

Figure S15. SEM imaging of carbon flake in (a) CC-Cu-Pt-Ga and (b) CC-Cu-Pt-Ga after mechanical stress testing (MS), (c, d) SEM-EDS elemental mapping of carbon product present in (c) CC-Cu-Pt-Ga and (d) CC-Cu-Pt-Ga after mechanical stress testing (MS).

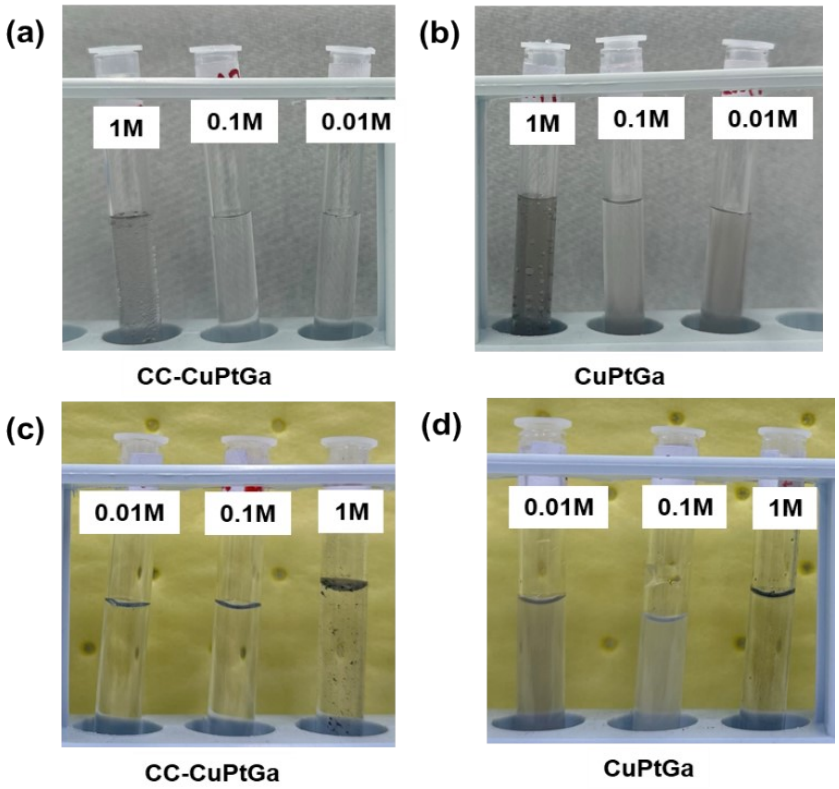

Figure S16. Cu-Pt-Ga and carbon-coated Cu-Pt-Ga (CC-Cu-Pt-Ga) nanoparticles after different hours of HCl exposure (a, b) 3hrs and (c, d) 48hrs.

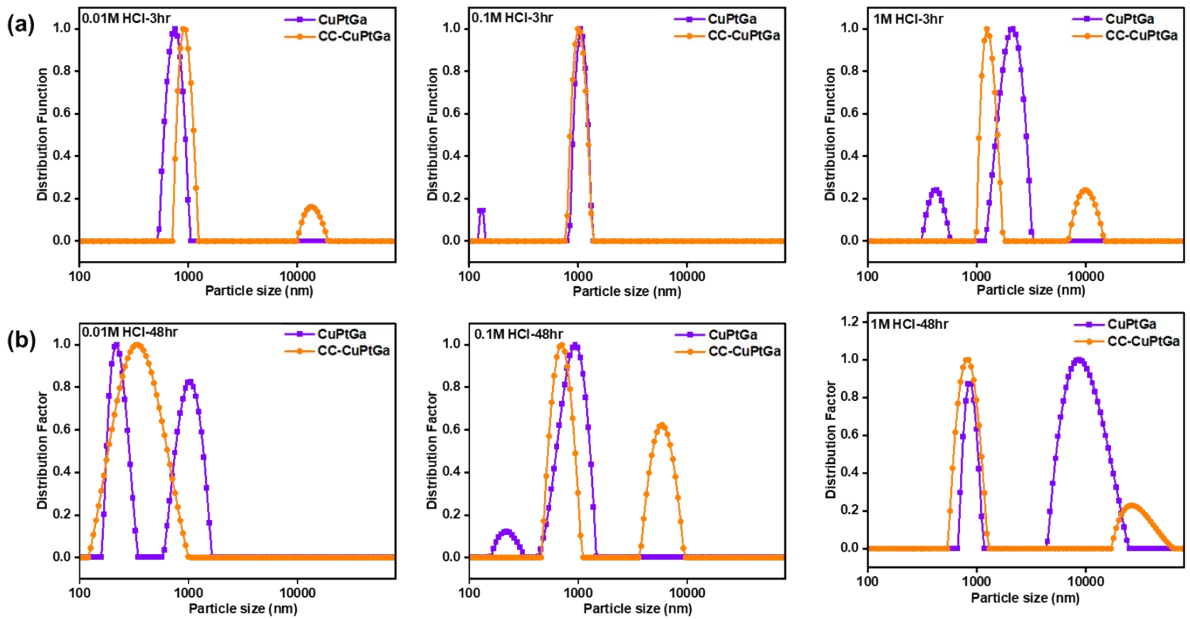

Figure S17. Dynamic light scattering (DLS) droplet size distribution of Cu-Pt-Ga and CC-Cu-Pt-Ga after following durations to acidic media (0.1M, 0.01M and 1M HCl solution): (a) 3 hrs exposure and (b) 48 hrs exposure.

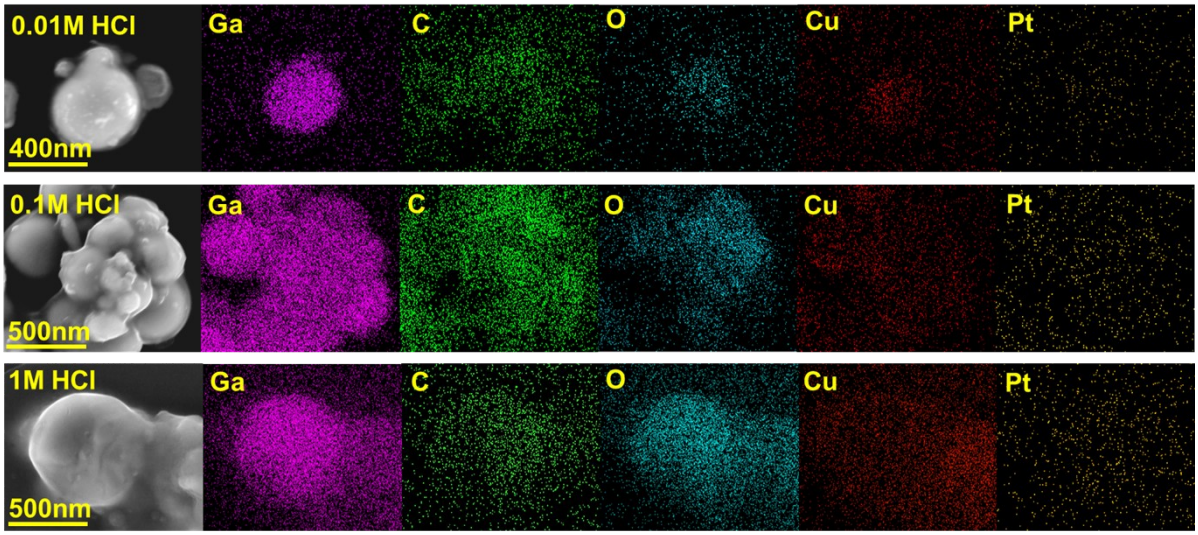

Figure S18. SEM-EDS of CC-Cu-Pt-Ga alloy NPs after 3 hrs. exposure to acidic media (0.01M, 0.1M and 1M HCl).