Supporting Information

Cascade growth and performance optimization of laminated heterointerface based on graphdiyne

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1. Experimental

$$\frac{1}{C_{SC}^2}A^2 = \left(\frac{2}{\varepsilon\varepsilon_P e N_D}\right)\left(E - E_{FB} - \frac{k_B T}{e}\right)$$

The M-S equation can be expressed by the above equation. Here, C_{SC} is the space-charge capacitance, A is the area, N_D is the carrier density, E is the applied voltage, E_{FB} is the flat band potential, k_B is Boltzmann's constant, T is the absolute temperature, e is the electronic charge, ε is the vacuum permittivity, and ε_p is the relative permittivity.

2. Supplementary Figures

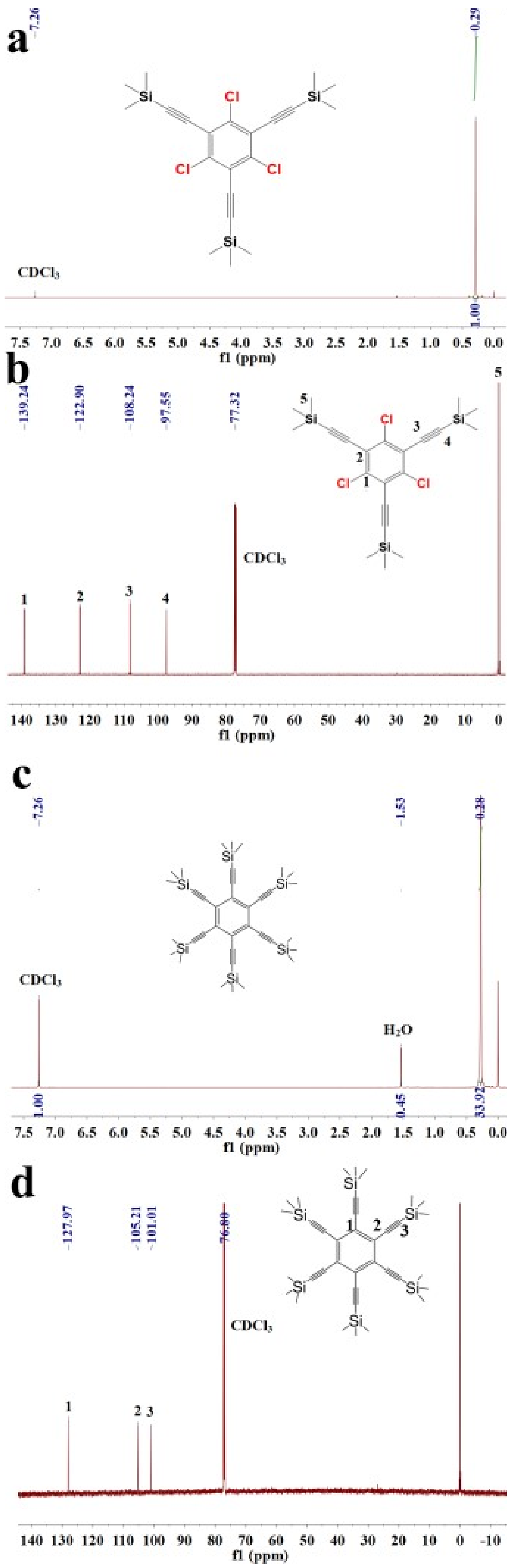


Figure S1 ^{1}H NMR (a) and ^{13}C NMR (b) of the precursor 1,3,5-(trimethylsilyl)ethynyl-2,4,6-trichlorobenzene. ^{1}H NMR (c) and ^{13}C NMR (d) of the precursor hexakis[(trimethylsilyl)ethynyl]benzene.

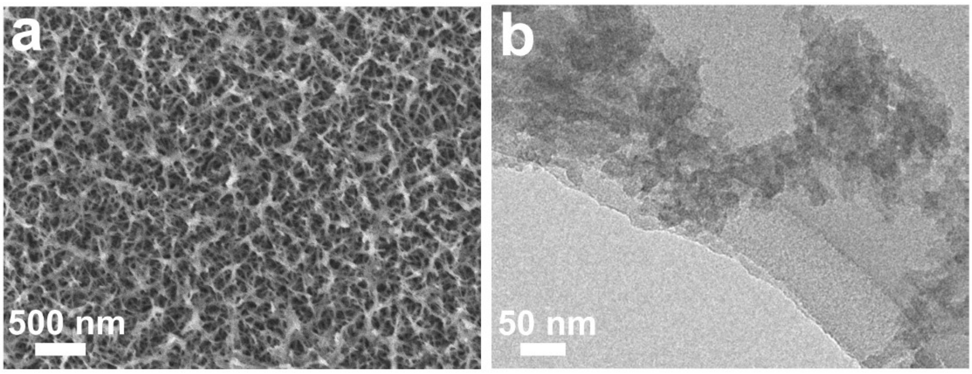


Figure S2. SEM (a) and (b)TEM images of reference Cl-GDY film on copper foil.

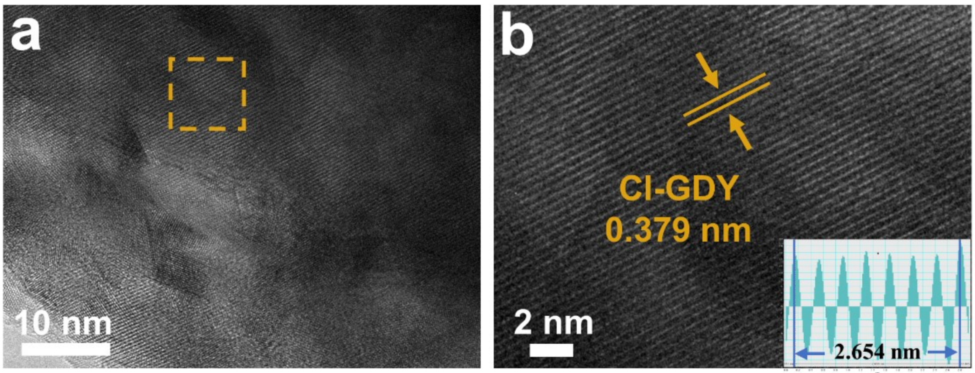


Figure S3. (a-b) HRTEM images of referencence Cl-GDY film exfoliated from copper substrates (The inset image is SAED pattern of Cl-GDY).

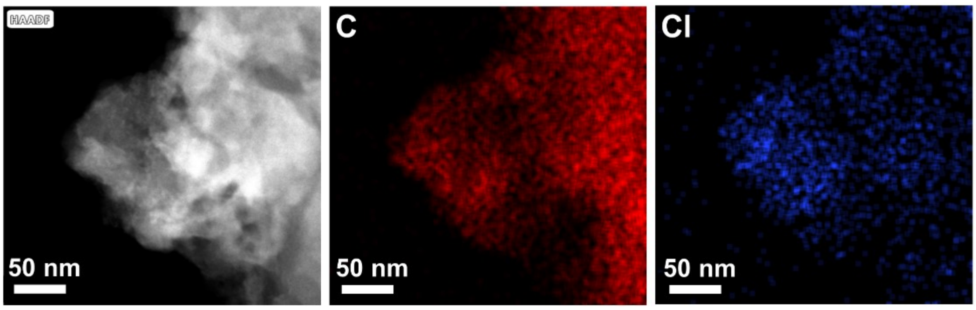


Figure S4. Overlapping and elemental mapping images of Cl-GDY film.

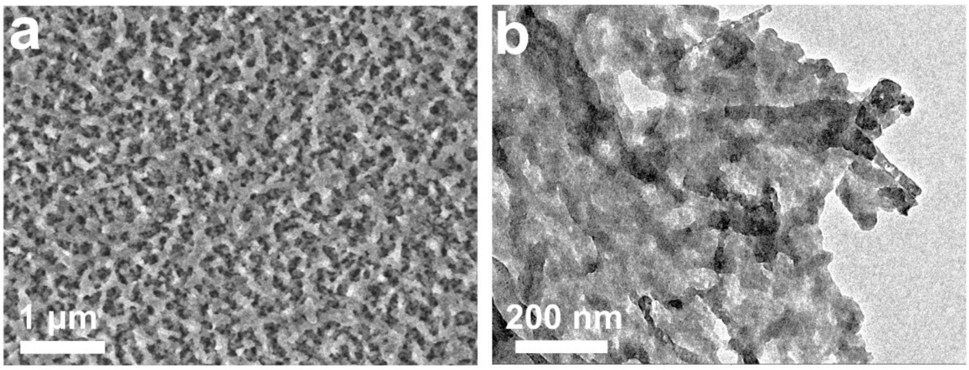


Figure S5. SEM (a) and (b)TEM images of pristine GDY film on copper foil.

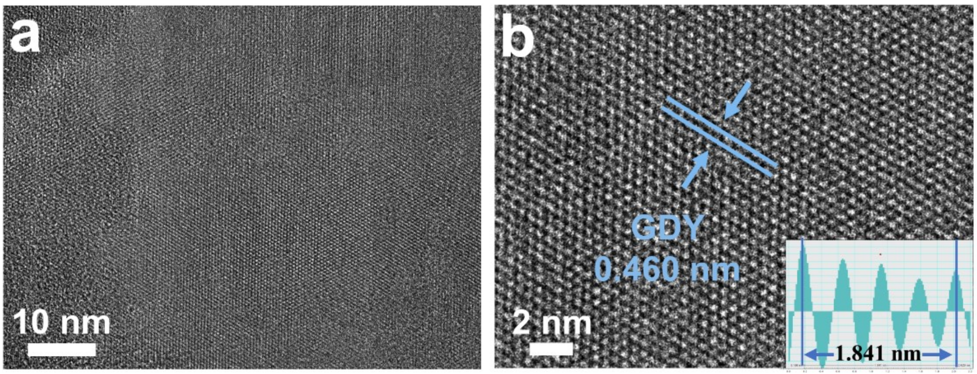


Figure S6. (a-b) HRTEM images of pristine GDY film exfoliated from copper substrates. And the lattice space is 0.46 nm.

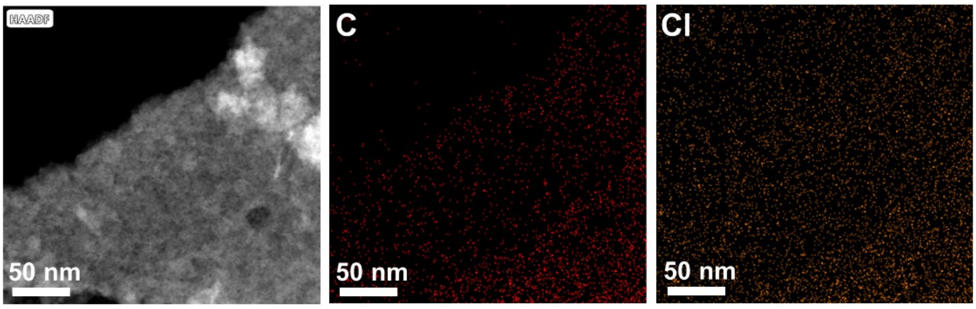


Figure S7. Overlapping and elemental mapping images of Cl-GDY/GDY film.

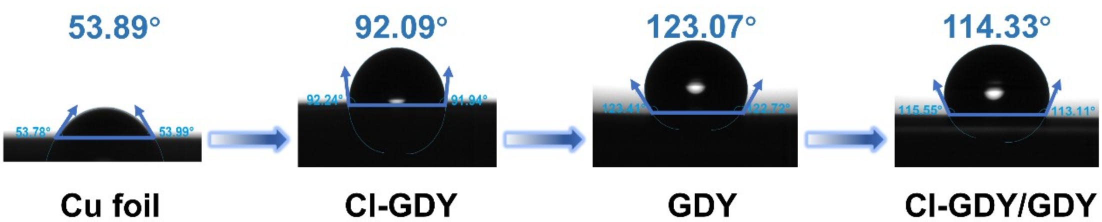


Figure S8. The contact angle of Cu foil, Cl-GDY, GDY, and Cl-GDY/GDY.

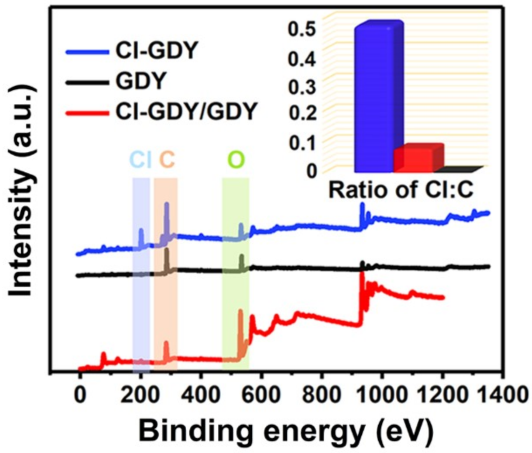
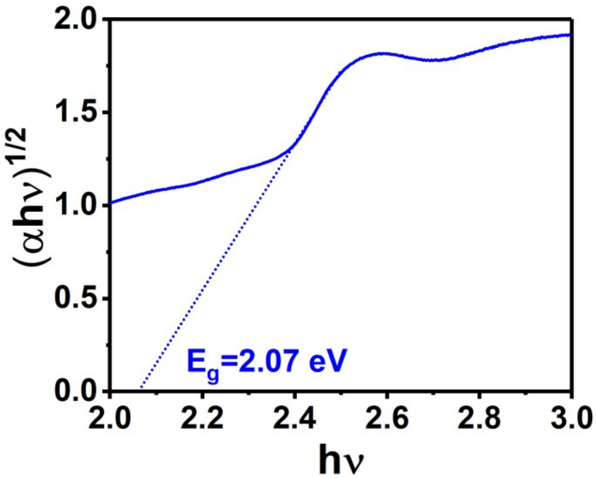
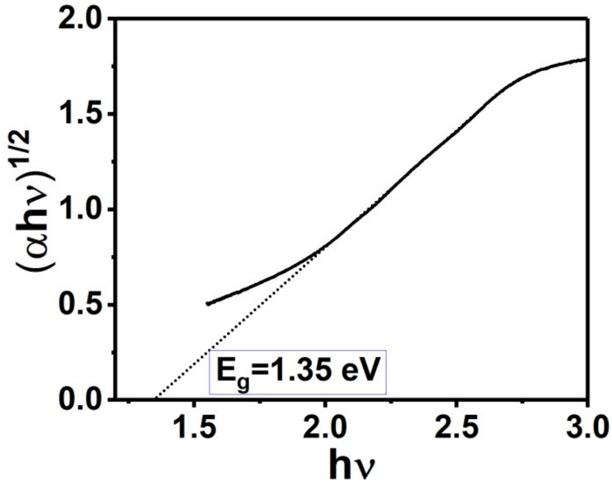


Figure S9. Comparison of XPS survey spectra of Cl-GDY, GDY, and Cl-GDY/GDY film. The inset graph shows the weight ratio of chlorine to carbon atoms.





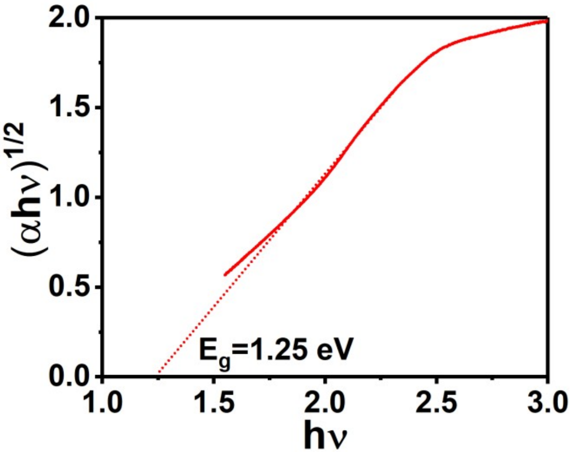


Figure S10. Tauc Plots for the optical band gaps. The blue, black and red line represent the curve of Cl-GDY, GDY, and Cl-GDY/GDY, respectively.

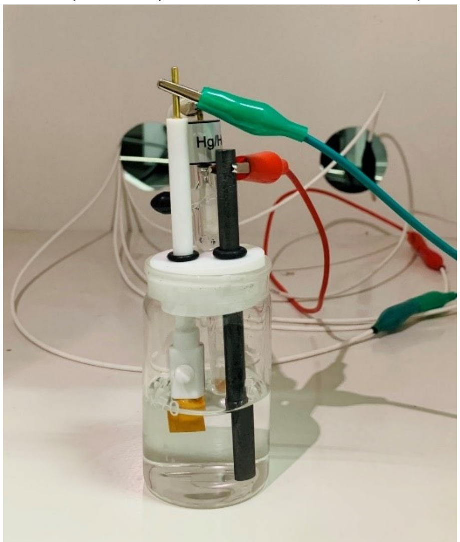


Figure S11. The photograph of the electrochemical cells with three electrodes, in which the copper foil serves as the working electrode, the carbon rod acts as the counter electrode, and the saturated calomel electrode (SCE) is used as the reference electrode.

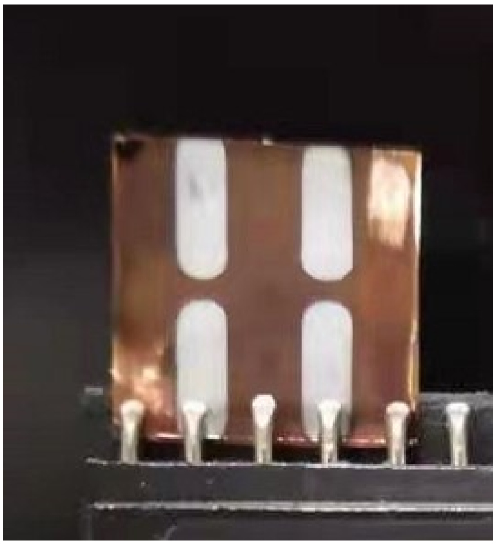


Figure S12. The photograph for the carbon-based heterojunction device.

3. Supplementary Tables

Table S1 Specific feeding ratio of experimental conditions.

Reagent	Cl-GDY	GDY
Precursor (mg)	15	15
THF (mL)	30	30
TBAF (mL)	0.6	0.6
Pyridine (mL)	1	1
TMEDA (mL)	10	10
Acetone used to dissolve the precursors (mL)	30	30
Acetone added in the reaction bottle (mL)	100	100
Color of film	Orange	Brown-red

Table S2 The summary of energy levels and band gaps from the method of optical method, and electrochemical method.

	Optical Method [a]			Electrochemical Method [b]		
	Valence Band	Conduction Band	Band Gap	22 Hz	100 Hz	1000 Hz
Cl-GDY	1.10	3.17	2.07	0.113	0.138	0.253
Cl-GDY/GDY	0.94	2.19	1.25	0.077	0.058	0.210
GDY	1.22	2.57	1.35	-	-	-

[a]The units is eV. [b]The units is V. The value of conduction band obtained from the measured E_{FB} under different applied frequency. All the data is compared to normal hydrogen electrodes (NHE).