

RESEARCH ARTICLE

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Ultrafast preparation of honeycomb N-doped carbon via flash joule heating for efficient capture of C₂H₄F₂ refrigerant

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Abstract

Developing advanced adsorbents for efficient capture of phase-out hydrofluorocarbons (HFCs) is imperative for reducing emissions of deleterious compounds (e.g., 1,1-difluoroethane, C₂H₄F₂). Here, honeycomb N-doped carbons (NPC-FJH) with hierarchical porosity were rapidly prepared (~20 s) using flash Joule heating (FJH) for C₂H₄F₂ adsorption. The structural and adsorption properties of NPC-FJH were compared with NPC-TF obtained via tubular furnace (TF). NPC-FJH have remarkable C₂H₄F₂ uptake capacities (12.37 mmol g⁻¹), selectivities (up to 420) at 25°C and 100 kPa than their NPC-TF counterparts. Density functional theory (DFT) revealed that N and adjacent C atoms enhance host-guest interactions. Moreover, NPC_{2.5}-FJH₈₀₀ demonstrates ultrafast adsorption kinetics (99 s to saturation) and a favorable diffusion coefficient ($D/r^2 = 3.49 \text{ ms}^{-1}$). Breakthrough tests demonstrated that NPC_{2.5}-FJH₈₀₀ achieved the selective separation of C₂H₄F₂ in ternary gas mixtures (N₂/C₂H₂/C₂H₄F₂), with negligible capacity loss over 10 cycles, demonstrating its excellent reusability and significant potential for practical applications.

KEYWORDS

absorption separation, C₂H₄F₂ capture, DFT, flash joule heating, novel prepared porous carbons

1 | INTRODUCTION

Over the past decades, hydrofluorocarbons (HFCs) have been considered as transitional substitutes to chlorofluorocarbons in industrial refrigeration applications due to their lower ozone depletion potential.^{1–3} Among them, 1,1-difluoroethane (C₂H₄F₂) has garnered particular attention within the industry owing to its favorable thermophysical properties and negligible ozone depletion potential, which supports its widespread deployment in various refrigerant systems.^{4,5} However, the global warming potential of HFCs has gradually aroused vigilance in the eco-environment protection field, with growing global concerns regarding greenhouse gas emissions. For example, C₂H₄F₂ has a global warming potential of ~138, which is over two orders of

magnitude higher than that of carbon dioxide (CO₂).⁶ This feature implies that there is a disproportionate climate impact per unit mass of C₂H₄F₂. In response, regulators in the European Union and the United States have mandated the phase-out of HFCs in favor of next-generation refrigerants such as hydrofluoroolefins, which exhibit low global warming potential and zero ozone depletion potential.⁷ Currently, the global inventory of in-use C₂H₄F₂ refrigerant is estimated at 2.8 million tons, which makes the development of eco-friendly treatment technologies critically urgent.⁴ In fact, it is preferable to separate and recycle C₂H₄F₂ rather than vent or incineration. However, C₂H₄F₂ separation still faces technical challenges because C₂H₄F₂ often coexists with gas mixtures (e.g., nitrogen, acetylene) that have comparable physicochemical properties in actual situations. Thus, the development of efficient reclamation technologies is a key step toward sustainable refrigerant management.

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To date, porous adsorbents such as metal organic frameworks and porous organic cages have been explored for HFCs adsorption and separation.^{8–12} For example, under higher pressure (488 kPa) and 15°C condition, a three-dimensional honeycomb MOF-1 showed a 1,1,1,2-tetrafluoroethane ($C_2H_2F_4$) uptake capacity of 1.10 g g^{-1} ($\sim 10.78\text{ mmol g}^{-1}$).¹¹ Similarly, Ogoshi et al. reported that pillar[6] arene-based porous organic cages selectively adsorbed 1,1,1,2,2-pentafluoroethane (C_2HF_5) with only a capacity of $\sim 1.27\text{ mmol g}^{-1}$ at 100 kPa and 25°C.¹² However, such adsorbents are often constrained by challenges including complex multi-level spatial structures, tedious synthesis routes, and limited reusability. In contrast, nitrogen-doped porous carbons (NPC) have emerged as attractive alternatives for HFCs or greenhouse gases capture, featuring facile and economical preparation and excellent recyclability.^{13–16} Importantly, nitrogen doping modifies the local electronic environment of adjacent carbon atoms, resulting in enhanced electrostatic and hydrogen-bonding interactions with polar fluorinated gases such as $C_2H_4F_2$.¹⁷ Therefore, the rational design and optimization of high-performance NPC-based adsorbents may provide a promising strategy for sustainable $C_2H_4F_2$ reclamation and resource recovery.

At present, various strategies for the preparation of NPC adsorbents, among which traditional tubular furnace (TF) pyrolysis remains widely employed.¹⁸ Nonetheless, the TF method typically suffers from several inherent problems, including prolonged thermal treatment durations, limited heat transfer efficiency, and sparse heteroatoms doping within the carbon framework, which hinder the performance of NPC adsorbents. This is because the heat transfer nature of the TF method relies on external heat conduction from surface inward of the calcined precursors.¹⁹ The pronounced temperature gradients between the core and periphery slow the carbonization kinetics of the internal zones, thereby necessitating prolonged thermal treatment. By contrast, flash Joule heating (FJH), which applies ultrafast thermal processing via high-intensity electrical pulses, induces rapid carbonization of precursors on the mere seconds timescale.^{20–22} Meanwhile, Guo et al.²³ reported that this rapid thermal process has a distinctive thermal profile wherein the internal regions of the material undergo a more rapid temperature rise than the surface, thereby promoting more uniform internal carbonization and imparting the resulting NPC adsorbents with unique structural and functional attributes. However, the application of NPC adsorbents prepared by FJH in selective $C_2H_4F_2$ adsorption remains largely underexplored.

Herein, we report the rapid preparation of a series of honeycomb NPC-FJH adsorbents via FJH and use them for $C_2H_4F_2$ adsorption, wherein carbonization and activation of the precursors are achieved in the single step within 20 s. In addition, to elucidate the advantages of FJH, comparative analyses were conducted against benchmark NPC-TF samples prepared via TF. The precursor comprised biomass-derived glucose, urea, and potassium carbonate, serving respectively as the carbon source, nitrogen source, and activator. By modulating the activator dosages and carbonization parameters, NPC-FJH with hierarchical porosity, high specific surface area, and well-distributed adsorption sites were obtained. Furthermore, the adsorption mechanism was investigated through DFT and NBO calculations.

Additionally, the adsorption behaviors of NPC-FJH were analyzed using the Langmuir–Freundlich adsorption model, while the diffusion coefficient of NPC-FJH was calculated from $C_2H_4F_2$ adsorption kinetics by correlating with the Fick model. Finally, hydrocarbon/nitrogen selectivity, dynamic breakthrough performance, and adsorbent reusability were also studied to evaluate the industrial viability of NPC-FJH for gas separation applications.

2 | EXPERIMENTAL

2.1 | Materials

D-glucose (AR), urea (99%), and hydrochloric acid (HCl, 36%) were purchased from Shanghai Macklin Biochemical Co., Ltd. Potassium carbonate (K_2CO_3 , 99%) was obtained from Shanghai Titan Scientific Co., Ltd., and anhydrous ethanol (98%) was provided by Shanghai Wohua Chemical Co., Ltd. The adsorption gases of nitrogen (N_2 , 99.999%), ethane (C_2H_6 , 99.99%), ethylene (C_2H_4 , 99.99%), acetylene (C_2H_2 , 99.99%), and argon (Ar, 99.999%) were supplied by Jiangxi Huahong Special Gas Co., Ltd., while helium (He, 99.999%) was sourced from Nanchang East China Special Gas Co. Ltd. 1,1-difluoroethane ($C_2H_4F_2$, 99.99%) and the binary gas of $C_2H_4F_2/N_2$ (v:v, 85:15) were purchased from Dalian Special Gases Co., Ltd. All experimental reagents and solvents were of analytical grade and not further purified.

2.2 | Preparations

In this work, a series of NPC samples were prepared via two distinct methods: rapid FJH and traditional TF treatment. The resulting samples are designated as NPC_X-FJH_T and NPC_X-TF_T, where X refers to the mass ratio of K_2CO_3 to D-glucose, and T represents the final carbonization temperature. Detailed operational programs of FJH and TF are provided in the Supporting Information S1.1. In a typical preparation of NPC_{2.5}-FJH₈₀₀, urea (2 g), D-glucose (2 g) and K_2CO_3 (5 g) were dissolved in 30 mL of deionized water in a round-bottom flask and the reaction system was stirred at 80°C for 5 h. Then, the water was removed using a rotary evaporator to obtain a brown viscous precursor. Next, the above precursor was transferred into a graphite-pressed sample boat ($10 \times 2 \times 0.2\text{ cm}$) for FJH treatment, and physical images are provided in Figure S1. Subsequently, the system was heated to the target temperature (800°C) within 20 s under a preprogrammed protocol. After cooling to room temperature, a black honeycomb-like solid was obtained. Finally, the resulting samples were washed with HCl (2 M, $3 \times 20\text{ mL}$) and deionized water ($5 \times 50\text{ mL}$) to remove residual K_2CO_3 or inorganic impurities, followed by drying in a vacuum oven at 80°C for 8 h. Several other NPC_X-FJH_T samples were synthesized by a similar process. In addition, NPC_X-TF_T samples were prepared via TF at identical reactant ratios and carbonization temperatures as the corresponding NPC_X-FJH_T samples. The preparations of selected reference porous samples

(e.g., N-free porous carbon (PC_{2.5}-FJH₈₀₀), graphene oxide-derived carbon (GO-JFH₈₀₀), N-doped graphene oxide (NGO-JFH₈₀₀), Polydivinylbenzene (PDVB), representative metal-organic framework (Zeolitic Imidazolate Framework-8, ZIF-8)) are summarized in the Supporting Information S1.2. Additionally, details of characterizations and corresponding instrumental parameters are presented in the Supporting Information S1.3.

3 | RESULTS AND DISCUSSION

FJH was selected for the preparation of the porous carbon host due to the following reasons: (I) It provides ultrafast thermal shock that enables the concurrent carbonization and heteroatom doping processes within several seconds, which makes it unnecessary to perform

complex pre-treatment steps; (II) It allows for the synthesis of porous activated carbon samples with tunable porosity and structural features, which enhances adsorption capacities for various gases molecules; (III) it offers scalability for the production of large quantities of porous carbon with uniform physicochemical characteristics, which is critical for industrial deployment.

3.1 | Structure and texture properties

Based on the above, the rational utilization of the instantaneous thermal shock induced the carbonization-activation process to produce carbon-based C₂H₄F₂ adsorbents. The schematic diagram of the preparation pathways for NPC_X-FJH_T and NPC_X-TF_T samples via FJH and TF, respectively (see Figure 1A). The SEM images (Figure 1B,C) clearly

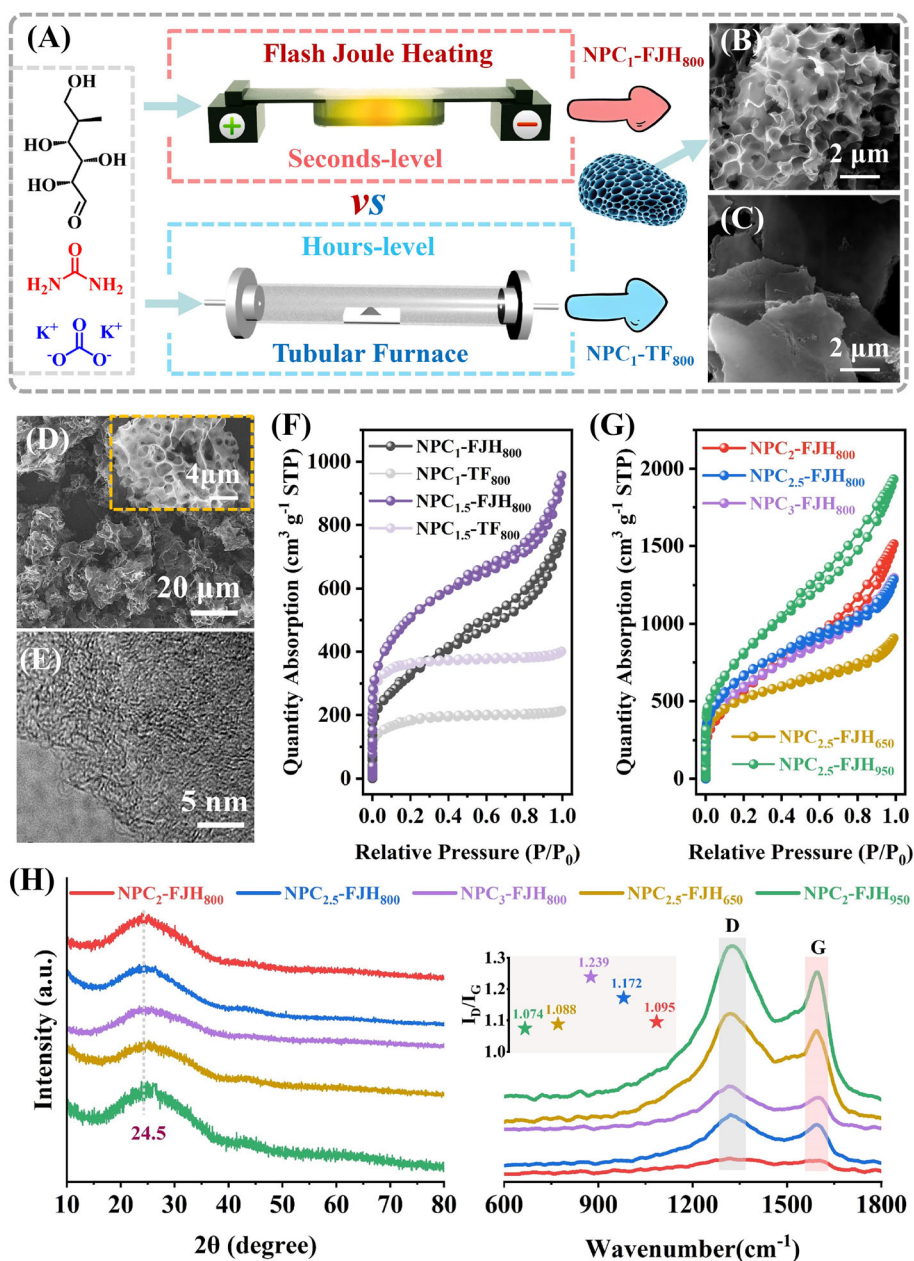


FIGURE 1 (A) Comparison schematic diagram of the preparation pathways of NPC₁-FJH₈₀₀ and NPC₁-TF₈₀₀ samples using FJH and TF; SEM image of (B) NPC₁-FJH₈₀₀ and (C) NPC₁-TF₈₀₀; (D) SEM and (E) TEM images of NPC_{2.5}-TF₈₀₀; (F, G) N₂ adsorption-desorption isotherms of (b) NPC_X-FJH_T and NPC_X-TF_T samples; (H, left) XRD patterns, (H, right) Raman spectra and (H, inset) intensity ratios of NPC_X-FJH_T sample.

show the morphological differences between the NPC₁-FJH₈₀₀ and NPC₁-TF₈₀₀ samples. It can be observed that the NPC₁-FJH₈₀₀ sample presents a honeycomb-like three-dimensional network structure, which means that it has rich porosity characteristics. For comparison, NPC_X-TF_T samples were obtained via TF at a ramp rate of 8°C min⁻¹ to 800°C for carbonization and activation. The resulting NPC₁-TF₈₀₀ sample displays a thick sheet-like morphology with smooth surfaces. Furthermore, NPC_{2.5}-TF₈₀₀ samples obtained with higher activation dosages have similar honeycomb structures (Figure 1D), while TEM image (Figure 1E) confirmed the presence of porous structures with thin pore walls. The N₂ adsorption-desorption curves (Figure 1F) of NPC_X-FJH₈₀₀ and NPC_X-TF₈₀₀ (X = 1 and 1.5) were tested, and the relevant structural data are calculated and summarized in Table S1. Notably, surface areas and pore volumes of NPC₁-TF₈₀₀ (580.6 m² g⁻¹, 0.249 cm³ g⁻¹) and NPC_{1.5}-TF₈₀₀ (1111.6 m² g⁻¹, 0.172 cm³ g⁻¹) were significantly lower than those of the corresponding NPC_X-FJH_T (NPC₁-FJH₈₀₀ (1156.3 m² g⁻¹, 1.070 cm³ g⁻¹) and NPC_{1.5}-FJH₈₀₀ (1738.2 m² g⁻¹, 1.150 cm³ g⁻¹)) samples prepared under the same conditions. Overall, the differences mentioned above are attributed to the instantaneous achievement of elevated internal temperatures within the raw materials via the FJH, which causes outward expansion from interior of the raw material. In this process, the K₂CO₃ rapidly decomposes and reacts with carbon to release gaseous products, which promotes rapid expansion and rupture of the carbon matrix, ultimately yielding interconnected honeycomb-like pore structures. In contrast, the TF method transfers heat from the exterior to the interior, resulting in delayed internal reactions and forming dense block-like mesoporous structures. Besides, the comparison results suggest that a controlled increase in activator dosage improves the surface area and pore volume of NPC₁-FJH₈₀₀ and NPC_{1.5}-FJH₈₀₀, which may promote more efficient mass transport of the adsorbates.

Subsequently, to evaluate the effects of the activator dosages and pyrolysis temperatures, NPC_X-FJH_T (NPC₁-FJH₈₀₀, NPC₂-FJH₈₀₀, NPC_{2.5}-FJH₈₀₀, NPC₃-FJH₈₀₀, NPC_{2.5}-FJH₆₅₀ and NPC_{2.5}-FJH₉₅₀) samples were synthesized under varying conditions, with structural parameters summarized in Tables 1 and S1. SEM images of NPC_X-FJH_T samples are shown in Figures 1B–D and S2. It can be found that all NPC_X-FJH_T samples exhibit a characteristic honeycomb-like structure morphology, which supports the excellent generality and stability of the FJH process. Notably, elevated pyrolysis temperatures further contributed to the development of larger surface area, which suggests

that high-temperature thermal shock is instrumental in maximizing surface area of NPC samples. Meanwhile, the specific surface area reached a maximum value of 2930.2 m² g⁻¹ for NPC_{2.5}-FJH₉₅₀, followed by a gradual decrease with further increases in activator dosage. Moreover, the N₂ adsorption-desorption isotherms of NPC_X-FJH_T samples revealed the Type IV hysteresis loop, indicating the presence of a hierarchical porous structure within the NPC_X-FJH_T samples (see Figure 1G).²⁴ By comparison, NPC_X-TF₈₀₀ samples displayed Type I hysteresis loops (Figure 1F), characteristic of microporous carbons, which indicated that the FJH method was more inclined to form a hierarchical pore structure. In addition, activator dosage was also found to significantly affect surface area, particularly at elevated pyrolysis temperatures, as exemplified by the substantial increases observed in NPC_{2.5}-FJH₉₅₀. Additionally, the N₂ adsorption-desorption isotherms of the porous materials selected (PC_{2.5}-FJH₈₀₀, GO-JFH₈₀₀, NGO-JFH₈₀₀, PDVB, ZIF-8) as control groups are shown in Figure S3, with their specific surface area and pore parameters listed in Table S1. It is evident that the porous characteristics of these materials are consistent with those reported in the literature. As expected, XRD patterns revealed that NPC_X-FJH_T samples have broad (002) diffraction peaks centered at 24.5°, which are consistent with their amorphous nature (Figure 1H, left).¹⁵ The structural properties of NPC_X-FJH_T samples were examined using Raman (Figure 1H, right). The results show that NPC_X-FJH_T samples displayed prominent D bands and G bands at around 1350 cm⁻¹ and 1580 cm⁻¹, which correspond to disordered sp³ carbon and graphitic sp² domains, respectively.²⁴ The calculated intensity ratio (I_D/I_G) result (Figure 1H, inset) reveals that a monotonic increase with increasing dosage but exhibits a non-monotonic trend with respect to pyrolysis temperature, decreasing at higher temperatures. In short, these findings highlight the efficacy of the FJH strategy in the preparation of honeycomb-like three-dimensional network of NPC materials with tunable porosity and enhanced surface area.

3.2 | C₂H₄F₂ adsorption capacity and contribution analysis

The gases separation potential of NPC_X-FJH_T samples were evaluated by measuring their C₂H₄F₂ adsorption isotherms at 25°C and 100 kPa (see Figure 2A–C) and the operational details of the adsorption tests

TABLE 1 The structural features of NPC_X-FJH_T samples and their C₂H₄F₂ uptake capacities and IAST selectivities at 25°C and 100 kPa.

Samples	S _{BET} (m ² g ⁻¹)	V _p (cm ³ g ⁻¹)	Elemental analysis (wt%)			C ₂ H ₄ F ₂ uptake (mmol g ⁻¹)	Sel ^a (IAST)
			C	N	O		
NPC ₂ -FJH ₈₀₀	2154.2	2.317	71.04	4.49	21.45	10.96	211.84
NPC _{2.5} -FJH ₈₀₀	2345.6	1.769	66.54	4.22	25.95	12.37	223.70
NPC ₃ -FJH ₈₀₀	2134.9	1.806	70.94	4.98	22.06	9.18	217.21
NPC _{2.5} -FJH ₆₅₀	1729.4	1.248	70.90	5.84	20.23	10.70	322.12
NPC _{2.5} -FJH ₉₅₀	2930.2	2.729	52.06	5.36	38.71	7.59	185.84

^aIdeal Adsorbed Solution Theory (IAST) binary-gas adsorption selectivity (C₂H₄F₂/N₂, v/v = 85/15).

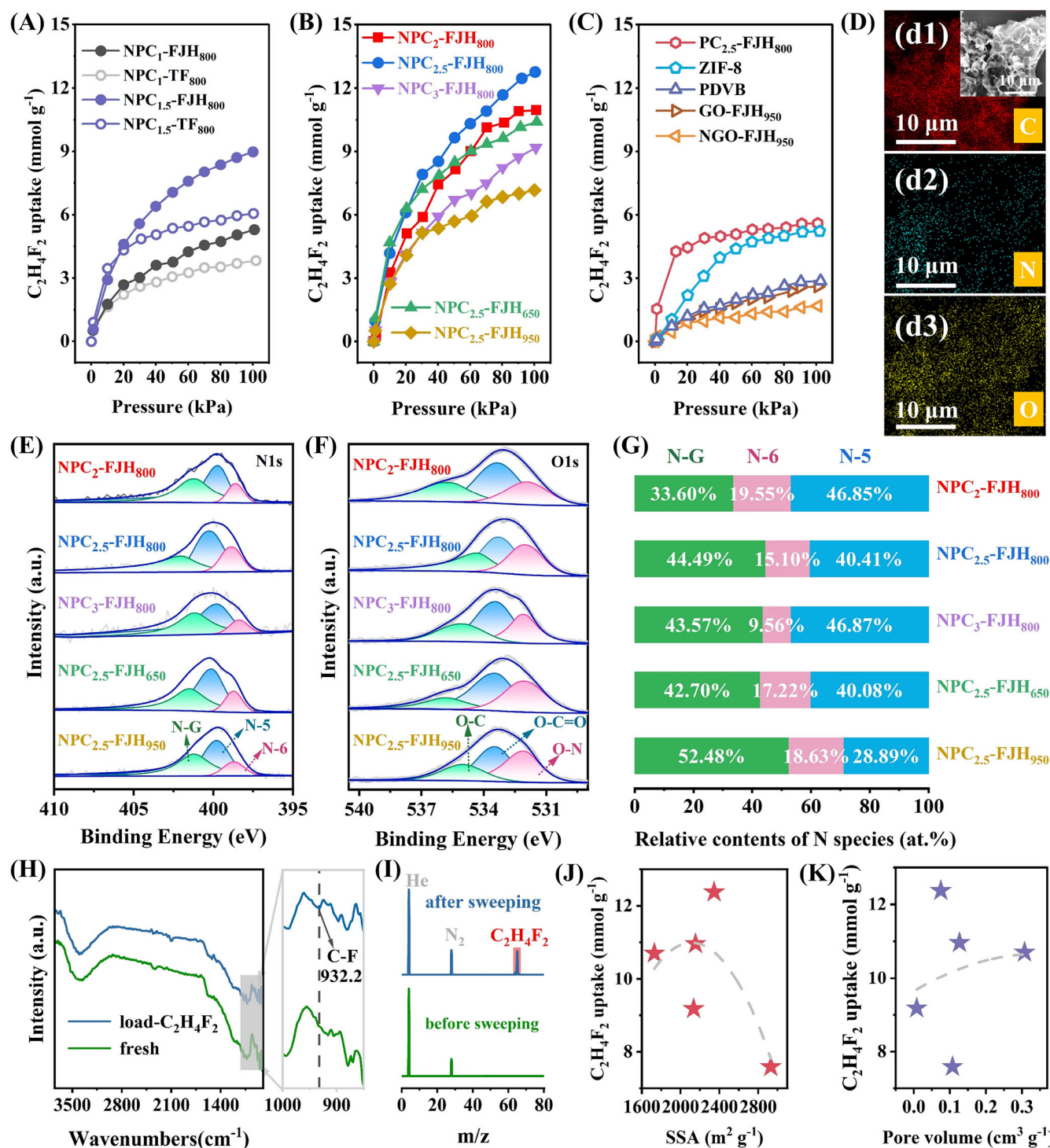


FIGURE 2 (A–C) The C₂H₄F₂ adsorption isotherms of NPC_x-FJH₇ samples, NPC_x-TF₇ samples and selected reference porous samples at 25°C and 100 kPa; (D) SEM-EDS mapping of NPC_{2.5}-FJH₈₀₀ sample; (E) N1s spectra, (F) O1s spectra and (G) elemental composition of NPC_x-FJH₇ samples; (H) FTIR spectra of load-C₂H₄F₂ and the fresh NPC_{2.5}-FJH₈₀₀ sample; (I) mass spectra of NPC_{2.5}-FJH₈₀₀ sample before and after sweeping in dynamic test; (J) the correlation between C₂H₄F₂ uptake and surface area and (K) pore volume.

are given in the Supporting Information S1.4. The C₂H₄F₂ adsorption isotherms were compared across NPC samples prepared under varying conditions (e.g., activator dosage and carbonization temperature), through distinct preparation methods (FJH vs. TF),

and against selected reference porous samples, to elucidate the physicochemical factors governing adsorption capacity contribution. As shown in Figure 2A, NPC_{1.5}-FJH₈₀₀ exhibited a C₂H₄F₂ adsorption capacity of 8.98 mmol g⁻¹, outperforming TF prepared

counterparts (NPC₁-FJH₈₀₀, 5.29 mmol g⁻¹ and NPC_{1.5}-TF₈₀₀, 6.06 mmol g⁻¹). These results confirm the superior efficiency of the FJH process in enhancing adsorption performance of NPC samples for C₂H₄F₂. Furthermore, Figure 2B shows the effects of activator dosages and carbonization temperatures on C₂H₄F₂ adsorption capacities of NPC_X-FJH_T samples, and the corresponding adsorption data are listed in Table 1. Specifically, the maximum C₂H₄F₂ adsorption capacity was obtained by NPC_{2.5}-FJH₈₀₀ (12.37 mmol g⁻¹), which was higher than that of NPC₂-FJH₈₀₀ (10.96 mmol g⁻¹) and NPC₃-FJH₈₀₀ (9.18 mmol g⁻¹). It can be inferred that the formation of hierarchically porous networks and optimal C₂H₄F₂ adsorption sites in the NPC_{2.5}-FJH₈₀₀ sample can be attributed to the moderate K₂CO₃ dosage and FJH treatment at 800°C, which endows the sample with superior affinity for C₂H₄F₂ under the same conditions. Furthermore, it is important to note that deviations in carbonization temperature, either elevated to 950°C (NPC_{2.5}-FJH₉₅₀) or reduced to 650°C (NPC_{2.5}-FJH₆₅₀), resulted in diminished C₂H₄F₂ uptakes. This result may stem from pore collapse or escaped heteroatoms during the preparation of NPC_X-FJH_T samples, both of which can diminish the interaction affinity toward polar gas molecules.

Figure 2C and Table S1 show extended comparison of C₂H₄F₂ adsorption performances of various porous samples at 25°C and 100 kPa, including N-free porous carbon (PC_{2.5}-FJH₈₀₀), graphite oxide (GO-FJH₉₅₀), N-doped graphite oxide (NGO-FJH₉₅₀), and benchmark MOF (ZIF-8). The results demonstrate that PC_{2.5}-FJH₈₀₀ possesses a micro- and mesoporosity structure and achieves a C₂H₄F₂ adsorption capacity of 5.59 mmol g⁻¹, which surpasses those of ZIF-8 (5.22 mmol g⁻¹) and also exceeds the lower C₂H₄F₂ capacities of PDVB, GO-FJH₉₅₀, and NGO-FJH₉₅₀ samples (<3 mmol g⁻¹). However, its performance remains inferior to that of NPC_X-FJH_T samples. These findings emphasize the critical role of heteroatom doping and hierarchical porosity in the NPC samples and highlight the value of FJH in the preparation of engineering porous carbon materials.

Moreover, the elemental distributions of NPC_{2.5}-FJH₈₀₀ were examined via SEM-EDS mapping (Figure 2D). It can be observed that the C, N, and O elements are uniformly distributed throughout the surface of honeycomb-like NPC_{2.5}-FJH₈₀₀ material, which is a key property for maximizing accessible adsorption sites and avoiding local saturation effects. Furthermore, XPS analyses were carried out to analyze the contribution of surface chemical features of NPC_X-FJH_T samples to C₂H₄F₂ adsorption, as shown in Figure 2E,F. The XPS full spectrum (Figure S4a) reveals the presence of C, N, and O elements on the surface of NPC_X-FJH_T samples, which confirms the observation results of SEM-EDS. The corresponding elemental content on the surface of NPC_X-FJH_T samples, as well as the relative proportions of various chemical species of N, C, and O, are summarized in Figures 2G and S4b–d, respectively. Particularly, the N 1s spectra (see Figure 2E) were deconvoluted into pyridinic N (N-6, 398.7 eV), pyrrolic N (N-5, 399.8 eV), and graphitic N (N-G, 401.2 eV).²³ These N species are known to enhance gas adsorption by adsorbents through electrostatic and hydrogen bond interactions. Given the strong electron-withdrawing features of C₂H₄F₂ molecules from the

high electronegativity of F atoms, intramolecular charge redistribution arises along the C–F bonds.^{25,26} This polarization effect induces partial positive charges (δ⁺) on C atoms and partial negative charges (δ⁻) on F atoms. As a result, the polarized C₂H₄F₂ molecules enable F (δ⁻) atoms to interact electrostatically with electron-deficient carbon sites in N-6, while the molecular dipole moment of C₂H₄F₂ facilitates dipole–dipole interactions with N-5 species. Among all samples, NPC_{2.5}-FJH₈₀₀ possesses favorable adsorption sites (N-6 and N-5) and porosity, which are the most effective for adsorbing polar molecules (see Figure 2G). In addition, the O 1s spectra (Figure 2F) further confirmed the presence of various O groups such as O=N (532.1 eV), O=C=O (533.5 eV), O–C (534.9 eV), and their corresponding relative species contents are shown in Figure S4d.²⁴ Interestingly, these O species exhibited minimal variation across NPC_X-FJH_T samples, suggesting that the N species played a more dominant role in C₂H₄F₂ adsorption performance.

FTIR spectra were collected before and after C₂H₄F₂ loading to further validate their possible interaction with NPC_{2.5}-FJH₈₀₀ sample (Figure 2H). The magnified view reveals a new vibrational band at 933.2 cm⁻¹ that appeared upon adsorption, which is ascribed to the C–F bond stretching vibration of C₂H₄F₂.^{27,28} In addition, mass spectrometry was employed to compare the mass spectra of the NPC_{2.5}-FJH₈₀₀ sample before and after C₂H₄F₂ adsorption under dynamic binary gas C₂H₄F₂/N₂ test conditions (Figure 2I). It can be found that a discernible signal corresponding to C₂H₄F₂ (*m/z* = 66) was detected after the sweeping process, which supports the fact that the NPC_{2.5}-FJH₈₀₀ sample effectively adsorbed C₂H₄F₂ and subsequently released it during desorption. Therefore, these observations confirm the successful C₂H₄F₂ capture and its interactions with the NPC_{2.5}-FJH₈₀₀ sample. Besides, previous studies have shown that the surface area and pore volume of porous materials play an important role in determining their gas adsorption performance.^{29–31} As shown in Figure 2J, C₂H₄F₂ adsorption capacities of NPC_X-FJH_T samples exhibit a volcano-shaped relationship with surface area, which means an optimal surface area range that balances active site accessibility and surface area. In other words, a moderate surface area of NPC_X-FJH_T samples facilitates optimal exposure of the active sites for improved C₂H₄F₂ adsorption. Further increases in surface area are often achieved by higher carbonization temperatures, which can adversely affect the integrity of active sites in NPC_X-FJH_T materials. In contrast, Figure 2K reveals a nearly linear correlation between pore volume and C₂H₄F₂ uptake, which means that pore volume (0.1 – 0.3 cm³ g⁻¹) serves as minor adsorption domains for C₂H₄F₂ molecules. Overall, the excellent C₂H₄F₂ adsorption performance of NPC_{2.5}-FJH₈₀₀ arises from the synergistic effect of activated N species and hierarchical porosity, which enhances the molecular interactions and facilitates high-capacity uptake of C₂H₄F₂.

3.3 | C₂H₄F₂ adsorption heats and selectivity

The C₂H₄F₂ adsorption isotherms on three NPC_X-FJH₈₀₀ samples (*X* = 2, 2.5 and 3) were measured at 100 kPa and 15 ~ 35°C to

evaluate their adsorption capacities and ideal adsorption solution theory (IAST) selectivities. The results are presented in Figure 3. In all cases, the $C_2H_4F_2$ adsorption isotherms conformed well to the Langmuir–Freundlich isotherm model ($R^2 > 0.99$), which indicates the reversible monolayer adsorption (see Figure 3A–C).³² As expected, an increase in test temperature led to a decline in $C_2H_4F_2$ adsorption capacity, consistent with an exothermic adsorption process. Consequently, as the adsorption temperature decreased from 25°C to 15°C, the $C_2H_4F_2$ capacity of NPC_{2.5}-FJH₈₀₀ sample increased from 12.37 mmol g⁻¹ to 13.64 mmol g⁻¹ at 100 kPa. Additionally, the $C_2H_4F_2$ isosteric heats (Q_{st}) in NPC_X-FJH₈₀₀ samples were analyzed via the Clausius–Clapeyron equation (see Supporting Information S1.6, Equation S2), and the results are presented in Figure S5. It can be found that NPC₃-FJH₈₀₀ exhibits the highest Q_{st} value (~50 kJ mol⁻¹) at low $C_2H_4F_2$ adsorption capacity, yet Q_{st} values declined rapidly with increasing $C_2H_4F_2$ uptakes, suggesting a limited number of high-affinity sites.³³ In contrast, NPC₂-FJH₈₀₀ and NPC_{2.5}-FJH₈₀₀ maintain relatively invariant Q_{st} values under the examined adsorption range, which

supports that the dominance of physisorption with chemisorption plays a minor role.

Moreover, Figure 3D shows the single-component adsorption isotherms for $C_2H_4F_2$, C_2H_6 , C_2H_4 , and C_2H_2 on NPC₂-FJH₈₀₀ sample at 25°C and 100 kPa. Notably, the $C_2H_4F_2$ adsorption capacity of NPC_{2.5}-FJH₈₀₀ (12.37 mmol g⁻¹) is significantly higher than those for C_2H_6 , C_2H_4 , and C_2H_2 gases (<4 mmol g⁻¹), which implies that its potential for selective $C_2H_4F_2$ capture. Meanwhile, in order to assess the separation performance in binary gas mixtures ($C_2H_4F_2/C_2H_6$, $C_2H_4F_2/C_2H_4$, $C_2H_4F_2/C_2H_2$, and $C_2H_4F_2/N_2$), IAST calculation (see Supporting Information S1.7, Equation S3.4) was employed to evaluate the adsorption selectivities of NPC_{2.5}-FJH₈₀₀ sample for various gases (see Figure 3D–F). The results reveal that NPC₂-FJH₈₀₀ exhibits high selectivity for $C_2H_4F_2$ relative to the other gases, consistent with the preferential adsorption observed in the single-component isotherms (Figure 3D). Specifically, NPC₂-FJH₈₀₀ achieved remarkable IAST selectivities of 419.93 ($C_2H_4F_2/C_2H_6$), 29.61 ($C_2H_4F_2/C_2H_4$), and 240.52 ($C_2H_4F_2/C_2H_2$), respectively (Figure 3E). As discussed in Section 3.2, the polarity for $C_2H_4F_2$ molecule arises from the

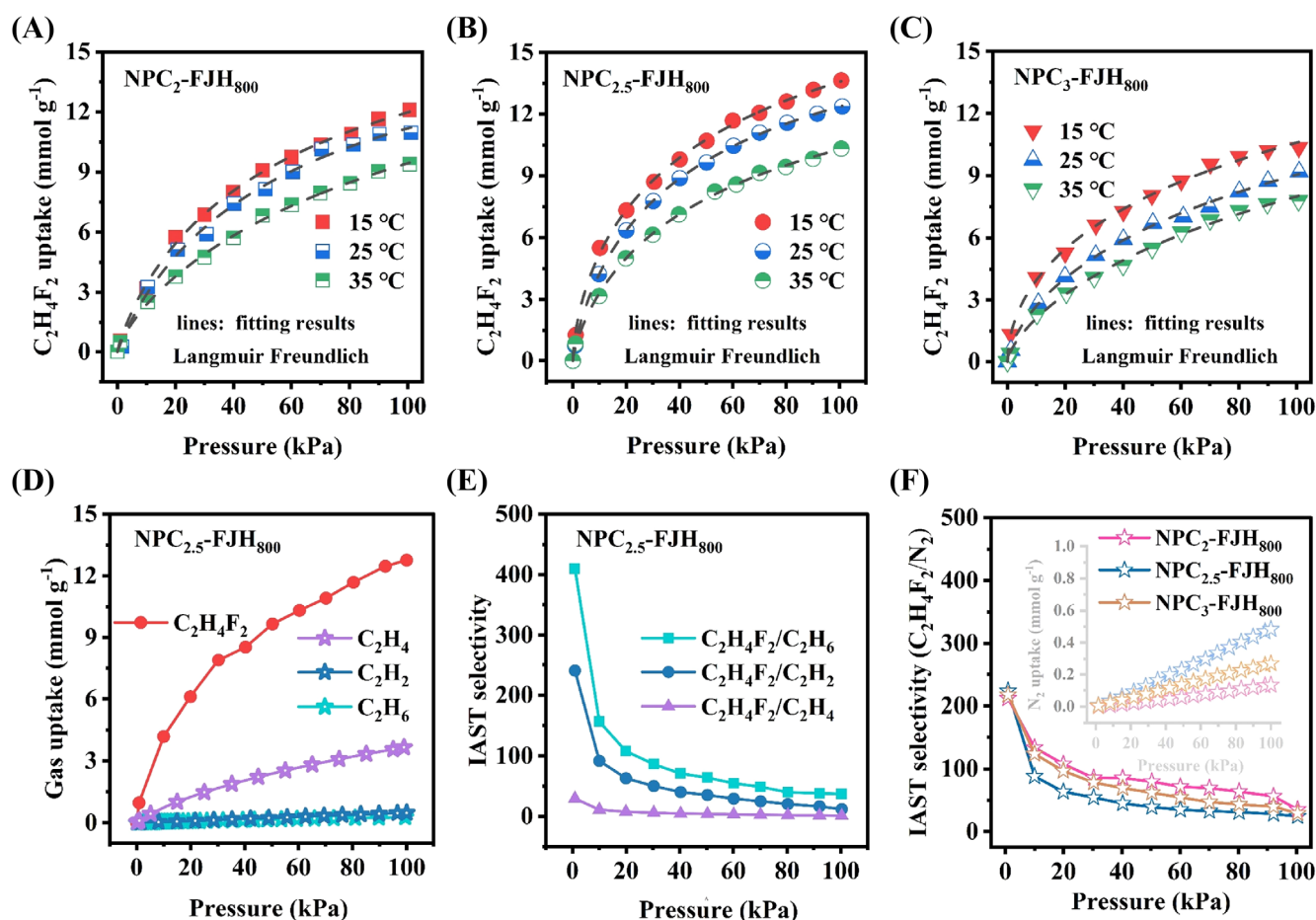


FIGURE 3 The $C_2H_4F_2$ adsorption isotherms of (A) NPC₂-FJH₈₀₀, (B) NPC_{2.5}-FJH₈₀₀ and (C) NPC₃-FJH₈₀₀ samples at 100 kPa and different temperatures (15°C, 25°C, and 35°C); (D) The $C_2H_4F_2$, C_2H_6 , C_2H_4 and C_2H_2 adsorption isotherm of NPC₂-FJH₈₀₀ sample at 100 kPa and 25°C; (E) Ideal adsorption solution theory (IAST) selectivities of NPC₂-FJH₈₀₀ sample for $C_2H_4F_2/C_2H_6$, $C_2H_4F_2/C_2H_4$, and $C_2H_4F_2/C_2H_2$ at 25°C, and (F) IAST selectivities of NPC₂-FJH₈₀₀, NPC_{2.5}-FJH₈₀₀ and NPC₃-FJH₈₀₀ samples for $C_2H_4F_2/N_2$, with inset is the N_2 adsorption isotherms at 100 kPa and 25°C.

electron-withdrawing nature of F atoms, which confers the molecule with an enhanced capacity to interact with polar heteroatoms present in the NPC₂-FJH₈₀₀ sample. It is inferred that the presence of N-6 and N-5 species facilitates localized electrostatic and hydrogen bonding interactions with the electron-deficient regions of C₂H₄F₂. The specific adsorption mechanisms underlying these interactions will be elucidated in the following section. As shown in the inset of Figure 3F, the NPC_x-FJH₈₀₀ samples exhibit negligible N₂ adsorption capacities (<0.6 mmol g⁻¹) with nearly linear adsorption isotherms over a wide pressure range. The calculated IAST selectivities of C₂H₄F₂/N₂ (~215, Table 1) suggest that N₂ adsorption capacities are governed by surface area rather than specific adsorbate-adsorbent interactions, as supported by other similar porous carbon material studies.^{16,34–36} Thus, the excellent C₂H₄F₂ adsorption performance of the NPC₂-FJH₈₀₀ sample as well as resistance to competing gas interferences rendering it a promising candidate for selective C₂H₄F₂ separation applications.

3.4 | Adsorption mechanism

To gain deeper insights into the experimentally observed inverse correlation between C₂H₄F₂ adsorption capacity and the N species in NPC samples (Table 2), DFT and NBO calculations were conducted to systematically assess the adsorption behavior of C₂H₄F₂ on undoped C and C models doped with N-5, N-G, and N-6. The atomic structures of NPC material models and their corresponding guest gas C₂H₄F₂ adsorption complexes are illustrated in Figures S6–S10. For comparison, the adsorption energies of C₂H₂, C₂H₄, and N₂ on these models were also computed to evaluate their gas selectivities (Table 2). The adsorption energies of C₂H₄F₂ on graphite carbon (G-C), N-6 doped C, and N-5 doped C are −16.97, −23.78, and −15.54 kJ·mol⁻¹, respectively. It can be found that N-6 doped C has the most negative among all tested gases, demonstrating the strongest preferential affinity for C₂H₄F₂ on these sites. This is attributed to the formation of weak hydrogen bonds between C₂H₄F₂ and the N–H moiety on the N-5 doped carbon surface, with both F atoms located at a distance of 2.42 Å from H atom (Figure S7).³⁷ This interaction distance is notably shorter than the closest F atom separations observed on other NPC samples, measured as 2.81 Å for G-C (Figure S8), 3.17 Å for N-6 doped C (Figure S9), and 3.12 Å for N-G doped C (Figure S10). In contrast, N-G doped C exhibits a higher affinity for C₂H₄ (−18.85 kJ·mol⁻¹) than for C₂H₄F₂

(−15.47 kJ·mol⁻¹), theoretically confirming the reduced selectivity of N-G sites toward C₂H₄F₂, in line with the experimental observations.

To reveal the underlying electronic interactions, NBO analysis was performed combined with second-order perturbation theory.³⁸ On N-5 doped C surface, the strongest interaction was identified between the lone pair orbitals LP(3) of F41 (or F42) and the antibonding orbital BD*(1) of the H16–N34 bond. As shown in Figure 4A,B, the isosurface map of valence NAOs of LP(3) F41 (composed of F41(sp¹)) and BD*(1) of H16–N34 (composed of H16(s) + N34(sp^{2.26})) reveals delocalization of the lone pair electrons of F41 atom toward C surface, partially overlapping with the antibonding orbital of H16–N34. On the G-C model, the dominant interaction occurs between the LP(3) orbital of F44 and the BD*(2) antibonding orbital of the C28–C29 bond (Figure 4C,D), similarly involving F atomic lone pair to C orbital overlap.

In the C₂H₄F₂---N-6 system, however, the strongest orbital interaction shifts away from the F atoms. Instead, it is observed between the bonding orbital BD(1) of the C36–H37 bond and the antibonding orbital BD*(2) of the C7–C9 pair (Figure 5A,B). The F-related sites (F42...C3–C5 and F43...C3–C34) display negligible orbital overlap (Figure 5C,D), indicating that N-6 shows weak affinity for F atoms and prefers interacting with the −CH moiety of C₂H₄F₂. A more distinct preference for C atoms is observed on N-G. In the C₂H₄F₂---N-G doped C system, the most significant interaction arises between the lone pair antibonding orbital LP*(1) of C41 and the bonding orbital BD(2) of the C4–C5 pair (Figure S11a,b). Meanwhile, the overlap between LP(2) orbital of F43 and the BD*(2) orbital of C3–C6 remains minimal (Figure S11c,d). These C atoms preferences are further reflected in the analysis of C₂H₂ and C₂H₄ adsorption. In the case of C₂H₂, the BD(3) orbital of the C37–C39 bond strongly interacts with the singly excited orbital RY*(3) of N36 (Figure S12). For C₂H₄, four comparable orbital interactions involving the C3–C6 and C7–C8 regions of N-G are evident (Figure S13), with an additional interaction between BD(1) of C37–C40 and the RY*(3) orbital of N36 (Figure S14). Overall, the adsorption of C₂H₄F₂ on N-5 doped C is primarily facilitated through interactions involving F atoms, leading to higher adsorption energy (−23.78 kJ·mol⁻¹). In contrast, G-C, N-5 and N-G favor interactions with C atoms rather than F atoms, resulting in weaker C₂H₄F₂ adsorption (−16.97, −15.54 and −15.47 kJ·mol⁻¹). Particularly, N-G exhibits enhanced affinity for C₂H₂ and C₂H₄, which compromises its selectivity toward C₂H₄F₂, in agreement with experimental observations.

Species	Guest gases			
	C ₂ H ₂ (kJ·mol ⁻¹)	C ₂ H ₄ (kJ·mol ⁻¹)	C ₂ H ₄ F ₂ (kJ·mol ⁻¹)	N ₂ (kJ·mol ⁻¹)
G-C	−12.45	−12.87	−16.97	−7.35
N-5	−16.77	−16.36	−23.78	−7.31
N-G	−16.01	−18.85	−15.47	−8.18
N-6	−12.05	−13.69	−15.54	−5.08

TABLE 2 The adsorption energies of the constructed NPC samples with the adsorbed guest gases.

FIGURE 4 Isosurface map (isovalue = 0.03 au) of valence NAOs of F41 LP(3) interacting with the H16–N34 BD*(1) orbitals in N-5 doped C–C₂H₄F₂ cluster: (A) top and (B) side view; Isosurface map (isovalue = 0.03 au) of valence NAOs of F44 LP(3) interacting with the C28–C29 BD*(2) orbitals in G-C–C₂H₄F₂ cluster: (A) top and (B) side view.

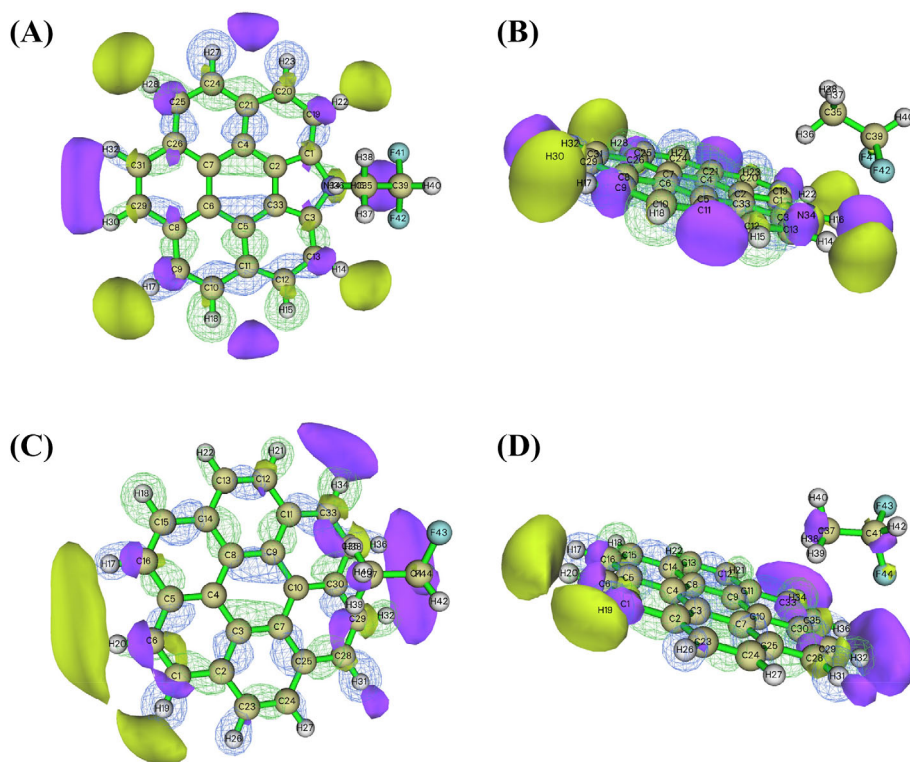
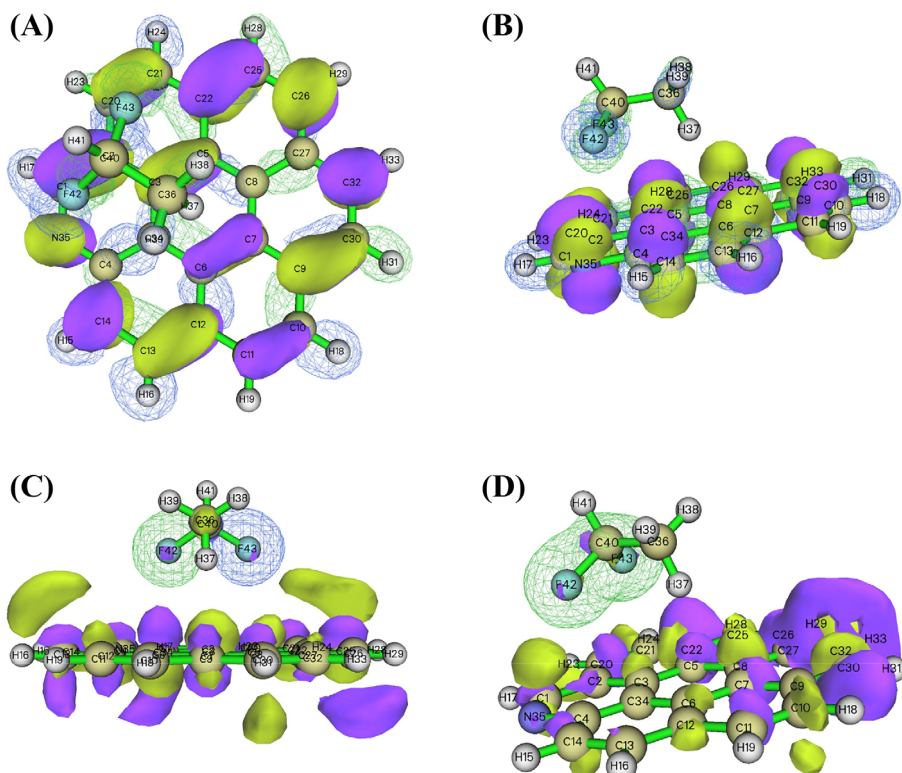


FIGURE 5 Isosurface map (isovalue = 0.03 au) of valence NAOs of (A, B) C36–H37 BD(1) interacting with the C7–C9 BD*(2) orbitals, (C) F42 LP(1) interacting with the C3–C5 BD*(1) orbitals, and (D) F43 LP(1) interacting with the C3–C34 BD*(1) orbitals in N-6 doped carbon–C₂H₄F₂ cluster.



3.5 | Adsorption kinetics and diffusion analysis

In industrial gas separation applications, the adsorption rate of specific adsorbates is a key parameter for the assessment of the practical viability of the adsorbents.³⁹ The C₂H₄F₂ adsorption kinetics of

NPC_X-FJH_T and selected reference samples (PC_{2.5}-FJH₈₀₀, ZIF-8, and PDVB) were carried out at 25°C and 100 kPa, and the results are shown in Figures 6 and S15. The results show that all tested NPC_X-FJH_T samples exhibited rapid adsorption rates of C₂H₄F₂ within 1 min, followed by a progressive approach to saturation,

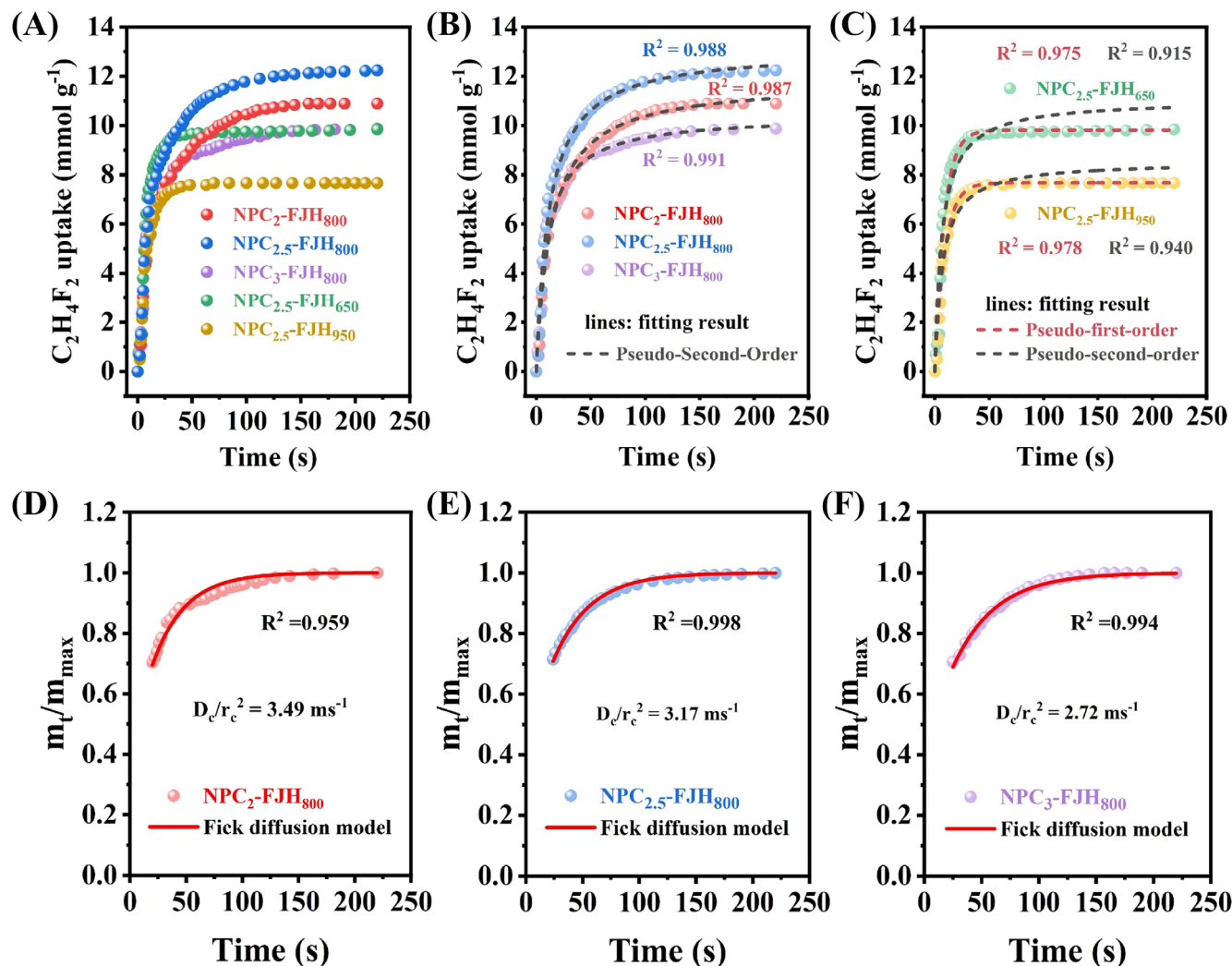


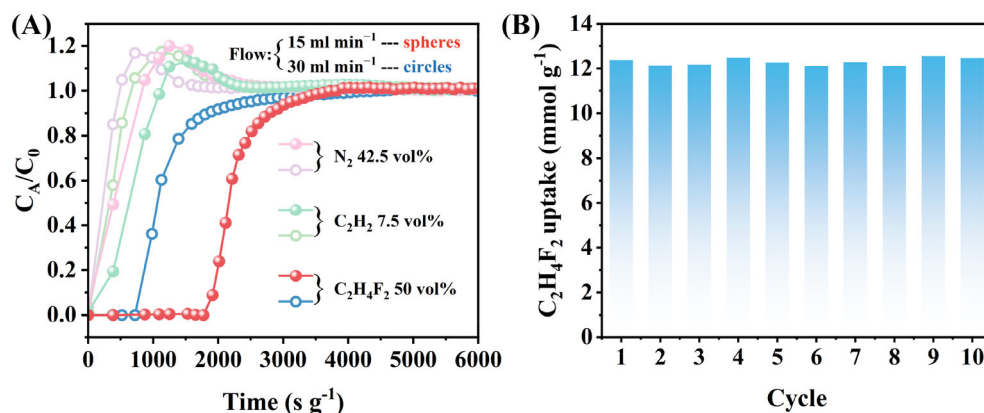
FIGURE 6 (A) $C_2H_4F_2$ adsorption rate curves for NPC_x-FJH_T samples and (B, C) adsorption kinetics curves fitted using PFP and PSO models at 25°C and 100 kPa; (D–F) diffusion fitting curves based on the Fick diffusion model at 25°C and 100 kPa.

which means that they have the potential to effectively capture $C_2H_4F_2$ (Figure 6A). In contrast, the reference porous samples (PC_{2.5}-FJH₈₀₀, PDVB, and ZIF-8) showed comparatively slower $C_2H_4F_2$ adsorption rates and lower $C_2H_4F_2$ adsorption capacities (Figure S15a). Among the samples, NPC_{2.5}-FJH₈₀₀ and NPC_{2.5}-FJH₆₅₀ achieved the highest adsorption capacity (12.37 mmol g⁻¹) and fastest equilibrium rate (99 s, 1.65 min), respectively. Conversely, NPC₃-FJH₈₀₀ displayed the slowest adsorption kinetics (197 s, 3.28 min), while NPC_{2.5}-FJH₉₅₀ demonstrated the lowest $C_2H_4F_2$ uptake capacity (7.59 mmol g⁻¹). These performance limitations can be ascribed to excessive graphitization and partial pore collapse arising from elevated K_2CO_3 dosages and high FJH temperatures, which together reduce diffusivity and accessible active sites. Next, the adsorption data were fitted using pseudo-first-order (PFO) and pseudo-second-order (PSO) kinetic models (see Figure 6B,C). The fitting results reveal that NPC_x-FJH_T samples synthesized at 800°C (NPC₂-FJH₈₀₀, NPC_{2.5}-FJH₈₀₀, and NPC₃-FJH₈₀₀) as well as selected reference porous samples (PC_{2.5}-FJH₈₀₀ and

PDVB) showed adsorption behaviors that more closely followed the PSO model ($R^2 > 0.98$), which implies that their interactions with $C_2H_4F_2$ are mainly governed by chemisorption.^{40,41} Conversely, NPC samples (Figure S15a) synthesized at either higher (NPC_{2.5}-FJH₉₀₀) or lower (NPC_{2.5}-FJH₆₅₀) temperatures deviated from the PSO model ($0.91 < R^2$), while exhibiting strong agreement with the PFO model ($R^2 \approx 0.98$). Similarly, the adsorption kinetics of the benchmark MOF ZIF-8 were well described by the PSO model ($R^2 = 0.995$). Therefore, these results mean that $C_2H_4F_2$ adsorption on NPC_x-FJH_T samples involves a combination of chemisorption and physisorption, which is consistent with the results obtained from the Langmuir–Freundlich isotherm model, isosteric heats, and DFT calculations.

Moreover, the effects of synthesis parameters (activator dosages and pyrolysis temperatures) on the diffusion resistance of various NPC_x-FJH_T and reference porous samples were studied by fitting adsorption rate data within the 70 ~ 100% total $C_2H_4F_2$ adsorption capacities using the Fick diffusion model (Figure 6D–F).^{42,43} The

FIGURE 7 (A) Breakthrough curves of $C_2H_4F_2/C_2H_2/N_2$ (v:v: v = 50:7.5:42.5) separation on NPC_{2.5}-FJH₈₀₀ at 25°C, 15 mL min⁻¹ (solid spheres) or 30 mL min⁻¹ (hollow circles); (B) reusability of NPC_{2.5}-FJH₈₀₀ sample for $C_2H_4F_2$ adsorption.



results show that the experimental data exhibited excellent agreement with the Fick model, with determination coefficients $R^2 > 0.95$. Notably, as K_2CO_3 dosage increases, the diffusion coefficients (D/r^2) followed the order NPC₂-FJH₈₀₀ (3.49 ms^{-1}) > NPC_{2.5}-FJH₈₀₀ (3.17 ms^{-1}) > NPC₃-FJH₈₀₀ (2.72 ms^{-1}), which highlights that a moderate K_2CO_3 dosage optimizes pore structure and facilitates enhanced mass transfer. Nonetheless, the diffusion performance of NPC_{2.5}-FJH₈₀₀ was slightly inferior to that of NPC₂-FJH₈₀₀, the former achieved a superior $C_2H_4F_2$ adsorption capacity ($11.48 \text{ mmol g}^{-1}$) within 1 min, surpassing the 9.53 mmol g^{-1} observed for NPC₂-FJH₈₀₀ under identical conditions. Additionally, the estimated D/r^2 values for NPC_{2.5}-FJH₆₅₀ and NPC₃-FJH₈₀₀ were 8.93 ms^{-1} and 8.42 ms^{-1} , respectively (Figure S15b,c), yet both samples exhibited lower $C_2H_4F_2$ adsorption capacities within the same time frame. In short, these findings suggest that NPC_{2.5}-FJH₈₀₀ offers a favorable balance between diffusion kinetics and adsorption capacity. Furthermore, given the multiple demands of industrial gas separation for rapid kinetics, high adsorption capacity and gas selectivity, NPC_{2.5}-FJH₈₀₀ emerges as a highly feasible application for efficient $C_2H_4F_2$ capture and separation.

3.6 | Dynamic separation performance and reusability

Breakthrough experiments were conducted to evaluate the dynamic separation performance of NPC_{2.5}-FJH₈₀₀ at varying $C_2H_4F_2$ concentrations (see Figures 7A and S16). The breakthrough tests were carried out using $N_2/C_2H_2/C_2H_4F_2$ ternary gas mixtures at 25°C, with gas volume ratios (42.5/7.5/50 vol% and 55/12/33 vol%) and flow rates of 15 mL min⁻¹ (solid spheres) and 30 mL min⁻¹ (hollow circles). It can be found that competing gas components of N_2 and C_2H_2 exhibit almost immediate breakthroughs ($\sim 23 \text{ s g}^{-1}$) and then the displacement of the previously adsorbed N_2 and C_2H_2 by $C_2H_4F_2$ becomes obvious at $\sim 480 \text{ s g}^{-1}$ ($C_A/C_0 > 1$). These findings demonstrate that NPC_{2.5}-FJH₈₀₀ preferentially captures $C_2H_4F_2$ from multi-component gas mixtures, consistent with the DFT results. Furthermore, based on previously reported literature,^{44,45}

the actual $C_2H_4F_2$ adsorption capacities in the breakthrough experiments were calculated, and the results are listed in Table S2. The results showed that while NPC_{2.5}-FJH₈₀₀ exhibited a high static capacity of $12.37 \text{ mmol g}^{-1}$ for $C_2H_4F_2$ at 25°C and 100 kPa, it also maintained effective $C_2H_4F_2$ capture performance under continuous flow conditions. For $N_2/C_2H_2/C_2H_4F_2$ (v:v:v, 42.5/7.5/50 vol %) mixture, NPC_{2.5}-FJH₈₀₀ achieved a selective adsorption capacity of 6.33 mmol g^{-1} for $C_2H_4F_2$ at 15 mL min⁻¹. In contrast, when $C_2H_4F_2$ concentration was decreased to 33 vol%, $C_2H_4F_2$ capacity increased to 7.86 mmol g^{-1} at the same flow rate. In addition, increasing the gas flow rate to 30 mL min⁻¹, $C_2H_4F_2$ capacities decreased slightly to 5.46 and 6.48 mmol g⁻¹, respectively. This result suggests that the efficiency of the adsorbent is somewhat flow rate-dependent, likely due to mass transfer limitations at higher flow velocities. Meanwhile, the adsorption capacities of competing gases (N_2 and C_2H_2) remained below 0.8 mmol g^{-1} under the same conditions. Therefore, these results suggest that NPC_{2.5}-FJH₈₀₀ can selectively adsorb $C_2H_4F_2$ from mixed gas, demonstrating its potential for industrial applications in the separation of diluted $C_2H_4F_2$.

In addition to breakthrough performance, the practical deployment of adsorbents in industrial settings demands exceptional separation performance and robust regenerative stability across multiple adsorption-desorption cycles. The recyclability of the NPC_{2.5}-FJH₈₀₀ sample for $C_2H_4F_2$ capture was further evaluated through consecutive adsorption-desorption cycles at 25°C and 100 kPa, with regeneration carried out at 60°C and 10 kPa (Figure 7B). Notably, the reused NPC_{2.5}-FJH₈₀₀ sample retained its adsorption capacity over multiple cycles without significant degradation, which supports its high stability and potential for long-term use in industrial gas separation applications. Furthermore, the crystal structure and morphology feature of the recycled NPC_{2.5}-FJH₈₀₀ sample closely resembles that of its freshly prepared counterpart, suggesting the absence of major structural changes during adsorption-desorption cycles (Figure S17a-c). Overall, the breakthrough and cycling data affirm the promise of NPC_{2.5}-FJH₈₀₀ as a high-performance adsorbent for selective $C_2H_4F_2$ capture under dynamic flow conditions.

4 | CONCLUSION

In summary, we have successfully developed the ultrafast preparation of honeycomb-like nitrogen-doped porous carbon (NPC_X-FJH_T) within 20 s via FJH strategy. Compared with microporous NPC_X-TF_T samples obtained from traditional TF, the inside-out thermal gradient inherent to FJH process affords NPC_X-FJH_T samples with hierarchical porous structures, which significantly improves 1,1-difluoroethane (C₂H₄F₂) adsorbates mass transfer efficiency within the porous network. Furthermore, the well-distributed heteroatoms in NPC_X-TF_T effectively tune the electronic structure of pyrrolic N–C bonds, thereby facilitating preferential adsorption of polar C₂H₄F₂ molecules. This selective C₂H₄F₂ adsorption behavior aligns well with the Langmuir–Freundlich model and involves both physisorption and chemisorption, compared to the predominantly physisorption observed in typical porous materials. In addition to electronic interactions, the synergistic contributions of high specific surface area and optimized hierarchical pore structure further support C₂H₄F₂ uptake. These structural and chemical features together confer excellent resistance to competitive adsorption for NPC_{2.5}-FJH₈₀₀ sample, affording record-high C₂H₄F₂ capacity (12.37 mmol g^{−1}) and IAST selectivities (1,1-difluoroethane/ethane (C₂H₄F₂/C₂H₆, ~420) and 1,1-difluoroethane/nitrogen (C₂H₄F₂/N₂, ~215), respectively) under complex gas mixture conditions. Additionally, dynamic column breakthrough and recyclability tests show that NPC_{2.5}-FJH₈₀₀ sample retains superior adsorption performance and structural stability over multiple C₂H₄F₂ adsorption–desorption cycles. Therefore, this work provides a sustainable and scalable strategy for the selective capture and recovery of C₂H₄F₂, with strong potential for industrial implementation.

NOMENCLATURE

δ ⁺	partial positive charges
δ [−]	partial negative charges
C ₂ H ₂	acetylene
C ₂ H ₂ F ₄	1,1,1,2-tetrafluoroethane
C ₂ H ₄	ethylene
C ₂ H ₄ F ₂	1,1-difluoroethane
C ₂ H ₆	ethane
CO ₂	carbon dioxide
D/r ²	diffusion coefficients
DFT	density functional theory
FJH	flash Joule heating
G-C	graphite carbon
HCl	hydrochloric acid
HFCs	hydrofluorocarbons
IAST	ideal adsorption solution theory
K ₂ CO ₃	potassium carbonate
N ₂	nitrogen
N-5	pyrrolic nitrogen
N-6	pyridinic nitrogen

NBO	natural bond orbital
N-G	graphitic nitrogen
NPC	Nitrogen-doped porous carbon
NPC-FJH	NPC prepared by flash Joule heating
NPC-TF	NPC prepared by tubular furnace
PFO	pseudo-first-order
PSO	pseudo-second-order
Q _{st}	isosteric heats
TF	tubular furnace

AUTHOR CONTRIBUTIONS

Fan-Peng Meng: Investigation; formal analysis; visualization. **Wei Hui:** Project administration; Resources, Supervision; Funding acquisition; Methodology; Writing – original draft; Writing – review & editing. **Duan-Jian Tao:** Project administration; conceptualization; writing – original draft; funding acquisition. **Ming-Shuai Sun:** Formal analysis; investigation; data curation; software; visualization. **Yan Zhou:** Investigation; visualization. **Dan-Dan Cai:** Methodology; investigation. **Fei-Feng Mao:** Validation; formal analysis. **Jia-Yin Zhang:** Formal analysis; investigation.

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CONFLICT OF INTEREST STATEMENT

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

DATA AVAILABILITY STATEMENT

The numerical data from Figures 1–3, 6, 7, and Figures S2, S4, S5, and S15–S17 are tabulated in a .zip file. All experimental data were obtained from individual measurements. The data that support this study are available from the corresponding author upon reasonable request.

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

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