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Supporting Information

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The Hallmarks of Copper Single Atom Catalysts in Direct Alcohol Fuel Cells and Electrochemical CO₂ Fixation

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Figure S1. X-ray diffraction diagrams from of pristine fluorographene (FG, the precursor of G-CN), cyanographene (G-CN) and copper-loaded cyanographene (G(CN)-Cu). X-Ray radiation used: Co K-alpha (1.789 Å).

Discussion: The catalyst powder (as well as the rest of the solids) lacks completely reflections from any inorganic nanoparticles. Only the very broad reflection at 30.4 degrees is present, typical of non-restacked graphene powders and present in carbon materials. The extent of the parallel stacking of graphenes (thickness of the crystallites, Lc) can be estimated from the Scherrer formula.^[1,2] In the case of the G(CN)-Cu is 1.8 nm, which confirms that the catalyst is composed of few layer graphene stacks, which are in a disordered state between each other.

^[1] T. Kyotani, N. Sonobe, A. Tomita, Nature 1988, 331, 331.

^[2] F. Sun, X. Liu, H. B. Wu, L. Wang, J. Gao, H. Li, Y. Lu, Nano Lett. 2018, 18, 3368.

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Figure S2. High resolution and deconvoluted spectra of (b) C 1s, and (b) N1s peaks.



Figure S3. High-resolution AFM image for G-CN – the 3D model, topography and Young's modulus map.

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A higher-resolution AFM image of a region containing both soft and stiff nanoplate, is shown in **Figures S4-S5**. It is evident that the soft nanoplate also has significantly higher roughness compared to the stiff nanoplate. Since YM of the soft nanoplate is comparable to that of the soft amorphous phase, it is quite possible that material comprising the soft amorphous phase can also deposit on nanoplates. This conclusion would explain the heterogeneity in terms of roughness and YM values between the nanoplates.



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Figure S4. High-resolution AFM image for G(CN)-Cu – the 3D model, topography, and Young's modulus map.