Synthesis of carbon frameworks with N, O and S-lined pores from gallic acid and thiourea for superior CO₂ adsorption and supercapacitors

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ABSTRACT “C₂N”-species have emerged as a promising material with carbon-like applications in sorption, gas separation and energy storage, while with much higher polarity and functionality. Controlled synthesis of “C₂N” structure is still based on complex and less-sustainable monomers, which prohibits its broader industrial application. Here we report a class of well-defined C₂(N_xO_yS_z)₁ carbons with both high content of N/O/S heteroatoms and large specific surface area of up to 1704 m² g^{−1}, which can be efficiently synthesized through a simple additive condensation process using simple gallic acid and thiourea as the building blocks, without subtractive activation. This 1,4-para tri-doped C₂(N_xO_yS_z)₁ structure leads to sufficient CO₂ adsorption capacity (3.0 mmol g^{−1} at 273 K, 1 bar) and a high CO₂/N₂ selectivity (47.5 for a 0.15/0.85 CO₂/N₂ mixture at 273 K). Related to the polarity, the polar frameworks can be used as supercapacitor electrodes, with record specific capacitances as high as 255 F g^{−1} at 3.5 V for a symmetric supercapacitor in ionic liquid electrolyte. This work discloses a general way for preparing a novel family of multifunctional, high heteroatom-doped porous materials for various applications.

Keywords: carbon materials, structural pores, N/O/S doping, CO₂ adsorption, supercapacitor, ionic liquid

INTRODUCTION

Nanocarbons, including graphenes, carbon nanotubes, fullerenes and porous carbons, are used in a wide range of relevant fields, e.g., gas separation [1,2], electrocatalysis [3,4], and energy storage [5–7]. Particularly, porous carbon materials have the special advantages of large surface area, high porosity, good conductivity, high chemical and thermal stability, combined with potentially low costs [8]. Recently, many researchers have focused on developing

“designer carbons” with controlled local composition and structure. Functional sites for gas adsorption or energy carriers can be tailor-made, and the site density is much larger in designer carbons than that in carbons without defined local covalent structure.

“C₂N”-species is a subclass of porous carbonaceous frameworks with a nitrogen content of 33 mol%, and contains the maximum possible amount of structural pores, good chemical and thermal stability. Compared with amorphous activated carbon, we also find a rather well-defined structure. In short, there are two strategies for synthesizing “C₂N” using preorganized precursors. Mahmood *et al.* [9] reported the synthesis of C₂N holey two-dimensional (2D) crystal (C₂N-h2D) by a wet-chemical reaction between hexaaminobenzene trihydrochloride and hexaketocyclohexane octahydrate in *N*-methyl-2-pyrrolidone in the presence of a few drops of sulphuric acid or in trifluoromethanesulphonic acid. The resulting C₂N-h2D shows a 2D crystalline structure in surface layers which is a semiconductor, i.e., the bandgap was experimentally determined to be 1.96 eV, making it a potential photocatalyst for solar water splitting [10]. A field-effect transistor made by C₂N-h2D also shows an on/off current ratio of 10⁷. Fechner *et al.* [11,12] presented the use of a class of deep eutectic mixtures composed of hexaketocyclohexane and urea to synthesize a bulk C₂N structure. The resulting products exhibit lower overall order but well defined micropore structure at a high density, and the pores are tightly lined with pyrazinic nitrogen. Avoiding costly monomers which limit the practical applications of C₂N, we recently used simpler, more available, and sustainable starting synthons, gallic acid and urea, to produce N,O-dual doped C₂N_xO_{1–x} materials *via* direct ionothermal carbonization and ring

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closure [13]. The multifunctional material features an unusually high content of N/O heteroatoms (just below the theoretical limit 33 mol%) and a high specific surface area. The material also exhibits a defined microporosity, with pore size and shape well controlled by chemical condensation rather than etching. The introduced N/O in the carbon lattice allows varying the materials' properties, such as band positions, catalytic efficiency, chemical and thermal stability, and of course adsorption behavior [3].

Besides N and O, sulfur doping also provides valuable characteristics to the hosting carbon framework. A sulfur atom is much larger compared with O or N, so it easily protrudes out of the graphene plane, thus generating bent surface with unique properties, such as superconductivity [14]. However, precise S doping on a molecular scale and regularity is hard to realize. Here, we expand our previous "chemistry of simplicity" and prepare a new class of $C_2(N_xO_yS_z)_1$ using gallic acid and thiourea as monomers for controlled condensation (Scheme 1). Various functionalization scenes could be achieved by varying the synthetic temperature. Gallic acid is a well-known polyfunctional inexpensive natural chemical while simple thiourea serves as an effective nitrogen and sulphur source. The resulting $C_2(N_xO_yS_z)_1$ carbon materials (structure-defined ideally by 33 mol% heteroatom doping, but slightly below this value due to continued pore wall condensation, e.g., 29.5 mol%) are unusually polar, electrochemically active, and exhibit good adsorption and supercapacitance properties. This is due to not only the high specific surface area, but also the predominant paracheracter of the N/O/S doping patterns enabling simpli-

fied electronic communication between the single heteroatoms.

EXPERIMENTAL SECTION

Materials synthesis

Gallic acid (97.5%–102.5%) and thiourea (98%) were obtained from Sigma-Aldrich. Anhydrous zinc chloride ($ZnCl_2$, >97%) and hydrochloric acid (HCl, 1 mol L^{-1}) were purchased from Acros Organics and Merck KGaA, respectively. All the chemicals were used without any further purification.

Gallic acid (5 mmol), 5, 10, or 15 mmol of doping agent thiourea, and $ZnCl_2$ (in a weight ratio of precursor:salt of 1:10) were mixed, and the mixture was continuously heated to different temperatures (300, 400, 500, 800, 950°C) for 3 h under N_2 atmosphere. Once the samples were cooled down to room temperature, the resulting black carbon/salt product was washed with deionized water, HCl (1 mol L^{-1}) and ethanol, filtered and dried in vacuum at 80°C for 24 h.

All samples are named according to a simple code: GT1X-Y (G: gallic acid, T: thiourea, X: the molar ratio of thiourea:gallic acid, Y: preparation temperature).

Materials characterization

Fourier transformed infrared (FTIR) spectra were recorded on a Varian 600 FTIR spectrometer. Thermogravimetric analysis (TGA) was performed using a NETZSCH TGA 209 F1 device, in an air flow with a heating rate of 5°C min^{-1} . Scanning electron microscopy (SEM)

Scheme 1 Schematic illustration of the synthesis of the $C_2(N_xO_yS_z)_1$ for CO_2 adsorption and supercapacitors.

was carried out using an LEO 1550-Gemini microscope after sputtering with platinum. Energy-dispersive X-ray (EDX) investigations were detected by a Link ISIS-300 system (Oxford Microanalysis Group). X-ray diffraction (XRD) patterns were recorded on a Bruker D8 Advance instrument with Cu-K α radiation. High-resolution transmission electron microscopy (HRTEM) was performed on a JEOL ARM 200F instrument operated at 200 kV. Elemental analysis was accomplished as combustion analysis using a Vario Micro device. X-ray photoelectron spectroscopy (XPS) was investigated with a Thermo Scientific K-Alpha⁺ X-ray Photoelectron Spectrometer.

Pore structure properties of the samples were measured *via* nitrogen adsorption and desorption at 77 K using a volumetric technique on a Quantachrome Quadrasorb SI porosimeter. Prior to analysis, the carbons were degassed under vacuum at 200°C for 12 h. Brunauer-Emmett-Teller (BET) surface area was calculated in the relative pressure $P/P_0 < 0.2$. Total pore volume (V_t) was determined from the amount of nitrogen adsorbed at $P/P_0=0.995$. The pore size distributions of the carbons were obtained by quenched solid density functional theory (QSDFT) model with slit/cylindrical pore shape using nitrogen adsorption branch kernel.

CO₂ and N₂ adsorption measurements

CO₂ and N₂ adsorption were measured at pressures of up to 1 bar and 273 K (ice-water bath) on a Quantachrome Quadrasorb SI apparatus. Before analysis, the samples were degassed under vacuum at 200°C for 12 h. The CO₂/N₂ selectivity (S) (at 273 K, for CO₂/N₂ ratio of 15/85) was calculated by the ideal adsorption solution theory (IAST) and defined as

$$S_{\text{IAST}} = \frac{q_{\text{CO}_2}/q_{\text{N}_2}}{P_{\text{CO}_2}/P_{\text{N}_2}},$$

where q_{CO_2} and q_{N_2} are the molar loading of CO₂ and N₂, mmol g⁻¹; P_{CO_2} and P_{N_2} are the partial pressure of CO₂ and N₂, respectively.

Electrochemical measurements

The electrochemical performances of C₂(N_xO_yS_z)₁ were measured using a sandwich-type two-electrode testing cell at ambient condition. C₂(N_xO_yS_z)₁ electrodes in the form of round sheet were obtained by pressing a mixture film of 90 wt.% carbon materials and 10 wt.% poly(tetrafluoroethylene) (PTFE). The diameter of the electrode disk was 10 mm, and the areal density of each electrode was about 6 mg cm⁻². The 1-ethyl-3-methylimidazolium tetra-

rauoroborate (EMImBF₄) was used as electrolyte, two platinum foils were used as current collectors, and Dreamweaver Silver membrane was used as the separator. The two-electrode supercapacitors were assembled in an argon-filled glove box (H₂O < 0.1 ppm, O₂ < 0.1 ppm). A Biologic MPG-2 galvanostat/potentiostat was applied for electrochemical characterization. Cyclic voltammetry (CV) measurements were performed at sweep rates from 2 to 100 mV s⁻¹, from which the specific capacitance of the electrodes can be calculated according to the following equation

$$C = \frac{\int I dV}{v \times m \times V},$$

where I , V , v , and m represent the current, potential, scan rate, and the active mass in a single electrode. The galvanostatic charging/discharge tests were performed under different current densities between 0.1 and 5 A g⁻¹. The electrochemical impedance spectroscopy was performed at open-circuit potential with a sinusoidal signal over a frequency range from 20 kHz to 10⁻² Hz at an amplitude of 10 mV.

RESULTS AND DISCUSSION

Fig. S1 illustrates the synthesis of C₂(N_xO_yS_z)₁ from the condensation of gallic acid and thiourea in molten ZnCl₂. Gallic acid is considered as a promising unit for the synthesis of the ordered structure, because it can easily decarboxylate at higher temperatures to produce pyrogallol [15]. Therefore, the carboxylic acid unit can be seen as a “protecting/leaving group” to control preorganization, functionality, and N/O/S doping in the target carbon material. The three components together form a liquid crystalline, deep eutectic medium, very similar to Ref. [16], thus enabling a homogeneous starting situation. At the beginning, first massive foaming due to decarboxylation takes place. Thiourea decomposes at around 180°C under the formation of ammonia and thiocyanic acid, which both act as reactive linkers for the oligophenol [17–19]. Thiocyanic acid undergoes spontaneous thiocyanate formation with oxygen nucleophiles. The aromatic –OH groups can also be easily transformed into anilines and thiophenols, using the Lewis acid (ZnCl₂) catalyzed amination and Newman-Kwart-rearrangement. Simple thiourea is thereby expected to be an outstanding amination and thiolation agent for the condensation of –OH groups into oxazine, thiazine, and thiazole rings. The classical gallocyanine and methylene blue synthesis is using elemental sulfur for their very effective synthesis, resulting in the same structural motifs of

1,4-para-disubstitution, which comes from the minimization of ring energy of the condensing framework fragments, but with low molecular weight model compounds (Scheme S1) [20,21]. The resulting products are denoted as GT11-Y, GT12-Y and GT13-Y, with 11–13 indicating the molar ratio of gallic acid to thiourea and Y being the condensation temperature, respectively. The condensation of gallic acid and thiourea was confirmed by FTIR spectra of GT13-300, 400, 500 and 800 (Fig. S2). The two peaks at about 1040 and 1190 cm^{-1} , corresponding to C–O–C, C–N/S–C bonds [22], indicate the formation of oxazine, thiazine/thiazole rings. The samples could be burnt in oxygen with almost no residual mass at 700°C (Fig. S3) by TGA. The structure shows higher thermal stability than that of standard activated carbons. The high amount of heteroatoms incorporated in the structure resulted in an improvement in the oxidative stability, as reported in other Refs. [23,24]. The carbon morphology was visualized with SEM. GT13-300, 400 and 500 show aggregates of primary nanoparticles (Fig. S4). When the condensation temperature increases to 800°C , abundant mesopores can be observed in the GT13-800 (Fig. 1a). SEM elemental mapping images (Fig. 1b) show the homogeneous distribution of oxygen, nitrogen and sulphur, indicating the preservation of N/O/S functionality in GT13-800. To investigate the local ordering of the $\text{C}_2(\text{N}_x\text{O}_y\text{S}_z)_1$ structure, powder XRD and HRTEM were performed. The GT13-300 shows a broad feature centered

at $2\theta \approx 26^\circ$ (Fig. 1c), which is usually assigned to the stacking of aromatic systems [25]. This peak practically vanishes as the condensation temperature increases from 300 to 800°C , which means that the layer packing is fewer at high temperature than that at low temperature. HRTEM image (Fig. 1d) obtained from the GT13-800 reveals that the doping atoms are placed in relatively regular distances because of the band confinement where energy allows only most stable positions to be formed [3,26]. The bending of the layers visualizes that the aromatic systems with oxazine, thiazine/thiazole bridges are rather non planar and more undulated-2D.

Elemental analysis and XPS were used to investigate the carbon's chemical composition. All the samples have high content of N/O/S atoms (Table S1). Especially when the reaction is performed with gallic acid:urea in a 1:3 mole ratio at 500°C , the as-prepared GT13-500 has a stoichiometric composition of $\text{C}_2\text{N}_{0.43}\text{O}_{0.27}\text{S}_{0.14}$. The GT13-500 has thereby 29.5 mol% instead of 33 mol% of heteroatoms, which points to slightly ongoing wall condensation, as described before [11]. In spite of the simplicity of our starting products and the involved reaction cascade, the proximity to ideal behavior is, however, really remarkable and at least a positive surprise for us. H was bonded mainly with a secondary amine as shown in Fig. S1, which was confirmed by the N 1s spectrum. The overall condensation structure is also unusually thermally stable, and the amount of N/O/S is

Figure 1 (a) SEM image of GT13-800. (b) EDX mappings of carbon (red), nitrogen (green), oxygen (blue) and sulphur (yellow) for GT13-800. (c) XRD of the GT13-300, 400, 500, 800. (d) HRTEM image of GT13-800.

still near 30 mol% even after the sample is heated to 800°C.

The nature of the N/O/S species on the surface of carbon was further investigated using XPS characterization (Table S2). The N 1s spectrum can be deconvoluted into peaks located at 398.4 and 400 eV, which are assignable to a hydrogen bearing secondary amine in the para position of O/S and ternary nitrogen in aromatic rings, i.e., oxazine, thiazine/thiazole (Fig. 2a and b) [9]. Peaks for O 1s are found for the samples at 531.1, 532.2 and 533.4 eV corresponding to C=O, C–O–C, and O⁺, respectively (Fig. 2c and d). XPS spectra of the S 2p peak show that there are mainly three types of S, one with 163.8 eV, which can be assigned to the C–S–C in the

thiazine/thiazole rings [27], while the higher binding energy of 164.9 eV can be attributed to the presence of S–C–N binding motifs. Additionally, a broad peak centered at 168 eV indicates the presence of S⁺ or a sulfur oxide species. This peak is more prominent in the GT13-800 (Fig. 2e and f).

The synthesis scheme based on gallic acid and thiourea obviously creates aromatic systems with a (+)-charge at the O/S atom positions which is similar to many conjugated dyes in nature. Together with the total heteroatom content, these results indicate that N/O/S are integrated into the carbon framework, most probably in rings where both the 1,4-para positions in every second ring (note that the C₆ ring of gallic acid is expected to stay

Figure 2 Deconvoluted XPS spectra of the GT13-500 and GT13-800. (a, b) N 1s, (c, d) O 1s and (e, f) S 2p.

intact, while substitution and condensation reactions can fix six heteroatoms on this central, bridging to the next three phenyl cored by two heteroatoms, each). This simultaneous incorporation of N/O/S in a controlled manner with all heteroatoms facing micropores, indeed leads to a remarkable improvement of the performance in gas adsorption, electrocatalysis, and energy storage [28–30], and we will quantify the pore polarity and energy storage performance by gas sorption and supercapacitor experiments in the following. The specific surface area and pore size distribution of the obtained condensation products were evaluated by the nitrogen physisorption measurement at 77 K. (Fig. 3a and b, Figs S5–S8, Table S3). The low temperature condensation product GT13-300 has hardly any porosity accessible for nitrogen, while the isotherm of GT13-400 already exhibits typical type I character and reveals that it mainly has narrow micropores (< 1 nm), i.e., the structural framework pores have begun to form. With the increase of condensation temperature, the BET surface area (S_{BET}) of carbon increases from 516 to 798 $\text{m}^2 \text{g}^{-1}$ for GT13-400 and GT13-500. For GT11-500, GT12-500 and GT13-500, the additional rise at high pressure (P/P_0 close to 1) may be owing to the macropores formed between the primary, as-condensed particles, and the hysteresis loop indicates the

presence of bottleneck-mesopores. The proportion of mesopores decreases as the thiourea content increases, which means that higher thiourea content during condensation promotes the formation of more extended, well-defined microporosity with dense heteroatom lining. GT13-800 displays a high specific surface area of 1704 $\text{m}^2 \text{g}^{-1}$ with abundant micropores and mesopores (2–6 nm), which is due to the already discussed further wall condensation, but the resulting pore structure can provide a larger extent of binding sites and fast pore transport for the guest molecules.

To quantify the pore polarity, we also measured the CO_2 adsorption isotherms at 0°C for GT11-500, GT12-500 and GT13-500. Although the S_{BET} values of GT11-500 (1568 $\text{m}^2 \text{g}^{-1}$), GT12-500 (1065 $\text{m}^2 \text{g}^{-1}$) and GT13-500 (798 $\text{m}^2 \text{g}^{-1}$) decrease with the increase of thiourea to gallic acid mole ratio, the CO_2 adsorption capacities of them are similar at 1 bar. This means that the CO_2 is most effectively bound in the special structural framework pores, while the overall nitrogen surface area with its large outer surface area does not really contribute to the unusual and strong CO_2 binding. This sum balance of favourable structural pores overlayed by unspecific outer material surfaces was discussed in detail for a broad range of materials in a recent review [1].

Figure 3 (a, b) N_2 adsorption (filled symbols) and desorption (empty symbols) isotherms at 77 K and pore size distribution curves calculated by QSDFT for the GT13-300, GT13-400, GT13-500 and GT13-800. (c) CO_2 and N_2 adsorption isotherms of GT13-500 at 273 K. (d) CO_2/N_2 selectivity as calculated by the IAST method for a $\text{CO}_2/\text{N}_2=0.15:0.85$ gas mixture at 273 K.

GT13-500 exhibits an overall adsorption capacity of $66 \text{ cm}^3 \text{ g}^{-1}$ (3 mmol g^{-1}) (Fig. 3c). It should be pointed out that the CO_2 uptake of GT11-500, GT12-500 and GT13-500 are found to be 19.5, 21.2 and $24.1 \text{ cm}^3 \text{ g}^{-1}$ at 0.15 bar (Fig. 3c, Figs S9 and S10), i.e., the structural, heteroatom-lined micropores bind CO_2 rather strongly, with the CO_2 uptake at low pressure reflecting the affinity to CO_2 . In addition, the CO_2/N_2 selectivity for GT13-500 is up to 47.5 (Fig. 3d), which is significantly higher than most of the results reported for porous activated carbons, even those with “ordinary”, single site nitrogen doping [31–33]. The good CO_2 capture performance can therefore be attributed to the pores densely lined with heteroatoms and a rather defined micropore size predetermined by chemical condensation with a 12-covalent bond circumvented.

Given the polar character of these pores and their relative extension in relation with outer surface area, it is remarkably interesting to check the usefulness of our new material for application in supercapacitor electrodes. Here, high power density, long lifetimes, and rapid charge-discharge rates put high requests on the storage material [34,35]. The electrochemical performances of $\text{C}_2(\text{N}_x\text{O}_y\text{S}_z)_1$ were measured in a symmetric supercapacitor device by using ionic liquid (IL) of EMImBF₄ as the electrolyte. The CV curves and the specific capaci-

tances (calculated from CV curves) are displayed in Fig. 4a, b, and Figs S11–S13. The comparable lower performance of GT13-500 is mainly caused by its poor conductivity. Notably, GT13-800 shows the highest capacitance of 255 F g^{-1} at a scan rate of 2 mV s^{-1} . This is to our knowledge one of the highest specific capacitances ever reported for IL-based supercapacitors [36–40]. This high specific capacitance is attributed to the strong binding of electrolyte ions on the strongly polarized carbon surface in the presence of N/O/S. Furthermore, their well-developed micropores interact strongly with the electrolyte molecules, effectively enhancing the electrical double layer. On the other hand, the uniform mesopores provide the ion diffusion channel, which allows easy entry of the electrolytes into the inner micropores. Interestingly, further increase in condensation temperature to 950°C leads to the decrease of the performances, from 255 to 166.4 F g^{-1} , due to the loss of the important 1,4-para doped motif at this higher temperature. We take this as a proof that our material at 800°C is not only highly conductive, but also indeed makes the full use of its heteroatom content, while a usual N-doped carbon gained at higher temperatures cannot. As shown in Figs S14–S16, the specific capacitance at low scan rate of 2 mV s^{-1} increases from 193.5 to 255 F g^{-1} with increasing N/O/S content of the materials, which also indicates that

Figure 4 (a) CV curves of GT13-800 electrodes at different scan rates from 2 to 100 mV s^{-1} . (b) Specific capacitance values of GT13-800 calculated from CV curves at different scan rates. (c) Galvanostatic charge-discharge curves at various current densities from 0.1 to 5 A g^{-1} . (d) Nyquist plot of GT13-800.

the N/O/S functionalities are beneficial to the fast capture and efficient adsorption of IL electrolyte with a strengthened solid-liquid interfacial effect.

The CV curves of the GT13-800 display nearly rectangular shapes at different scan rates (Fig. 4a), underlining its high electronic conductivity. The observed peak at lower scan rates typically occurs during slow cycling in the IL electrolyte and was recently attributed to structural transitions of ions inside the micropores during charging/discharging [41–43]. The galvanostatic charging/discharge curves of GT13-800 are nearly symmetric (Fig. 4c), and the Nyquist plot exhibits a nearly vertical line in the low-frequency region (Fig. 4d). These results confirm that $C_2(N_xO_yS_z)_1$ (especially, GT13-800), in spite of its synthesis featured by simplicity and sustainability, is a very promising energy-storage material. This is attributed to the strong binding of electrolyte ions on the strongly polarized carbon surface promoted by the N/O/S heteroatoms, which continues and propagates recent observations in Refs. [44–46].

CONCLUSIONS

In summary, we successfully synthesized a new class of carbon-based microporous materials, $C_2(N_xO_yS_z)_1$, using gallic acid and thiourea as simple and available building blocks. The pores are formed due to structural condensation rather than subtractive etching processes, and the resulting structure is thermally stable and shows extraordinarily high N/O/S heteroatom content (<33 mol%) and a large specific surface area ($1704 \text{ m}^2 \text{ g}^{-1}$). Especially the high sulfur content which can be kept to comparably high temperatures is remarkable and rather unusual for classical microporous carbons. Owing to the structural porosity and heteroatom-controlled polarizability, the GT13-500 exhibits reasonably high CO_2 uptake of 3 mmol g^{-1} , meanwhile at an excellent CO_2/N_2 selectivity of 47.5 (273 K). The GT13-800 delivers in IL based-supercapacitor experiments a record high capacitance of 255 F g^{-1} @3.5 V. Considering monomer sustainability, feasibility of synthesis as well as the very promising figure of merit, this newly described carbon framework system can be scaled up and is suitable for a broad range of other energy and environmental applications.

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Author contributions Tian Z and Antonietti M conceived the idea and cowrote the paper. Tian Z and Lai F designed and performed the experiments and analyzed the results. Tobias H performed the HRTEM tests. All the authors discussed the results and commented on the manuscript.

Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Experimental details and supporting data are available in the online version of the paper.

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没食子酸和硫脲合成具有N, O和S排列孔的碳骨架用于高效CO₂吸附和超级电容器

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摘要 “C₂N”是一种很有前途的材料, 在吸附、气体分离和能量存储方面具有类似碳的应用, 且具有更高的极性和功能性。然而, “C₂N”的可控合成仍基于复杂且不可持续的单体, 阻碍了其更广泛的工业应用。本文报道了一种具有特定结构的C₂(N_xO_yS_z)₁碳材料, 即以简单的没食子酸和硫脲为结构单元, 通过简单的加成缩合制备, 无需活化。所制备的碳材料具有高的N/O/S杂原子含量和大的比表面积(高达1704 m² g⁻¹)。此1,4-对位三掺杂孔结构使得C₂(N_xO_yS_z)₁具有较好的CO₂吸附能力(在273 K, 1 bar下为3.0 mmol g⁻¹)和较高的CO₂/N₂选择性(在273 K, CO₂/N₂=0.15/0.85下为47.5)。此外, 由于碳骨架高极性的特征, 其在以离子液体为电解质的对称超级电容器中展现了高达3.5 V的电势窗口和255 F g⁻¹的比容量。本工作为制备新型多功能、高含量杂原子掺杂多孔材料提供了一种通用的方法。