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Tuning the Properties of Graphdiyne through Introducing Electron Withdrawing/Donating Groups

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Abstract: Herein, properties of graphdiyne (GDY) such as energy gap, morphology, and affinity to alkalis metals are efficiently adjusted through including electron withdrawing/donating groups. The experimental and theoretical results indicate that the push-pull electron ability and size differences of groups play a key role on the partial property adjusting of as-prepared GDY derivatives named MeGDY, HGDY, and CNGDY. Typically, cyano group with electron withdrawing ability and methyl group with electron donating characteristic have successfully reduced the band gap and increased the conductivity of the GDY network within an obvious range. Meanwhile, the inclusion of cyano and methyl groups affects the aggregation of GDY, thus providing a higher number of micropores and specific surface area. Moreover, the fascinating properties of these groups endow the original GDY additional advantages in following aspects： the stronger electronegativity of cyano groups increase the affinity of GDY frameworks to lithium atoms; the larger atomic volume of methyl groups increases the interlayer distance and provide more storage space and diffusion tunnels. These results provide an inspiring idea to design and synthesize novel functionalized GDY based materials for high stability, high conductivity and controllable functionality.

Carbon allotropes composed of three kinds of hybrid carbons (including sp, sp^2 and sp^3 carbon) exhibit varied structure and properties, such as fullerenes,[1] carbon nanotubes,[2] graphene,[3] graphdiyne (GDY),[4] have been widely applied in the fields of drug delivery,[5] sensors,[6] biomaterials,[7] catalysis,[8] energy storage and conversion[9]. Recently, the structure modification of strategies such as doping heteroatoms and including surface covalently groups have been developed for adjusting the electronic structure and related properties of carbon allotropes.[10] For example, oxygen reduction catalytic performance has been significantly improved by doping high content of nitrogen in graphene, which can be prepared by wet chemical reaction or annealing.[11] Furthermore, the infinitely adjustable D–A systems built through surface modified graphene with electron donating or electron withdrawing molecules, providing more opportunities for the preparation of high efficiency solar cells.[12] However, it is hard to precise control the number and position of heteroatoms or groups following above methods, which limits the systematic regulation of the structures and properties of carbon materials. Therefore, it is necessary to develop a new method to tailor the structure and modulate the properties of carbon materials. Studying the relationship between the functionality of the group and the properties of the carbon material is not only conducive to the preparation of new carbon materials with excellent performance, but also opens up new platforms for the theoretical and experimental research of carbon materials.

Recently, facile approach is developed to prepare heteroatomic substituted GDY films through a bottom-up synthetic strategy, such as fluorine-substituted GDY[13] and nitrogen-substituted GDY[14]. At the same time, the electronic structure and lithium storage properties of GDY derivatives are significantly tuned by precisely modifying the number and position of heteroatoms. Compare to heteroatoms, the structure variability of functional groups is distinct, and the characteristic difference of introduced functional groups can be an important positive factor to realize the systematical adjustment of the partial properties of GDY, such as charge density distribution, interlayer distance and morphology, etc. [15] In detail, the electrode materials modified by the electron withdrawing or the electron donating groups can decrease or increase the oxidation-reduction potential, respectively.[16] For example, a strong electron withdrawing group, cyano group has a lower lowest empty molecular orbital (LUMO) energy level and higher electrochemical stability than other electron donating groups, thereby greatly improves the potential range and cycle stability for high energy metal ion batteries.[17] On the other hand, methyl group is an electron donating group that provides a wider charge/discharge platform and better cycle stability for the organic polymer negative electrode.[18]

In this work, via a bottom-up synthetic strategy, methyl group substituted GDY (MeGDY), hydrogen substituted GDY (HGDY) and cyano group substituted GDY (CNGDY) were synthesized successfully by choosing the pentaethynyl-benzyl, pentaethynyl-benzene and pentaethynyl-benzonitrile as monomers, respectively. Moreover, the effects of electron withdrawing and electron donating groups on the basic properties of carbon materials were systematically studied. The experimental results and first-principles calculations indicate that cyano groups with electron withdrawing ability can increase conductivity by reducing the band gap of GDY. In addition, the microstructure, interlayer

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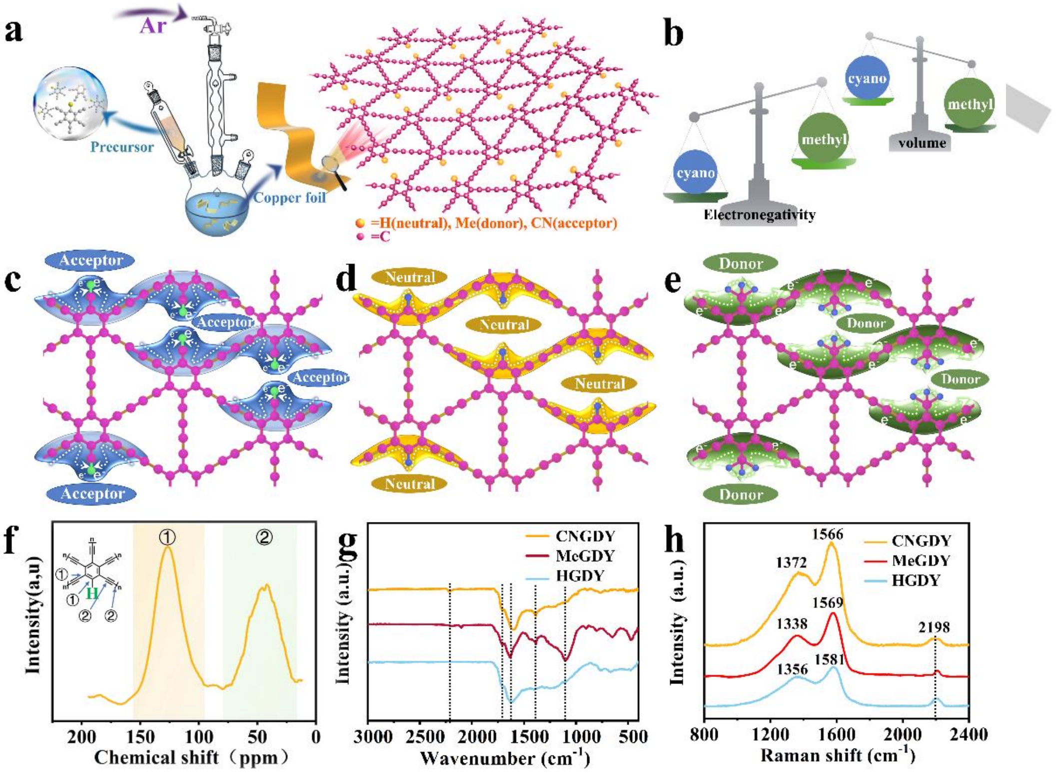

Figure 1. Synthetic route and characterizations of HGDY, MeGDY and CNGDY. (a) Synthetic route of HGDY, MeGDY and CNGDY. (b) Comparison of group properties, including electronegativity and volume. Chemical structure and electronic properties of (c) CNGDY, (d) HGDY, (e) MeGDY (Top to bottom). (f) Solid-state ^{13}C NMR spectrum of HGDY. (g) FT-IR spectrum of HGDY, MeGDY and CNGDY. (h) Raman spectrum of HGDY, MeGDY and CNGDY.

spacing and morphology of three GDY derivatives were found to be related to the size and coplanarity of the groups. At the same time, three GDY derivatives worked as anode material assembled lithium-ion battery showed a regular improvement in electrochemical properties, demonstrating the favorable regulation of the group on the properties of GDY. CNGDY exhibits the best electrochemical performance when applied to a negative electrode for lithium ion batteries. As a result, the high reversible capacity of 1612 mAh g^{-1} was achieved at a current density of 50 mA g^{-1}, and maintained more than 495 mAh g^{-1} at a current density of 2 A g^{-1} after 6000 cycles.

HGDY, MeGDY and CNGDY films are prepared on the surface of copper foil using group modified precursors, which are synthesized by simple chemical procedures including connecting alkyne and desiliconization (Figure 1a and Figure S1). As shown in Figure 1b, there is a clear difference between cyano and methyl groups in the aspects of electronegativity and group volume, so that the electronic structure and layer spacing of these GDY derivatives can be effectively tuned. Moreover, Figure 1c-e shows the electron transfer around the group and the adjacent benzene ring. It is expected that the electron density on the benzene ring will be decreased and increased after modifying the electron withdrawing group (cyano) and the electron donating group (methyl) on GDY planar, respectively. As shown in Figure 1f and Figure S2, the ^{13}C solid-state nuclear magnetic resonance (NMR) indicates the chemical structure of HGDY, MeGDY and CNGDY. Three GDY derivatives have two identical broaden peaks at 132 ppm and below 100 ppm, which correspond to $C(sp^2)$-$C(sp^2)$ on the benzene ring and the carbon atoms of butadiyne (including $C(sp)$-$C(sp^2)$ and $C(sp)$-$C(sp)$), respectively.[19] The broaden peaks below 100 ppm maybe due to the coexistence of multiple configurations in GDY derivatives. Precursors of three GDY derivatives with low molecular symmetry lead to complex coupling modes, thus producing a variety of configurations of GDY during the polymerization process. However, under the bottom-up strategy, the groups will be evenly distributed over the GDY planar. Notably, the peaks located around at 30 ppm (Figure S2g) and 126.6 ppm (Figure S2h) are the characteristic peaks of carbon atoms attached to the methyl group and the cyano group.[20] The carbon and nitrogen species were further characterized by X-ray photoelectron spectroscopy (XPS) (Figure S3-S5), which confirmed the components and structures of three GDY derivatives. FT-IR and Raman spectra are also used to characterize the components and structures of three GDY derivatives (Figure 1g and 1h). In their structures, adjacent aromatic rings are connected by diacetylenic bonds, while a group replaces one acetylenic bond on any aromatic ring (Figure 1a). The detailed XPS, FT-IR and Raman analysis are shown in Supporting Information.

With respect to the influence of different groups upon the microstructure of GDY, the morphologies and crystal structures of three GDY derivatives were characterized and discussed in

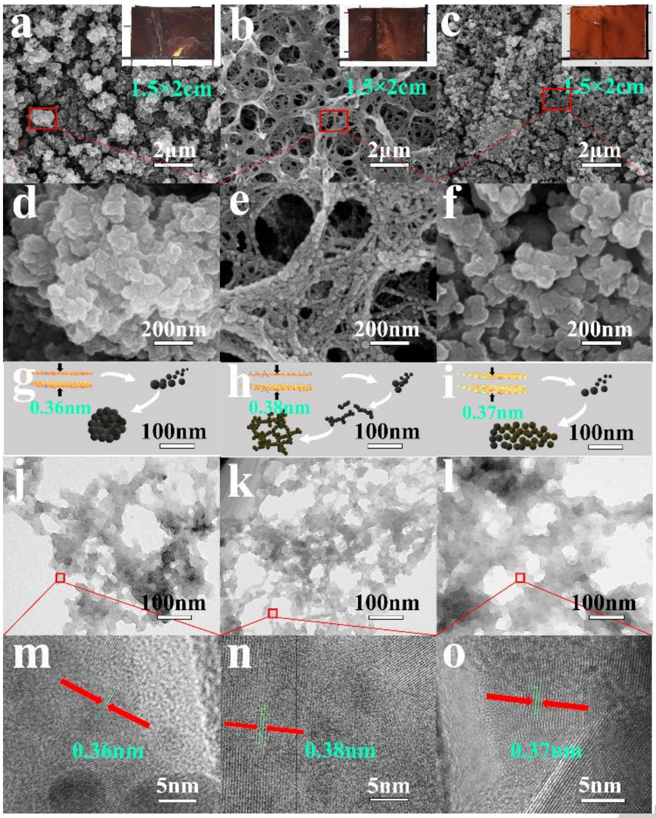

Figure 2. The top-view SEM images of (a) HGDY, (b) MeGDY and (c) CNGDY. The magnified SEM images of (d) HGDY, (e) MeGDY and (f) CNGDY. The formation process of (g) HGDY, (h) MeGDY and (i) CNGDY. The TEM images of (j) HGDY, (k) MeGDY and (l) CNGDY. The high-resolution TEM images of (m) HGDY, (n) MeGDY and (o) CNGDY.

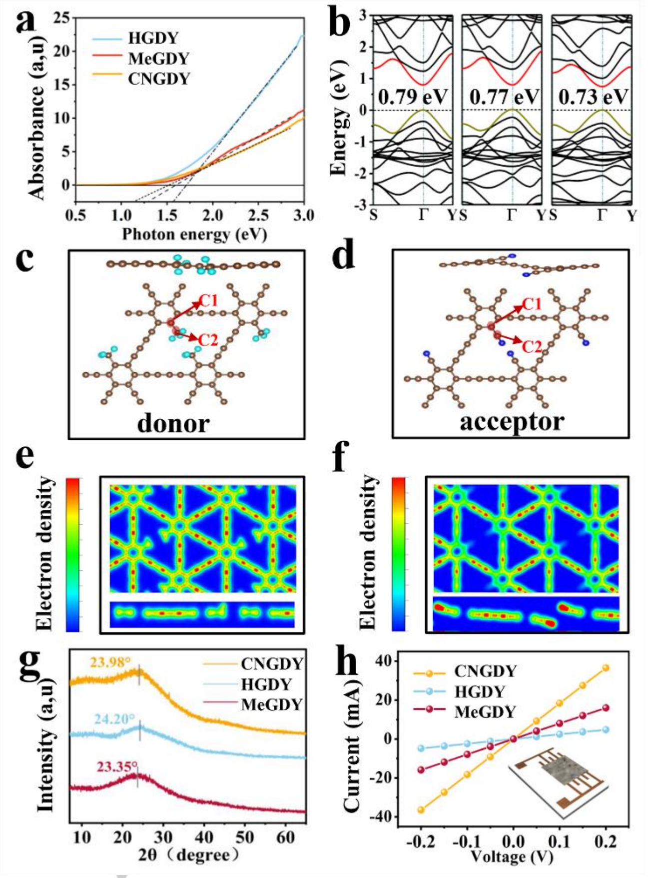

Figure 3. (a) UV-vis absorption spectrum of HGDY, MeGDY andCNGDY and plots of (Ahv)^2 versus photon energy (hv) (inset). (b) The Band structures of HGDY, MeGDY and CNGDY along the S-Γ-Y direction, respectively. The optimized configurations of (c) MeGDY and (d) CNGDY. The charge densities of (e) MeGDY and (f) CNGDY, the accumulation and loss of charge are represented by red and blue regions, respectively. (g) The X-ray diffraction patterns of samples. (h) The conductivity of different samples.

detailed, As shown in Figure 2a-f and S6, the scanning electron microscopy (SEM) images clearly show different morphologies about three GDY derivatives, all of them are composed of nanoparticles of different sizes. It is expected that the change in morphology is attributed to the size, coplanarity and electronegativity of groups. Figure S7 showed the diameter distribution of nanospheres of GDY derivatives under micro conditions. Subsequently, the simulated formation processes of microstructures are shown in Figure 2g–i. The electron donating group represented by methyl group not only reduces the size of the MeGDY nanospheres, but also provides an ordered porous structure by limiting the growth direction of the MeGDY nanospheres (Figure 2h). The electron withdrawing group represented by a cyano group tends to inhibit the aggregation of CNGDY nanospheres (Figure 2i).

Transmission electron microscopy (TEM) images present uniform films for HGDY, MeGDY and CNGDY, confirming the regular structure of GDY derivatives. Furthermore, layer spacings of GDY derivatives were obtained by high-resolution transmission electron microscopy (HRTEM) and X-ray diffraction (XRD). As shown in Figure 2m-o, HRTEM shows that the layer spacing of HGDY, MeGDY and CNGDY are 0.36nm, 0.38nm and 0.37nm, respectively. The SAED images (Figure S8) indicating the as-prepared GDY derivatives might contains polycrystal. At the same time the results of X-ray diffraction XRD (Figure 3g) are consistent with HRTEM, according to the different peak angles of three material (HGDY:MeGDY:CNGDY = 23.35°:24.20°:23.98°). Therefore, larger group could increase the layer spacing of GDY by a simple replacement process. Notably, the methyl provided the largest layer spacing for GDY, which can be attributed to the larger volume and non-planarity. The specific surface areas of HGDY, MeGDY and CNGDY powder calculated by nitrogen adsorption and desorption isotherms are 207 m^2/g, 559 m^2/g and 636 m^2/g, respectively (Figure S9). This confirms that the microstructure can be adjusted by changing the size and electronegativity of the group, predicting that unexpected results can be achieved in some areas. Moreover, three GDY derivatives exhibit a highly similar pore size distribution by the corresponding nitrogen adsorption-desorption isotherm (Figure S10).

As shown in Figure 3a and S11, the optical energy gap of HGDY, MeGDY and CNGDY, which calculated by ultraviolet-visible (UV-vis) and the formula of $\alpha \propto (hv - Eg)^{1/2}/hv$, were measured to be 1.73 eV, 1.62 eV and 1.53 eV, respectively (CNGDY< MeGDY < HGDY). As the optimized configurations shown in Figure 3 c-d, the methyl group is not planar since the angel of C1-C2-H is about 110° and the distance of C2-H is about

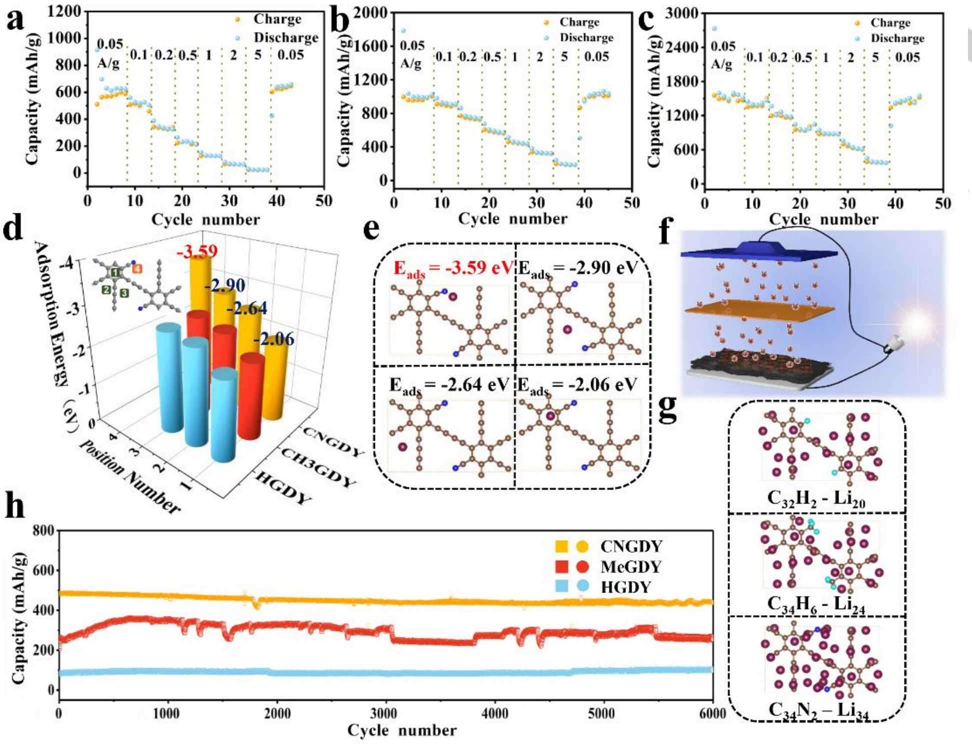

Figure 4. The electrochemical performance of HGDY, MeGDY and CNGDY electrodes in Li metal half-cell format. (a) (b) (c) The rate performance of HGDY, MeGDY and CNGDY, respectively. (d) The calculated adsorption energies of single Li occupied mode of different samples and corresponding storage sites. (e) The different adsorption configurations of single Li atom on the CNGDY with the adsorption energy marked. (f)The model of lithium ion battery. (g) The initial configurations of $C_{32}H_2$-Li_{20}, $C_{34}H_6$-Li_{24} and $C_{34}N_2$-Li_{34}, respectively. (h) The cycle performance of HGDY, MeGDY and CNGDY.

1.10 Å, which will observably increase the layer spacing. It is worth noting that the cyano group is collinear while the two cyano group in CNGDY protruded in up and down directions slightly due to the interaction of them. Therefore, the methyl group and the cyano group increased the layer spacing, which is consistent with the layer spacings of HGDY, MeGDY and CNGDY obtained from HRTEM are 0.36 nm, 0.38 nm, 0.37 nm, respectively.

To provide more insight difference of three samples, Figure 3e-f and Figure S12 show charge densities for different materials. Together with the bader analysis, the hydrogen and methyl group lose 0.12e and 0.13e while the cyano group gains 0.21e, confirming the cyano group has the high electronegativity as an electron withdrawing group while the methyl group and hydrogen have the low electronegativity as the electron donating group. The band structures of GDY modified with different groups along the high symmetry points Γ-X-S-Γ-Y-S are slightly different. The calculated direct band gaps of HGDY, MeGDY and CNGDY monolayers by PBE functional are predicted to be 0.79 eV, 0.77 eV and 0.73 eV, respectively (Figure 3b and S13), which is consistent with the changing trend of measured optical energy gap.[21] The narrower band gap will improve the conductivity, so it is often considered to provide better conductance and easier to overcome harsh experimental conditions. The current-voltage (I-V) curves of HGDY, MeGDY and CNGDY are in Figure 3h. the conductivity of HGDY and MeGDY are calculated by the conductivity formula (σ=L/RS) are about 1.14×10^{-2} s/m and 4.25×10^{-2} s/m while the conductivity of CNGDY can up to 5.41×10^{-1} s/m. Hence, the properties and performance for CNGDY should also be highly expected.

We further studied the lithium storage of three GDY derivatives as the LIB anode, including lithium storage mechanism, rate performance and cycle performance, etc. Figure 4a and 4b indicate the charge/discharge capacities of HGDY/MeGDY are 510.7/915.5 mAh g^{-1} with 55.7%, 997.3/1785.1 mAh g^{-1} with 55.87%, respectively. However, the first charge/discharge capacities of CNGDY are 1612 / 2420 mAh g^{-1} with the Coulombic efficiency (CE) of 66.6% (Figure 4c). It is notable that the electron withdrawing group has a stronger lithium storage ability and a higher coulombic efficiency. In addition, when the current density was reduced from 5 A g^{-1} to 0.05 A g^{-1}, all three materials completely recovered to the initial reversible capacity. The stability of three GDY derivatives were further characterized by long term cycling performances at a high current density (5 A g^{-1}) (Figure 4e), CNGDY shows the highest specific capacity (495 mAh g^{-1}) and stability when as the anode. Furthermore, we analyzed the lithium storage behavior of GDY by the cycling performance, cyclic voltammetry curves and capacity voltage curves, which were discussed in Supporting Information

(Figure S14 and S15). Moreover, in Supporting Information, we used the first principles to calculate the lithium storage characteristics of three GDY derivatives, including the adsorption energy at different sites and maximum theoretical capacity of lithium (Figure 4d, e, g and S16). The excellent electrochemical properties of CNGDY are mainly attributed to the reasonable adjustment of the physical and chemical properties, including the microscopic morphology, specific surface area, pore size distribution and interlayer spacing. The result of experiment and first-principles DFT calculations demonstrated the reduction of band gap (HGDY, 1.73 eV; MeGDY, 1.62 eV; CNGDY, 1.53 eV), the increase of volume or interlayer spacing （HGDY, 0.36 nm; CNGDY, 0.37 nm; MeGDY, 0.38 nm）, indicating a remarkable influence of group on the Li-storage capabilities. Expectedly, CNGDY with the stronger electronegativity ultimately increased the Li-storage capabilities through the increased reaction sites and the improved electron distribution environment. Moreover, MeGDY with larger interlayer distance provide more space for the storage and diffusion of lithium atom and ion, respectively. However, the improved layer spacing also acts unstable factors while the electrochemical process going on, causing low cycle stability of MeGDY based electrode (Figure 4h).

In summary, we synthesized a series of GDY derivatives with groups such as hydrogen, methyl and cyano through a novel cross-coupling reaction using chemically modified precursors. With the introduction of different groups, the properties including energy gap, layer spacing and microstructure, etc. would alter naturally. Expectedly, the regulation of the basic properties of GDY by these groups is beneficial to observe the change of lithium storage capacity in terms of reversible capacity, cycle stability and rate performance. Meanwhile, compared to those of GDY, HGDY and MeGDY, the improved electrochemical properties of CNGDY are released. Accurate control of energy gap, electron mobility, layer spacing, crystalline packing and specific surface area impacts the performance of lithium-ion storage. This in-depth investigation provides an effective strategy for precise regulation the electronic structure and properties of carbon materials and would leads to novel types of highly efficient carbon-based materials for energy storage.

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Keywords: graphdiyne • cyano • methyl • groups • lithium Ion battery

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Entry for the Table of Contents

COMMUNICATION

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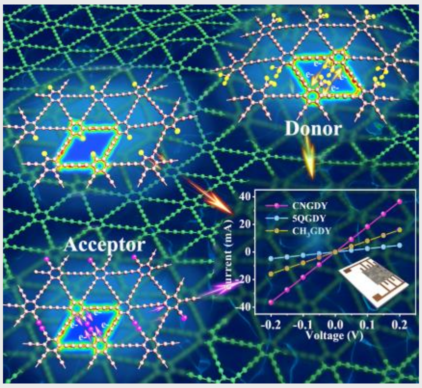

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Page No. – Page No.

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