

Dual-functional LDH interface engineered stable SiO₂@LDH-based porous liquids for efficient CO₂ capture

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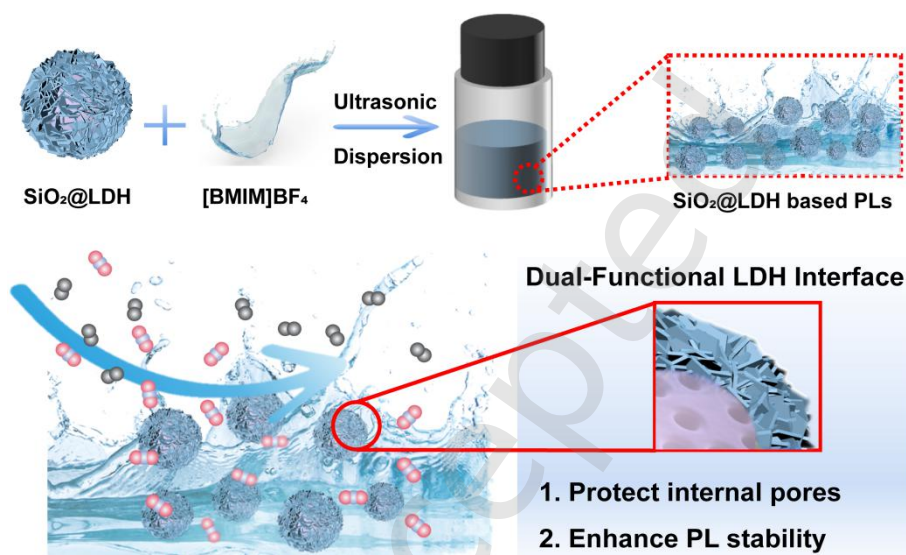
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Dual-Functional LDH Interface Engineered Stable $\text{SiO}_2@\text{LDH}$ -Based Porous Liquids for Efficient CO_2 Capture

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The dual-functional LDH interface achieved the following: (1) it prevented the IL from entering the pores, thereby preserving the porosity of the PL for CO_2 capture, and (2) the abundant hydroxyl groups on the surface of the LDH nanosheets enhanced the interaction with the IL, ensuring the long-term uniform stability of the PL.

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ABSTRACT: Porous liquids (PLs), as novel materials combining liquid fluidity with permanent porosity, demonstrate significant potential in gas capture and separation. However, the core challenge lies in achieving long-term stable dispersion of porous guests within sterically hindered solvents while maintaining pore accessibility. This work proposes a dual-functional layered double hydroxide (LDH) interfacial engineering strategy, successfully constructing stable porous SiO₂@LDH-based PLs. Cross-grown LDH nanosheets act as a physical barrier, effectively preventing steric solvent molecules from entering SiO₂ pores, thereby preserving abundant nanoscale cavities within the PL for CO₂ capture. Meanwhile, the abundant hydroxyl groups on LDH nanosheets form hydrogen bonds with the sterically hindered solvents, significantly enhancing the dispersion stability of the pore generators and preventing aggregation and sedimentation. The resulting PL exhibits outstanding long-term stability (>6 months without sedimentation) and highly efficient CO₂ capacity (1.51 mmol/g at 10 bar and 25 °C). Furthermore, this PL exhibits excellent selectivity (with a selectivity of 170 when simulating flue gas with a CO₂: N₂ ratio of 15%/85%) and cycle stability (retaining 97% of its maximum CO₂ capacity after 10 cycles). Isothermal adsorption models and adsorption kinetics models confirmed that the process primarily involves heterogeneous multilayer physical adsorption, thereby avoiding the capacity loss and energy consumption associated with regeneration caused by chemical absorption in traditional CO₂ absorption liquids. This study presents a novel and versatile interface design method for fabricating high-performance, stable PLs for CO₂ capture.

KEYWORDS: porous liquids, dual-function interface, layered double hydroxide (LDH) nanosheets, CO₂ capacity

1 Introduction

Under the Paris Agreement, limiting global warming to well below 2 °C necessitates stringent control of CO₂ emissions. Among various carbon capture and storage strategies, post-combustion capture stands as a crucial technological approach to address current CO₂ emission challenges, as it directly applies to flue gas treatment in existing power plants [1]. Advances in porous materials science have revealed the potential of numerous novel solid adsorbents for CO₂ capture [2]. However, their solid-phase morphology imposes limitations in continuous flow processes. Porous liquids (PLs) incorporate permanent pores into liquid matrices, combining the high selectivity of adsorbents with the pumpability and continuous operation potential of liquids, thereby providing a novel platform for gas capture [3,4].

Based on the combination of pore generators and sterically

hindered solvents, PLs are designed and classified into four types (I-IV) [5,6], ranging from intrinsic liquid porosity to dispersed frameworks, dissolved porous cavities, and molten porous structures. Among these, Type III PLs, with their porous hosts and sterically hindered solvents, offer significant advantages such as modular design, flexible regulation, and simple processing, and hold great potential for development into a new generation of green, transformative gas capture and conversion media. The construction of Type III PLs primarily relies on interfacial interactions between porous hosts and sterically hindered solvents [7-10]. However, this also introduces a fundamental trade-off during PLs construction: balancing system stability with pore accessibility. Using unmodified porous particles directly often results in aggregation and sedimentation, while sterically hindered solvents may invade and fill the pores [11,12]. To maintain stability, surface modification of porous hosts is required to match the solvent,

but this readily leads to pore blockage. Many studies have demonstrated that the gas capture and separation performance of Type III PL is significantly inferior to that of existing porous solid adsorbents [13,14]. Therefore, developing a strategy that simultaneously addresses particle dispersion stability and pore protection is a bottleneck in advancing PLs for CO₂ capture.

This work proposes a dual-function interface strategy that uses LDH nanosheets as an interlayer to selectively block barrier solvents from entering the internal pores of SiO₂, thereby preserving the pores in the liquid phase for efficient CO₂ capture. Specifically, a synthesis-followed-by-etching method is employed to first synthesize solid SiO₂@LDH. Then, taking advantage of the stability of LDH-NS in alkaline solutions, the solid SiO₂ was etched to form mesoporous SiO₂, yielding mesoporous SiO₂@LDH. Dispersing mesoporous SiO₂@LDH in an ionic liquid yields a PL system, driven by hydrogen bonding between the abundant hydroxyl groups on the LDH surface and the ionic liquid. The interfacial properties, long-term storage stability, and viscosity of the PL were systematically characterized and evaluated. Through comparative experiments and mechanistic analysis, the study focused on the influence of the LDH interface on enhancing CO₂ capture performance, selectivity, and cycle stability. Isothermal adsorption models and adsorption kinetics models were employed to thoroughly investigate the CO₂ capture mechanism of the SiO₂@LDH-based PL. This study not only developed a high-performance CO₂ capture material but, more importantly, proposed a universal interface engineering design strategy, paving a new path for the future development of functionalized and stable PL systems.

2 Experimental

2.1. Material preparation

2.1.1 Synthesis of SiO₂ microspheres

SiO₂ spheres were synthesized via a modified Stöber method. Anhydrous ethanol (128 g) and aqueous NH₃ solution (54 mL) were placed in a 500 mL round-bottom flask. After heating to 30 °C, 8.4 mL of tetraethyl orthosilicate (TEOS) was added, and the mixture was stirred for 1 h. The resulting SiO₂ spheres were collected by centrifugation (4900 rpm, 10 min) and washed five times with deionized water. After each washing cycle, the particles were dispersed in an ultrasonic bath for 10 min and subsequently separated by centrifugation. The final product was dried at 60 °C overnight to obtain the SiO₂ microspheres.

2.1.2. Synthesis of SiO₂@LDH

SiO₂@LDH composites were prepared by electrostatic self-assembly method using SiO₂ spheres and LDH nanosheets (**Fig. (1a)**). SiO₂ spheres (0.1 g) were ultrasonically dispersed in 20 mL of ethanol for 15 min. 0.01 g of sodium dodecylbenzene sulphate (SDBS) was added to the above solution and stirred for 1 h. After centrifugation, the supernatant was discarded. The precipitate was dispersed in 20 mL ethanol under sonication. A dispersion of 0.01 g LDH nanosheets was added to the above solution. The method for preparing LDH nanosheets is described in our group's previous work [15]. The solution was stirred for 5 h and then centrifuged. The precipitate was collected and dried at 70 °C overnight to obtain SiO₂@LDH composites (marked as SLDH-2).

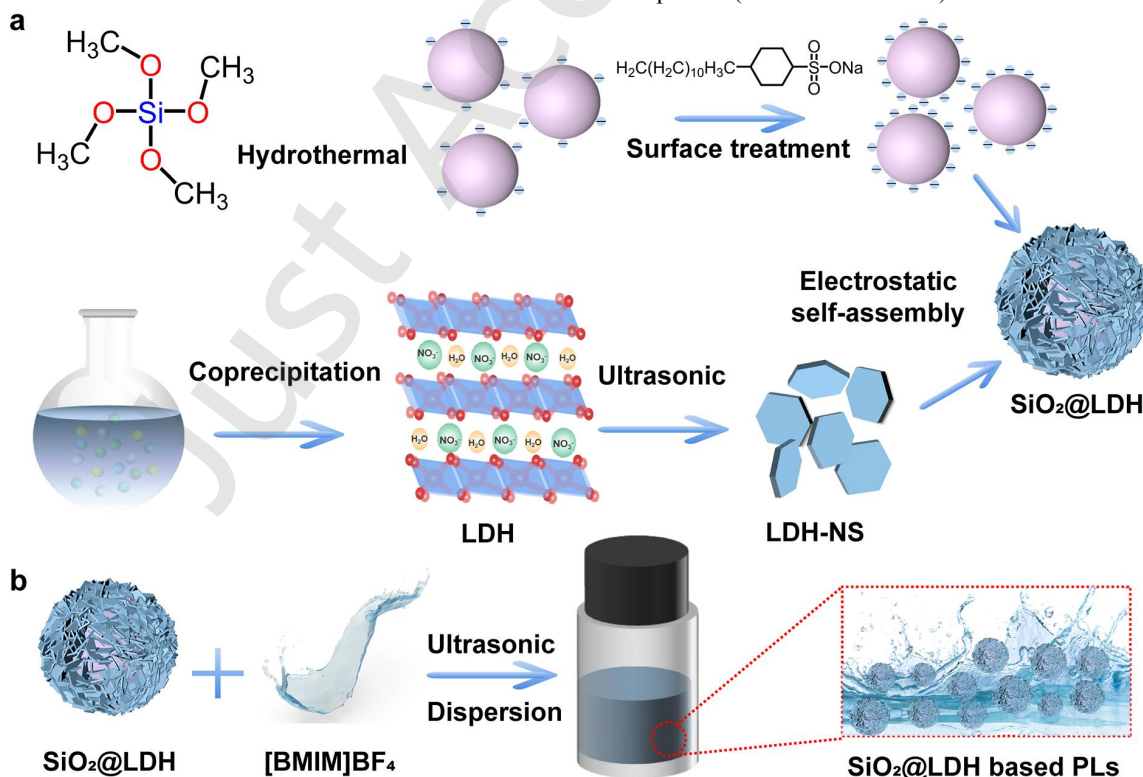


Figure 1 (a) Preparation of SiO₂@LDH by self-assembly method and (b) preparation of SiO₂@LDH-based nanofluids by direct dispersion method.

2.1.3. Preparation of mesoporous SiO₂@LDH

SLDH-2 was dispersed in 1 M NaOH solution and treated for different hours at room temperature to etch SiO₂ cores. The

resulting particles were then washed to neutrality with deionised water. Finally, the sample named SLDH-2-xh was obtained by drying at 70 °C, x represented the etched hour.

2.1.4. Preparation of PLs

A certain mass of SLDH-2-2h was placed in 10 mL ionic liquid ([BMIM][BF₄]) and fully sonicated for 10 min at room temperature in order to prepare SLDH-2-2h-based PLs (**Fig. (1b)**). Using the same methodology, SiO₂ and SLDH-2 were used as pore generators to obtain SiO₂ (SLDH-2)-based PLs, respectively. These PLs were denoted as SLDH-2-2h, SiO₂, or SLDH-2-*x*%BF₄ (*x* represents the mass fraction of pore generators in PLs).

2.2. Material characterizations

Detailed structural characterization of materials could be found in the Supporting Information.

2.3. Adsorption performance evaluation

The gas adsorption isotherms were determined using a Beishide high-temperature, high-pressure adsorption analyzer (3H-2000PH). All measurements were performed using ultra-high-purity N₂ and CO₂ (99.99%). A water bath was used to control the adsorption temperature (the test temperatures used in this study were 298 K, 308 K, and 318 K).

3. Results and discussion

3.1. Structural characterization of SiO₂@LDH

The crystalline structure of the samples was analyzed using powder X-ray diffraction (XRD). The XRD patterns of SiO₂ (PDF#39-142) and LDH (PDF#51-1525) both retain the crystal structure of SiO₂@LDH (**Fig. (2a)**). In addition, the (003), (006) and (110) characteristic diffraction peaks of the LDH phase are more pronounced, indicating that the surface of SiO₂ assembles the lamellar structure of LDH in core-shell SiO₂@LDH[16]. Mesoporous SLDH-2-2h treated by alkali etching shows no significant changes, demonstrating the stability of LDH-NS in an alkali solution and the maintenance of SiO₂@LDH core-shell structure. Fourier transform infrared spectroscopy (FTIR) was also used to characterize the chemical information on the surface of the samples (**Fig. (2b)**). Compared to the chemical composition of SiO₂, SLDH-2-2h maintained the distinct characteristic peaks at ~1110 and ~795 cm⁻¹, which is corresponding to the stretching vibrational peaks of Si-O-Si and O-Si-O bonds[17]. Meanwhile, the intensity weakening of SiO₂ characteristic

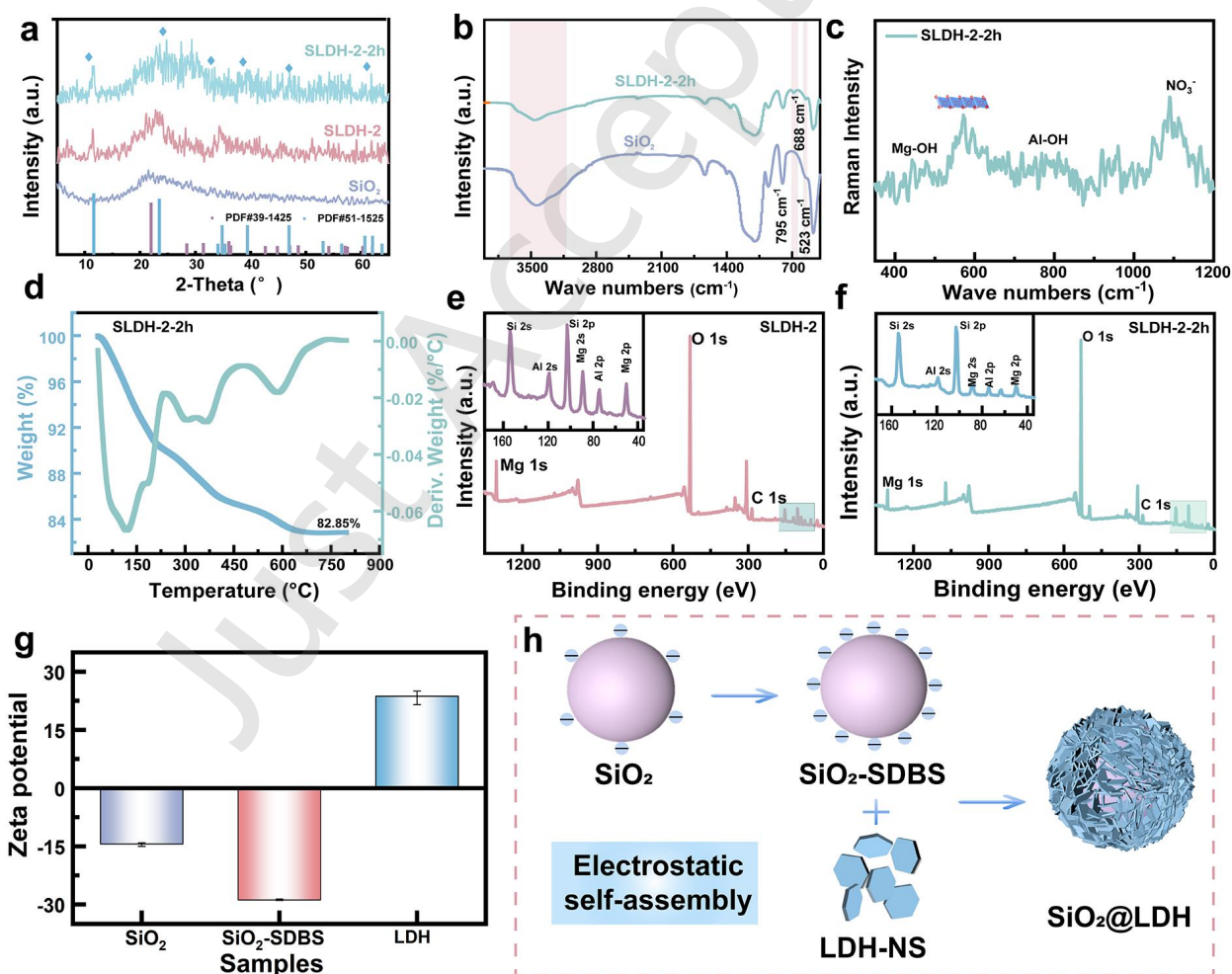


Figure 2 (a) XRD pattern, (b) FT-IR spectrum, (c) Raman spectra, (d) TG curve of SLDH-2-2h, XPS wide-scan spectra of (e) SLDH-2 and (f) SLDH-2-2h, (g) zeta potential values of SiO₂, SiO₂-SDBS and LDH, and (h) the schematic diagram of self-assembly preparation of SiO₂@LDH.

peaks and the appearance of the metal-O vibrational peaks (554 cm⁻¹ and 676 cm⁻¹) in the LDH confirm the confirmation of the LDH encapsulation on the surface of SiO₂[18]. In Raman spectra (**Fig. (2c)**), the LDH vibrational peaks of Mg-OH (443

cm⁻¹), Al-OH (780 cm⁻¹), NO₃⁻ (780 cm⁻¹) and (572 cm⁻¹) of the Mg₂Al-OH laminate were shown[19]. In the thermogravimetric (TG) curve of SiO₂ (**Fig. S1**), the major mass loss occurs below 175 °C, attributed to the removal of adsorbed water and

dehydroxylation of SiO₂[20]. There is a minor mass loss at 290 °C, followed by a continuous slow decrease in the mass of SiO₂. For solid SLDH-2 (Fig. S2), there is a slight increase in mass loss below 200 °C compared to SiO₂ alone, attributed to the loss of interlayer adsorbed water in the LDH[21]. For the mass loss between 200 °C and 400 °C attributed to the decomposition of metal-OH, the mass loss after 400 °C is attributed to the decomposition of interlayer anions. For mesoporous SLDH-2-2h after alkali etching (Fig. (2d)), the mass loss increases compared to SLDH-2, which is due to the partial removal of SiO₂ after alkali etching and the increase in the proportion of LDH in mesoporous SLDH-2-2h, which is greater than that of SiO₂ at the same temperature, and ultimately leads to an increase in mass loss of mesoporous SLDH-2-2h. Combined with the X-ray Fluorescence (XRF) analysis results for SLDH-2 and SLDH-2-2h (Table. S1), it was confirmed that the LDH structure was retained after alkali etching. To understand the chemical state of the catalyst surface, X-Ray photoelectron spectroscopy (XPS) under light was performed. Both the solid SLDH-2 and the etched mesoporous SLDH-2-2h (Figs. (2e) and (2f)) reveal signals from Mg, Al, Si, O, and N elements, indicating the successful preparation of the SiO₂@LDH core-shell structure. The O 1s XPS diagrams of SLDH-2-2h can be deconvoluted into three sub-peaks at binding energies of 531.2 eV, 532.1 eV, and 534.0 eV, corresponding to Mg-O, Al-O, and Si-O, which further confirmed the encapsulation of LDH on the SiO₂ surface (Fig. S3). In the high-resolution O 1s spectrum, signals from Mg-O and Al-O bonds are observable alongside the characteristic Si-O signal, with the Si-O signal exhibiting lower intensity relative to Mg-O and Al-O. This further confirms the coating of LDH on the SiO₂ surface[22]. First, SLDH-2 was prepared by electrostatic self-assembly method. The surface of SiO₂ spheres in ethanol is negatively charged with a zeta potential value of -14.50 mV, and LDH-NS is positively charged with a zeta potential value of 23.40 mV (Fig. (2g)). Therefore, electrostatic interactions may occur between the negatively charged SiO₂ spheres and the positively charged LDH-NS (Fig. (2h)). To further increase the difference in zeta potential values and enhance the interaction between SiO₂ and LDH-NS, anionic surfactants were also introduced to enhance the interaction between SiO₂ spheres and LDH-NS[23]. SiO₂, after treatment with anionic surfactants, undergoes electrostatic self-assembly with LDH-NS to form core-shell SiO₂@LDH.

3.2 Structural characterization of SiO₂@LDH-based PLs

The ¹H NMR spectrum of SLDH-2-2h-10%BF₄ (Fig. (3a)) displays the characteristic peaks of both surface -OH groups from SLDH-2-2h and the typical signals of [BMIM][BF₄], confirming the successful synthesis of the target material. Meanwhile, the Raman spectrum of SLDH-2-2h-10%BF₄ (Figs. (3b) and (3c)) exhibited vibrational peaks assignable to the

[BMIM]⁺ cation, including CN vibrations in CH₂(N) (624 cm⁻¹) and CH₃ (N) (652 cm⁻¹), out-of-plane modes of HCCH (755 cm⁻¹) and NC(H)NCH (843 cm⁻¹), an in-plane HCCH mode (1150 cm⁻¹), and C-H stretching vibrations of benzene rings (3120 and 3160 cm⁻¹). Additionally, the B-F stretching vibration of the [BF₄]⁻ anion was observed at 1030 cm⁻¹[24]. The vibration bands in the FTIR spectrum of SLDH-2-2h-10%BF₄ were similar to [BMIM][BF₄][25,26] (Fig. (3d)). The asymmetric stretching vibrational peaks of the imidazole ring CH₂-HCH at 3170 cm⁻¹, the symmetric stretching vibrational peaks of imidazole(N)CH₂-HCH at 3110 cm⁻¹, the stretching vibrational peaks of -CH₂- at 2970 and 2880 cm⁻¹, and the in-plane C-C and C-N stretching vibrational peaks of the imidazole ring at 1580 cm⁻¹ can be observed. In addition, the characteristic peaks near 1460 cm⁻¹ are attributed to the cyclic CH-CN stretching vibration, C-C stretching vibration, and asymmetric stretching vibration due to the C-H bond in the alkyl chain band, the characteristic peaks at 1030 cm⁻¹ are attributed to the in-plane vibration peaks of the alkyl chain, the characteristic peaks at 849 and 750 cm⁻¹ are attributed to the characteristic bands of B-F symmetric stretching vibration, the characteristic peaks at 653 and 623 cm⁻¹ are attributed to the bending vibration peaks of benzene ring, and the characteristic peak at 521 cm⁻¹ is attributed to the shear vibration of BF₄[27]. TG analysis of [BMIM][BF₄] and SLDH-2-2h-10%BF₄ (Figs. (3e) and (3f)) showed the same trend, originating from the decomposition of the ionic liquids at elevated temperatures, and the residue may be a solid carbide containing borates[28]. In particular, no considerable weight loss was shown below 300 °C, ascertaining its high thermal stability and low vapor pressure.

The surface elemental variations of SLDH-2-2h-10%BF₄ were studied deeply by analyzing XPS spectra. In the XPS broad scan spectrum (Fig. (3g)), the characteristic elements of [BMIM][BF₄] are observed, including C, B, and F elements. However, the characteristic elemental peaks of the pore generator SLDH-2-2h are significantly weakened, confirming the encapsulation of the pore generator by ILs. The XPS spectra (Fig. (3h)) indicated that C 1s peak at 284.80, 285.72, and 288.75 eV belonged to C=N, C-N and C-C groups, respectively[29]. The XPS signal peaks for the C element originate in the structure of ILs, further confirming the formation of SLDH-2-2h-10%BF₄. In the O 1s spectrum (Fig. (3i)), two peak strength signal peaks demonstrating the carboxyl group were observed at 531.1 eV (Mg-O), 532.1 eV (Al-O), and 533.4 eV (Si-O) as in the case of the nanoparticle SLDH-2-2h. This phenomenon indicated that the chemical environment of O elements in pore generators was not significantly altered after the encapsulation of ILs on the surface of SLDH-2-2h. Meanwhile, the stable existence of Si-O, Mg-O, and Al-O bonds proves the structural integrity of PLs and indicates that the coating of ILs did not destroy the original chemical bonding state of SLDH-2-2h.

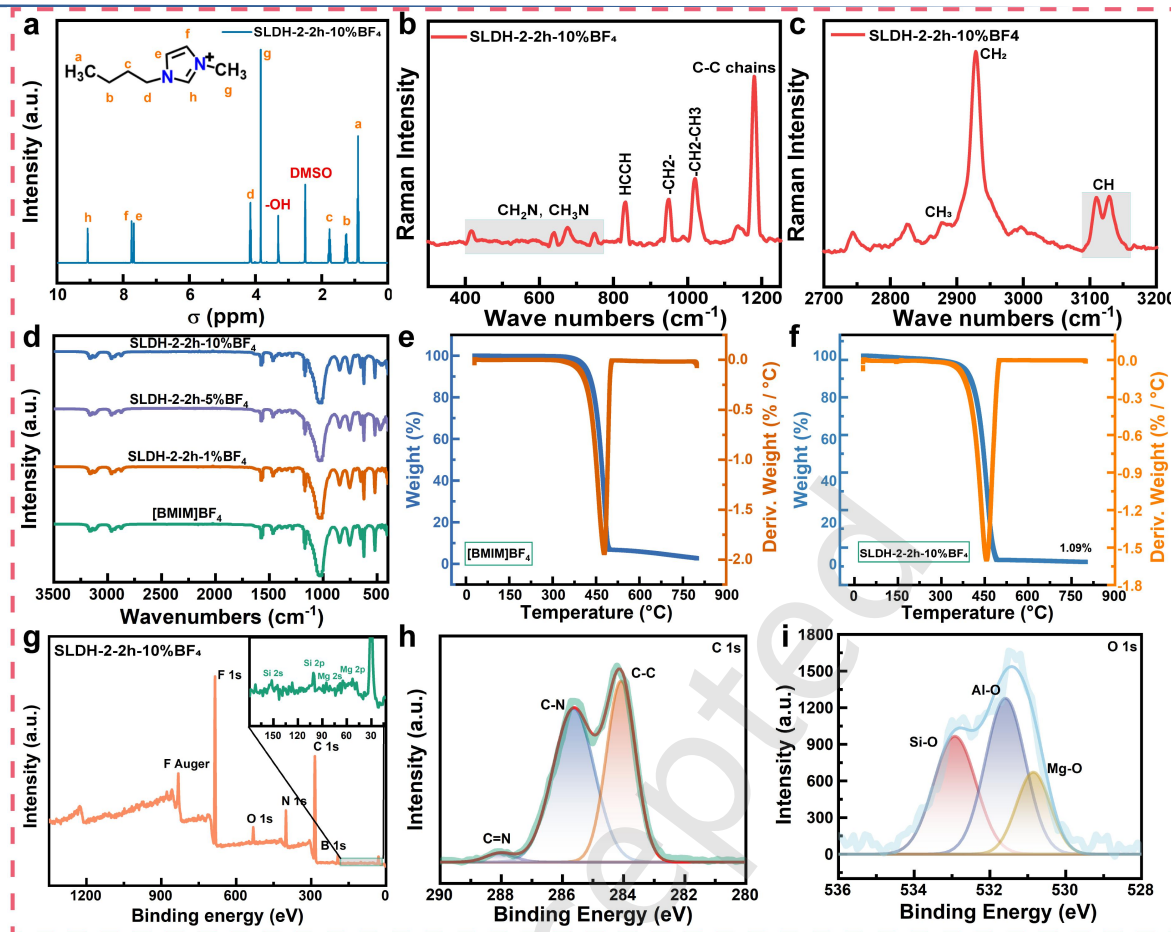


Figure 3 (a) ^1H NMR spectra of SLDH-2-2h-10% BF_4 , (b) low-wavenumber and (c) high-wavenumber Raman spectra, (d) FT-IR spectra of [BMIM][BF_4] and SLDH-2-2h-10% BF_4 , (e) TG curves of [BMIM][BF_4] and (f) SLDH-2-2h-10% BF_4 , (g) wide-scan spectra of SLDH-2-2h-10% BF_4 , (h) C 1s and (i) O 1s high-resolution spectra.

3.3 Morphology properties of $\text{SiO}_2\text{@LDH}$ and $\text{SiO}_2\text{@LDH}$ -based PLs

Furthermore, scanning electron microscope (SEM) was utilized to observe the sample morphology. The surface of the pristine SiO_2 nanospheres is smooth, and the particles are uniformly distributed and concentrated in size (Fig. (4a) and (4b)). Compared with the smooth SiO_2 nanospheres, the size of SLDH-2 was increased compared with that of SiO_2 , and the curled LDH-NS was dispersed on the surface of SiO_2 (Figs. (4c) and (4d)). The outer surface of SLDH-2 consisted of the edges of the LDH-NS, which agreed with the infrared mapping showing many exposed hydroxyl groups. The apparent morphology of SLDH-2-2h did not show any significant change after treatment with alkali (Figs. (4e) and (4f)). The LDH-NS on the outer surface remained, indicating that the exposed hydroxyl groups on the surface of SLDH-2-2h were not affected and used as sites for the subsequent interaction with sterically hindered solvents. The effect of alkali treatment on the morphology and pore structure of SLDH-2-2h was observed by TEM images. For the TEM image of solid SLDH-2 (Fig. (4g)), the internal SiO_2 presents a solid and uniform state. In contrast, the TEM image of mesoporous SLDH-2-2h (Fig. (4h)) presented many “white spots” in the internal SiO_2 , which are the pores inside the SiO_2 , indicating that the solid nanorods were etched into a porous structure. EDS analysis was used to characterize the elemental distribution of the SLDH-2 and

SLDH-2-2h, showing the composites have a representative core-shell structure. The elemental mapping shows the spatial distribution of Si, O, Mg, and Al. The core part is formed with strong Si and O signals, confirming the presence of the core ion SiO_2 . Meanwhile, Mg and Al elements are mainly distributed in the shell part, indicating the successful self-assembly of LDH-NS on the SiO_2 surface. Compare samples treated with alkali etching for 1 hour (SLDH -2-1h) and 4 hours (SLDH -2-4h). The surface of the SLDH -2-1h sphere is covered with a thin layer of LDH nanosheets, with a small number of nanosheets protruding at the edges (Fig. S4). The SiO_2 core has not been completely etched by the alkaline solution; no channels or cavity structures have formed, and thus the pore space required for carbon dioxide adsorption and storage is not available. The surface of the SLDH -2-4h microspheres is rough and uneven (Fig. S5). The originally ordered core-shell structure has completely collapsed, and the LDH no longer functions as an interface layer protecting the internal pores. ILs directly penetrate the exposed silica pores, hindering CO_2 capture (Fig. S6).

Both $\text{SiO}_2\text{-10\%BF}_4$ and SLDH-2-2h-10% BF_4 retain morphological features similar to SiO_2 nanoparticles (Fig. 5). For $\text{SiO}_2\text{-10\%BF}_4$, due to the thin IL coating layer on the SiO_2 surface, particle agglomeration persists, which will hinder the mass transfer process of CO_2 (Figs. 5(a-c)). Specifically, the IL coating on SLDH-2-2h-10% BF_4 is noticeably thicker. TEM images (Figs. 5(d-f)) reveal that the SLDH-2-2h core is

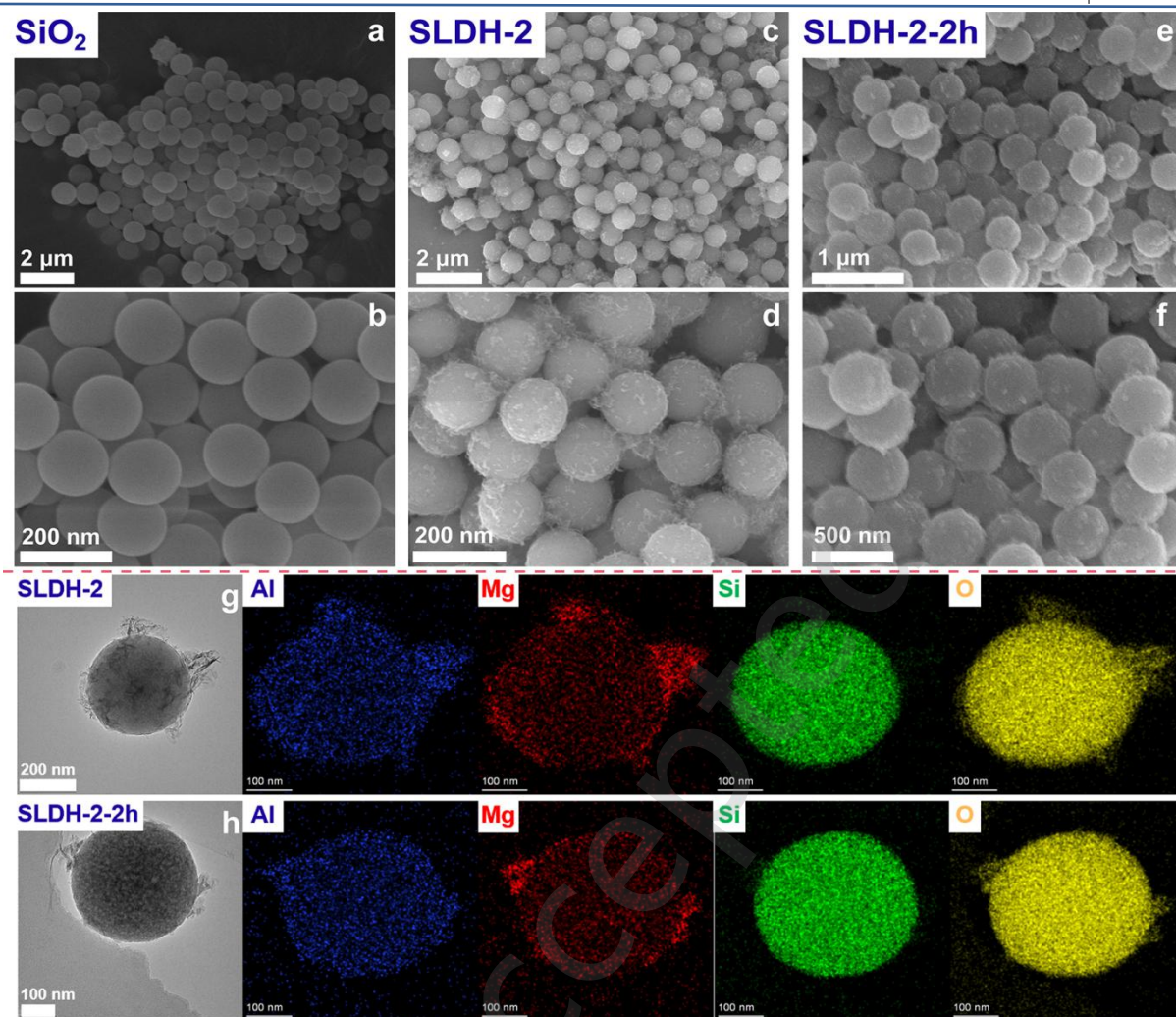


Figure 4 SEM images of (a, b) SiO_2 , (c, d) SLDH-2, and (e, f) SLDH-2-2h, and TEM images and element mapping diagram of (g) SLDH-2 and (h) SLDH-2-2h.

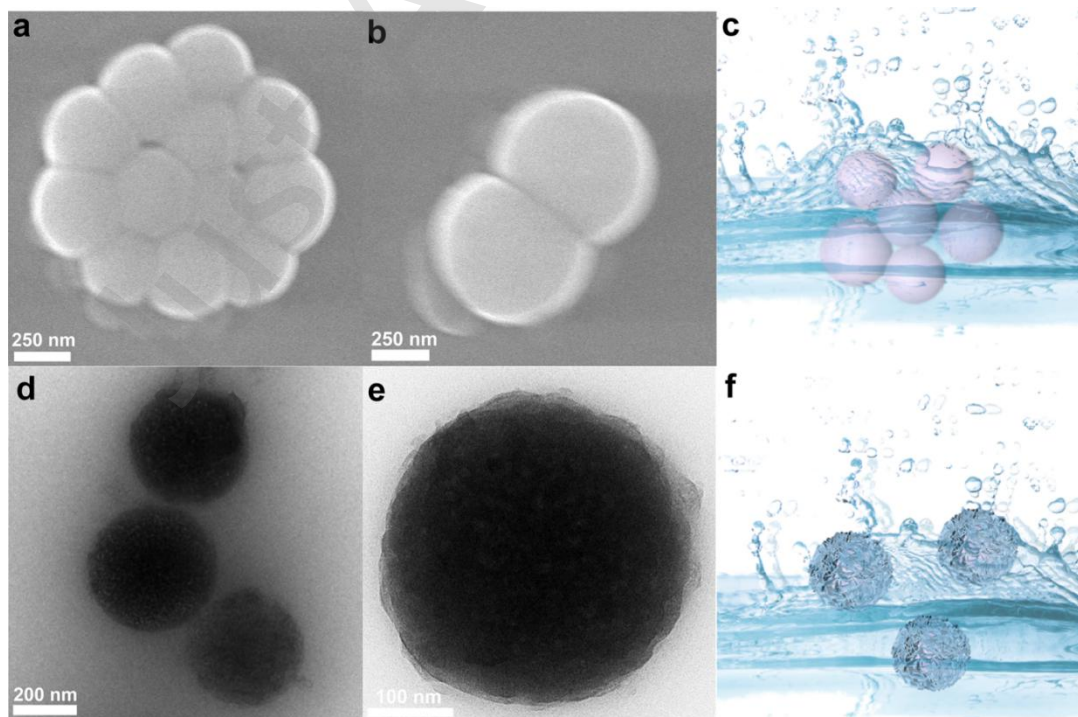


Figure 5 (a) (b) SEM images and (c) schematic of SiO_2 -10% BF_4 ; (d) (e) SEM images and (f) schematic of SLDH-2-2h-10% BF_4

encapsulated by a thicker IL shell, a phenomenon owing to hydrogen-bonding interactions between the abundant surface -OH groups of SLDH-2-2h and the IL. This thicker shell, combined with the spatial site resistance between IL molecules, results in relatively independent structures for SLDH-2-2h-10%BF₄₄, which helps preserve effective mass transfer pathways for CO₂ within the system. Furthermore, consistent with the observations for SLDH-2-h, “white dots” corresponding to the internal pore structure of mesoporous SiO₂ are also evident in TEM images of SLDH-2-2h-10%BF₄. This finding further confirms that the self-assembled LDH-NS layer on the SiO₂ surface functions as a protective shell, effectively preventing the ionic liquid from infiltrating the SiO₂ pores

3.4 Pore structure properties of SiO₂@LDH and SiO₂@LDH-based PLs

Furthermore, N₂ and CO₂ adsorption-desorption measurements were employed to analyze the influence of pore structure on the CO₂ capture performance of PLs. SiO₂, SLDH-2, and SLDH-2-2h exhibited type IV isotherms (Figs. (6a), (6d) and (6g))[30]. However, SiO₂ has the smallest N₂ capacity. The N₂ capacity of SLDH-2 increases slightly after coating LDH-NS on its surface. Then, N₂ capacity of SLDH-2-2h increases significantly after etching by alkali treatment, indicating that the SLDH-2-2h has greater pore volume and specific surface area than SLDH-2 and SiO₂ (Table S2 and S3). SiO₂, SLDH-2, and SLDH-2-2h exhibit

three distinct hysteresis loops (H2(b), H3, and H1, respectively), indicative of their distinct mesoporous structures (Figs. (6b), (6e) and (6h)). Specifically, the H2(b)-type loop for SiO₂ exhibits the presence of ink-bottle pores, typically observed in materials with limited pore connectivity (Fig. (6c)). The H3-type loop observed for SLDH-2 is associated with slit-like pores formed by plate-like particles, consistent with the LDH nanosheets within its structure (Fig. (6f)). Complementing this analysis, the pore size distribution reveals a predominant pore width of approximately 0.83 nm for SLDH-2-2h (Fig. (6i)), which is attributed to the interlamellar spacing of the cross-linked LDH nanosheets. This value aligns with the data presented in Table S4. Crucially, this pore size is larger than the kinetic diameter of a CO₂ molecule but smaller than that of the [BMIM][BF₄] ion pair. This unique pore size enables the LDH-NS to exclude the ionic liquid via a synergistic size-sieving and hydrogen-bonding effect, while simultaneously permitting the diffusion of CO₂.

SLDH-2-2h exhibits an H1+H3-type hysteresis loop, which is attributed to its uniform mesoporous structure of internal SiO₂ and the non-uniform pore structure of LDH-NS[31]. The ordered mesopores of SLDH-2-2h are mainly concentrated near 20 nm, consistent with the pore size distribution. The resulting mesopores are generated by the alkali etching process, which creates a porous silica structure. Since LDH-NS is stable in alkaline solution, OH⁻ passes through LDH-NS to etch SiO₂ into a porous structure. Due to the selective

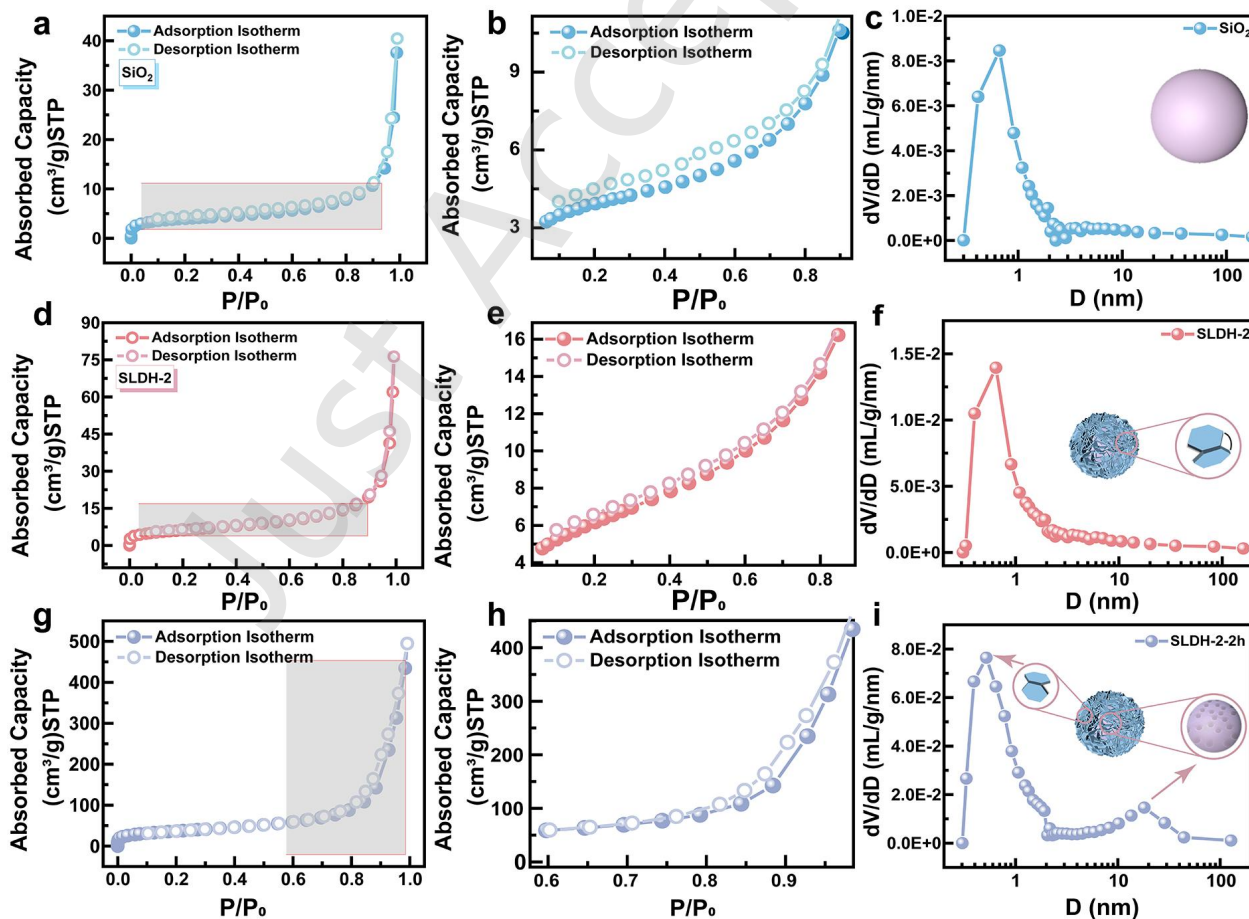


Figure 6 The (a) (d) (g) N₂ adsorption isotherms, (b) (e) (h) hysteresis loop, and (c) (f) (i) pore size distribution of SiO₂, SLDH-2 and SLDH-2-2h at 77 K.

permeability of LDH nanosheets, the enhanced porosity of SLDH-2-2h facilitates continuous mass transfer for CO₂ capture, which is supported by the comparative data in Table S4. The

post-synthetic method effectively mitigates gas mass transfer channel obstruction caused by LDH-NS coverage on the internal pore structure of SiO₂, an issue observed when porous SiO₂ is

synthesized before LDH-NS assembly. Therefore, the post-etching method preserves the inherent mesoporous structure of SiO_2 , which enhances gas mass transfer efficiency in PLs.

Since low-temperature N_2 adsorption techniques remain insufficient for accurately determining the specific surface area and pore volume of PLs (Detailed information is shown in SI), CO_2 was employed as a probe molecule to evaluate the ultramicroporous structure of SLDH-2-2h-10% BF_4 in this study. However, kinetic limitations in mass transfer at 273 K restricted the resolution of porosity data, preventing comprehensive structural analysis. As shown in **Figs. (7a) and (7b)**, the retention of ultramicropores centered at ~ 2 nm in SLDH-2-2h-10% BF_4 suggests partial mitigation of IL infiltration into the SiO_2 matrix, likely attributable to LDH-NS acting as a pore-gatekeeper. Since low-temperature N_2 adsorption techniques remain insufficient for accurately determining the specific surface area and pore volume of PLs (Detailed information is shown in SI), CO_2 was employed as a probe molecule to evaluate the ultramicroporous structure of SLDH-2-2h-10% BF_4 in this study. However, kinetic limitations in mass transfer at 273 K restricted the resolution of porosity data, preventing comprehensive structural analysis. As shown in Fig. 7a and 7b, the retention of ultramicropores centered at ~ 2 nm in SLDH-2-

2h-10% BF_4 suggests partial mitigation of IL infiltration into the SiO_2 matrix, likely attributable to LDH-NS acting as a pore-gatekeeper.

To further determine the geometric shape and size distribution of pores in PLs, wide-angle X-ray scattering (WAXS) analysis was performed on the curves of SLDH-2-10% BF_4 and SLDH-2-2h-10% BF_4 [32]. To visually observe the overall behavior of the scattering curves (**Figs. S7-S9**), $\log(I)$ - $\log(q)$ plots were first plotted, as shown in **Fig. (7c)**. The curves exhibited a general power-law decay trend, but the slope varied across different q intervals, suggesting distinct structural regions. Starting from $q \approx 0.033 \text{ \AA}^{-1}$, $I(q)$ decays rapidly with increasing q , a characteristic feature of porous materials. No distinct Bragg peaks were observed across the entire q range, indicating the absence of long-range ordered pore arrangements. Using Guinier analysis[33], a linear fit of $\ln I(q)$ versus q^2 was performed on data from approximately 0.03 to $0.06 \text{ \AA}^{-1}$. The calculated average diameter for SLDH-2-2h-10% BF_4 was approximately 21.5 nm, consistent with the average pore size of 22.7 nm for SLDH-2-2h. This confirms that the pore structure of SLDH-2-2h is preserved after PL formation. It is known that WAXS is sensitive to both open and closed pores,

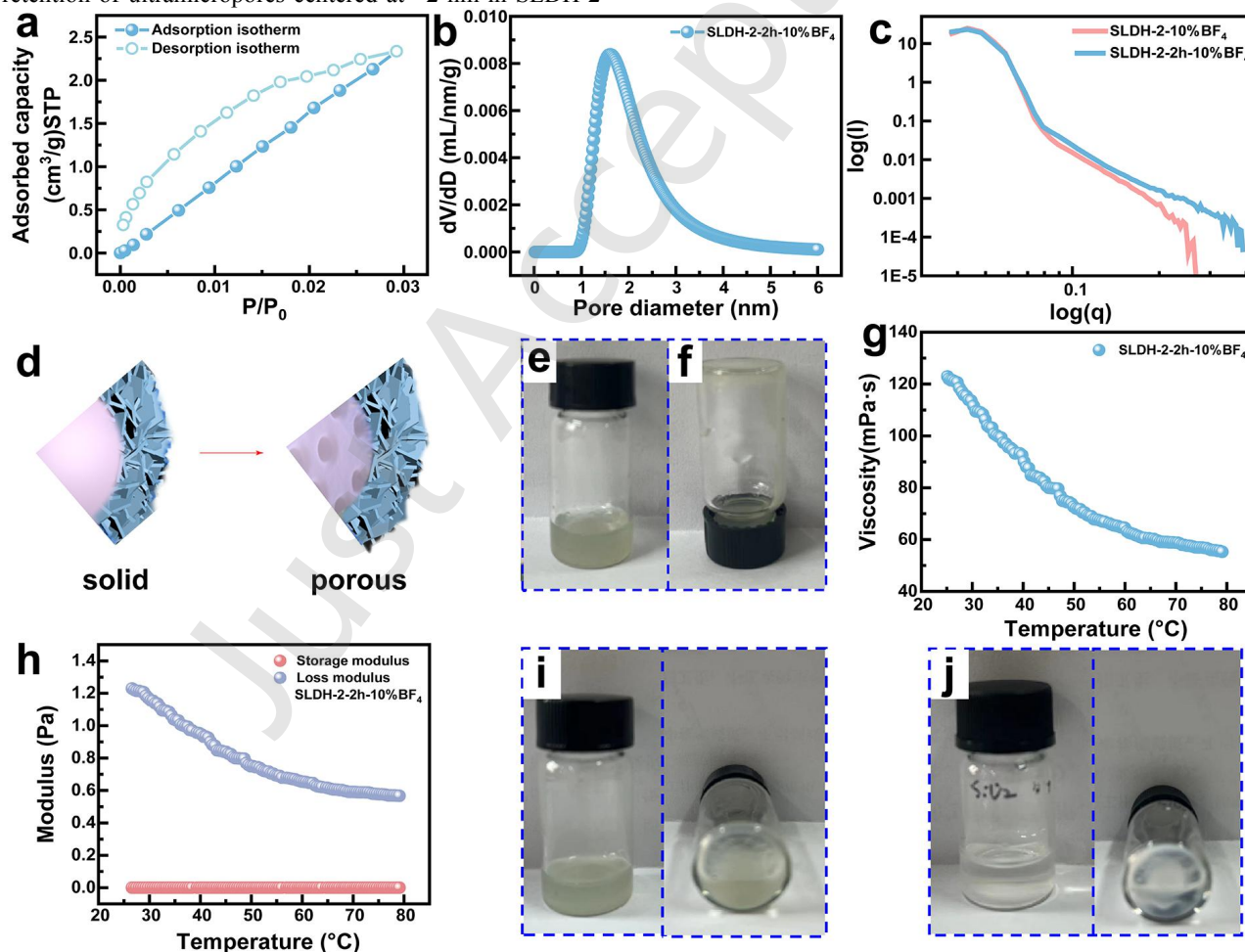


Figure 7 (a) CO_2 adsorption isotherms and (b) pore size distribution of SLDH-2-2h-10% BF_4 at 0 °C, (c) X-ray scattering results of SLDH-2-10% BF_4 and SLDH-2-2h-10% BF_4 in WAXS regions, (d) Pore diagrams of SLDH-2 and LDH-2-2h, (f) and (g) Flowability of SLDH-2-2h-10% BF_4 , (g) viscosity and (h) curve of modulus versus time of SLDH-2-2h-10% BF_4 , (i) SiO_2 -10% BF_4 and (j) SLDH-2-2h-10% BF_4 before and after one month of storage.

whereas BET typically detects only open pores. The absence of pore sizes larger than those detected by BET in the WAXS-calculated pore size distribution indicates that the pores in SLDH-2-2h-10% BF_4 are open. This is beneficial for enhancing

CO_2 mass transfer and improving the trapping capacity of PLs. Linear fitting of $\log I(q)$ and $\log q$ was further performed in the higher q range (e.g., $0.5 \text{ \AA}^{-1}$ to $2.5 \text{ \AA}^{-1}$), yielding a slope of $-\alpha$ [34]. The results reveal that the fractal characteristics ($\alpha \approx 3.0$)

of SLDH-2-2h-10%BF₄ strongly suggest a fractal internal structure, where pores and the solid framework are self-similar across multiple scales. In contrast, the solid SLDH-2-10%BF₄ exhibits a fractal dimension $\alpha \approx 3.8$ [35]. This indicates that the pore walls or pore-matrix interfaces possess rough, irregular self-similarity, while the overall framework remains compact (**Fig. (7d)**). This aligns with TEM observations, confirming the preservation of pores within SLDH-2-2h. SLDH-2-2h-10%BF₄ demonstrates good fluidity at room temperature (**Figs. (7e) and (7f)**). Rheological characterization further revealed that SLDH-2-2h-10%BF₄ has a viscosity of 0.12 Pa·s at 25 °C (**Fig. (7g)**), which is comparatively lower than that of many PLs reported in the literature (Table S5), highlighting its advantage in processability. Across the tested range, the loss modulus (G'') of SLDH-2-2h-10%BF₄ was significantly greater than its storage modulus (G') (**Fig. (7h)**), consistent with liquid-like behavior and confirming its excellent fluidity. After standing SiO₂-10%BF₄ at room temperature for one month, a significant precipitate was observed at the bottom of bottle (**Figs. 7e and 7f**). In contrast, SLDH-2-2h-10%BF₄ (**Fig. (7i)**) exhibited remarkable dispersion stability, showing no obvious precipitate under the same conditions. This enhanced stability is ascribed to the abundant hydroxyl groups (-OH) on the LDH nanosheets, which can form hydrogen bonds with both the BMIM⁺ cations and BF₄⁻ anions of ILs. The adsorption of these ions likely creates a stable solvation layer on the nanosheet surface, effectively preventing their close approach and aggregation (**Fig. (7j)**). Furthermore, the encapsulation by the ionic liquid also contributes to the favorable fluidity of SLDH-2-2h-10%BF₄. Furthermore, the TEM image (**Fig. S10**) of the samples after 6 months of storage confirmed that their core-shell nanostructures remained intact.

3.5 Evaluation of CO₂ Capture Performance

The SiO₂ showed a weak CO₂ capacity of 0.952 mmol/g at 10 bar and 25 °C (**Fig. (8a)**). After the self-assembly of LDH-NS on its surface, CO₂ capacity improved significantly. This trend aligns with changes in pore structure, indicating that the introduction of LDH-NS effectively optimized the SLDH-2's porosity characteristics, thereby enhancing CO₂ capacity (1.313 mmol/g). However, when the amount of LDH-NS was further increased, the CO₂ capacity of SLDH-3 decreased noticeably (0.514 mmol/g). This decline occurred because the excess LDH-NS failed to assemble properly on the SiO₂ surface and instead agglomerated by itself, which ultimately led to the significant drop in performance. By comparing the performance of the three PLs (SiO₂, SLDH-2, and SLDH-2-2h-based PLs), this study systematically investigated the influence of porous hosts structure on the overall performance of PLs. In SLDH-2-based PLs, CO₂ capture performance gradually improves with increasing SLDH-2 loading (**Fig. (8b)**). A further comparison of the PLs derived from SiO₂, SLDH-2, and SLDH-2-2h (**Fig. (8c)**)

shows their CO₂ capacities at 25 °C and 10 bar follow the sequence (**Fig. 8c**): SiO₂-10%BF₄ (0.952 mmol/g) < SLDH-2-10%BF₄ (1.075 mmol/g) < SLDH-2-2h-10%BF₄ (1.519 mmol/g). This trend suggests that the internal porous structure of SLDH-2-2h was effectively maintained into PLs, thereby creating a more favorable environment for CO₂ capture. As a result, the CO₂ capacity of SLDH-2-2h-10%BF₄ is higher than that of the previously reported PLs (Table S6). Furthermore, owing to the exceptionally low solubility of N₂ in ILs, all PLs exhibited weak N₂ capacity (**Figs. (8d) and (8e)**), with values generally below 0.150 mmol/g. This characteristic contributes to their enhanced CO₂/N₂ selectivity, achieving a value of 170 under simulated flue gas conditions with a CO₂:N₂ ratio of 15%/85% (**Fig. (8f)**). SLDH-2-2h-10%BF₄ shows excellent cycling stability at 25 °C and 10 bar. When regenerated only by vacuum at room temperature, it retains 97% of its original CO₂ capacity after 10 cycles (**Fig. (8g)**). This improvement comes from rich porous structures of SLDH-2-2h, which provides plenty of space for CO₂ physical adsorption storage. Also, the low viscosity of SLDH-2-2h-10%BF₄ helps CO₂ move more easily inside porous structures, which allows faster CO₂ diffusion and capture. It is worth noting that after 10 cycles, the LDH shell remained intact, confirming the structural stability of SLDH-2-2h-10%BF₄ (**Fig. S11**).

To further investigate the mechanisms involved in CO₂ capture, this study employed four classical isothermal adsorption models (Langmuir, Freundlich, Dubinin-Radushkevich (D-R), and Temkin) to fit experimental data from three samples ([BMIM][BF₄], SLDH-2-10%BF₄, SLDH-2-2h-10%BF₄). For the Langmuir model (**Fig. (9a)**), which assumes uniform monolayer adsorption, the R² values for all three samples (0.563–0.861) were relatively low, and both $q_{\max}$ and b_L exhibited negative values (Table S6). This indicates that CO₂ adsorption behavior on the sample surfaces does not conform to the uniform monolayer adsorption assumption, confirming significant surface heterogeneity[36]. The Freundlich model exhibited R² > 0.995 for all three samples, indicating multi-layer adsorption on a non-uniform surface (**Fig. (9b)**). This result supports a multi-layer adsorption mechanism occurring on a heterogeneous surface. The Freundlich constant k_F represents the adsorbent's capacity to adsorb CO₂ (Table S7). The k_F value for SLDH-2-2h-10%BF₄ (1.589) was higher than that for SLDH-2-10%BF₄ (1.230) and [BMIM][BF₄] (0.329), confirming that CO₂ adsorption is most favorable on SLDH-2-2h-10%BF₄[37].

For the D-R model (**Fig. (9c)**), the R² values for all three samples exceeded 0.98. The maximum adsorption capacity $q_{\max}$ calculated via this model for SLDH-2-2h-10%BF₄ reached 3.155 mmol/g (Table S8), representing an approximately 33% increase compared to SLDH-2-10%BF₄. This result confirms that the porous structure of SLDH-2-2h is preserved in SLDH-2-2h-10%BF₄. Moreover, this porous structure provides more CO₂ mass transfer pathways and

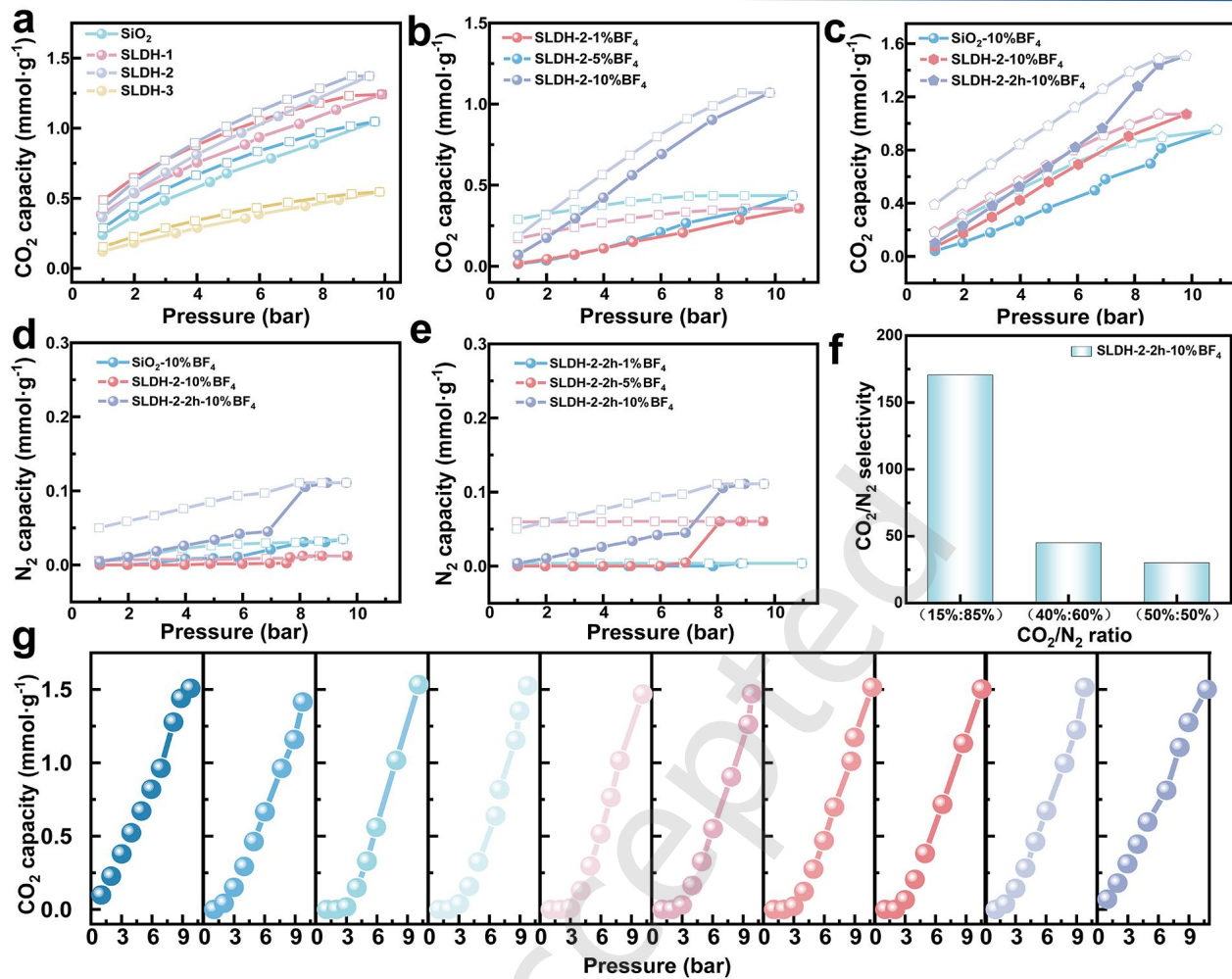


Figure 8 (a) (b) (c) CO₂ capacity of different materials, (d) (e) N₂ capacity of different materials, (f) CO₂/N₂ selectivity of SLDH-2-2h-10%BF₄, and (g) CO₂ cyclic capacity performance of SLDH-2-2h-10%BF₄.

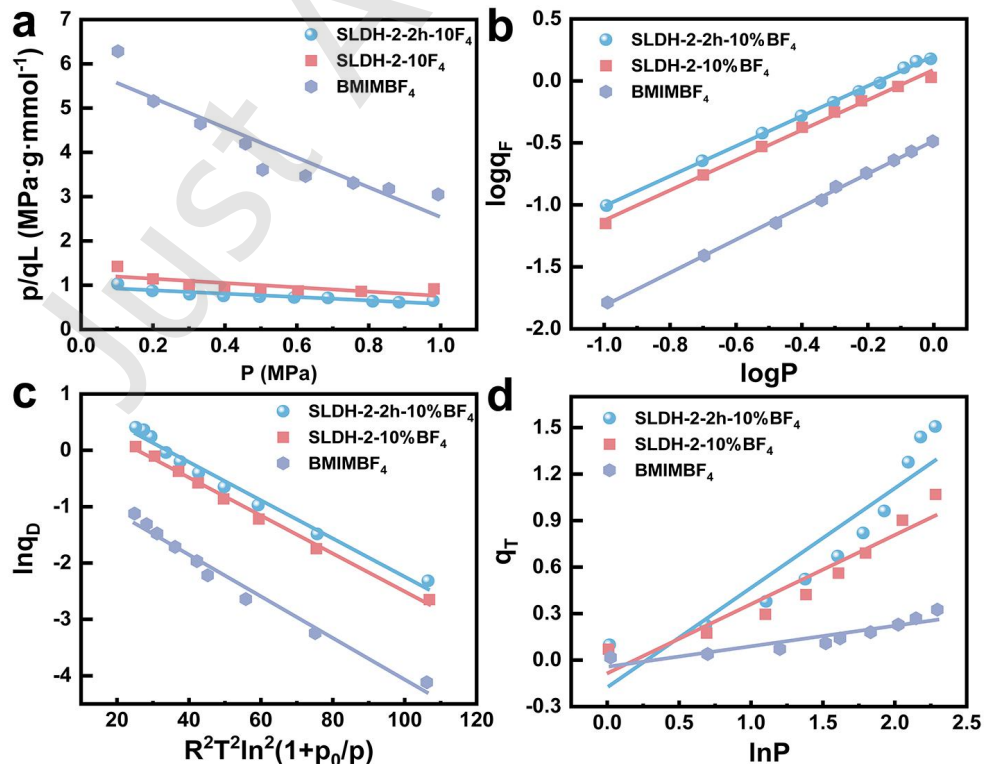


Figure 9 (a) Langmuir, (b) Freundlich, (c) D-R, and (d) Temkin model fitting of SLDH-2-2h-10%BF₄.

accessible CO₂ adsorption sites. Furthermore, the average adsorption free energy (*E*) values estimated using the D-R model ranged from 3.68 to 3.85 kJ/mol, falling within the typical range for physical adsorption (generally *E* < 8 kJ/mol)[38]. Among these, SLDH-2-2h-10%BF₄ exhibited a slightly higher *E* value, indicating marginally stronger adsorption forces. Regarding the Temkin model, the *R*² values for all three samples were moderate (0.845–0.909), reflecting interactions between adsorbate and adsorbent but not as the dominant mechanism (Fig. (9d)). The Temkin constant *b_T* exceeded 1 for all samples (Table S9), suggesting an exothermic adsorption process consistent with the conclusion that physical adsorption predominates[39]. The results confirm that [BMIM][BF₄] exhibits limited CO₂ capacity, but the porous structure within SLDH-2-2h-10%BF₄ significantly enhances CO₂ capacity and efficiency. The adsorption process is primarily characterized by non-uniform multilayer physical adsorption. This study provides crucial insights into the mechanisms and material design strategies for developing highly efficient CO₂ adsorbents.

4 Conclusion

This work successfully developed a stable PL based on bifunctional LDH interfacial engineering, resolving the long-standing core contradiction in PLs between pore accessibility and system stability. While maintaining high grafting efficiency on LDH, mesoporous SiO₂ was prepared within the LDH structure via post-etching, yielding a core-shell SiO₂@LDH-based PLs (SLDH-2-2h-10%BF₄). SLDH-2-2h-10%BF₄ exhibits significantly higher adsorption capacity (1.51 mmol/g at 10 bar and 25 °C) and excellent CO₂/N₂ selectivity compared to pure ionic liquid solvents. It also demonstrates outstanding cycling stability, retaining 97% of its maximum CO₂ capacity after 10 cycles. Research on the adsorption mechanism further indicates that the system follows multilayer adsorption according to the Freundlich isotherm model. Concurrently, the kinetic model confirms that the adsorption process is primarily governed by physical interactions. In summary, this work not only demonstrates the feasibility of constructing stable, highly efficient PLs through LDH interfacial engineering but, more importantly, provides a novel porous material design concept of “in-situ pore formation under interfacial protection”. Future work may further expand the application potential of such materials in gas separation, homogeneous catalysis, and chemical sensing by optimizing the chemical composition of LDH, controlling the microstructure of nanosheets, and matching functionalized solvents.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and/or the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

Xiaoqian Ju: Writing - original draft, Methodology, Data curation, Model fitting, Conceptualization. Xinbo Duan: Visualization, Methodology, Investigation. Xiangbo Feng: Supervision, Funding acquisition, Software, Resources. Xinghao He: Investigation, Formal analysis. Rundi Cui: Methodology, Data curation. Zhiyuan Yang: Supervision, Funding acquisition, Resources, Methodology, Investigation. Pu Guo: Investigation, Data curation. Huizhi Xue: Formal analysis, Data curation. Bowei Duan: Methodology, Investigation. Jiaming Hu: Methodology, Data curation. Chi He: Project administration, Methodology, Investigation, Funding acquisition, Conceptualization.

Use of AI statement

None.

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Electronic Supplementary Material

Dual-functional LDH interface engineered stable

SiO₂@LDH-based porous liquids for efficient CO₂ capture

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1. Materials

Tetraethyl orthosilicate (AR), sodium dodecyl benzene sulfonate (AR), NaOH (AR), $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ($\geq 99\%$), $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ ($\geq 99\%$), and $\text{NH}_3 \cdot \text{H}_2\text{O}$ (25%~28%) were purchased from Macklin. 1-Butyl-3-methylimidazolium tetrafluoroborate ($\geq 97\%$) were purchased from Aladdin Chemical Reagent Co., Ltd., China. N_2 and CO_2 with a purity of 99.99% were supplied from Shaanxi Kuai Te Gas Equipment Co., Ltd., China. All other chemicals were used directly without further purification.

2. Methods

2.1 X-Ray Diffraction (XRD)

XRD patterns were recorded on a Bruker D8 Advance diffractometer with Cu K radiation ($\lambda = 1.5406 \text{ \AA}$) at room temperature. Data were measured with 0.05° increments and 40 kV.

2.2 Fourier Transform Infra-Red Spectroscopy (FT-IR)

FT-IR measurements were performed using a Tensor 27 infrared spectrometer with a diamond-based attenuated total reflection unit in the wavenumber range of 400 cm^{-1} to 4000 cm^{-1} . The sample is mixed with potassium bromide (KBr) in a 100:1 ratio.

2.3 Raman Spectroscopy

Raman spectra of the samples were measured using a Renishaw Invia Raman Microscope. A green laser (532 nm) excited wavenumbers from 150 to 3200 cm^{-1} , with the excitation source being a He-Ne laser.

2.4 Nuclear magnetic resonance (NMR)

The structure of porous liquids was analyzed using nuclear magnetic resonance (NMR) spectroscopy. The measurements were performed on a Bruker Avance NEO 600 MHz spectrometer in Germany, with deuterated DMSO as the solvent.

2.5 X-ray photoelectron spectroscopy (XPS)

The elemental composition of the solid samples was verified using X-ray photoelectron

spectroscopy (XPS) (Kratos Axis Ultra DLD). The binding energy range was 0–700 eV with an energy step size of 0.050 eV, using an Al K α X-ray source. XPS testing of porous liquids was performed on a Thermo Scientific ESCALAB 250Xi instrument, with high-resolution scans conducted for C and O elements. The binding energy range was 0–800 eV with an energy step size of 0.050 eV, and the X-ray source was Al K α .

2.6 Thermogravimetric Analysis (TGA)

TGA was performed under nitrogen atmosphere with a heating rate of 20 °C/min using a SDT Q600 V8.0 Build 95 thermal analyzer. The samples were heated to 750 °C using N₂ (100 mL/min) as the protective gas.

2.7 Scanning Electron Microscope (SEM)

Morphological analysis was performed using a field emission SEM model JSM-7610F from JEOL Ltd. An acceleration voltage of 15 kV was selected, and magnification was adjusted according to the sample dimensions.

2.8 Transmission Electron Microscope (TEM)

Morphological characterization was performed using a FEI Talos F200X TEM. A 200 kV electron beam acceleration voltage was employed, with magnification adjusted according to sample dimensions. Selected area electron diffraction (SAED) analysis of TEM images enables in situ correlation between microstructural observations and crystallographic properties, facilitating simultaneous characterization of morphological features and crystallographic attributes in crystalline samples.

2.9 Rotational Rheometer

The storage modulus, loss modulus, and viscosity of nanofluids were measured using a DHR-20 rotational rheometer from the United States. The angular frequency was set at 10 rad/s, with the temperature ramped from 25 °C to 80 °C at a heating rate of 0.15 °C/s. The oscillation displacement was 0.000255 rad.

2.10 Wide-Angle X-ray Scattering (WAXS)

Wide angle X-ray scattering (WAXS) experiments were carried out in the laboratory using a Cu anode X-ray generator (Xeuss 3.0) equipped with a Dectris Pilatus 2D detector. The data was then radially averaged to 1D. Analysis of the scattering scans was done after careful background correction.

2.11 Gas Uptake

The samples were dried overnight at 100 °C in a vacuum oven prior to testing. Gas adsorption isotherms were determined using a Best High-Temperature High-Pressure Adsorption Analyzer (3H-2000PH). All measurements employed ultra-high-purity (99.99%) N₂ and CO₂. A water bath was used to control adsorption temperatures (298 K, 308 K, and 318 K were employed in this study).

2.12 Nitrogen adsorption-desorption testing at -196 °C

Samples were analyzed using N₂ adsorption-desorption at -196 °C. A Micromeritics ASAP 2460 adsorption analyzer (USA) was employed, with vacuum degassing at 120 °C for 6 h. Sample loading was approximately 0.2 g.

2.13 CO₂ adsorption-desorption testing at 0 °C

Microporosity analysis of samples was performed using CO₂ adsorption-desorption at 0 °C. Testing was conducted on a Beijing BSD-660M A3MB3M adsorption analyzer. Prior to testing, samples underwent vacuum degassing at 60 °C for 2 h. Dead volume in the temperature zone was measured before testing, followed by vacuum degassing to eliminate He contamination.

3. WAXS Parameter Analysis

3.1 Guinier analysis

Guinier analysis is applicable for estimating the size of isolated scatterers[1] (Eq. S1). Perform a linear fit of $\ln I(q)$ versus q^2 for data in the range of approximately 0.03 to 0.06 Å⁻¹.

$$\ln(I) \approx \ln(I(0)) \times e^{(-q^2 R_g^2 / 3)} \quad (\text{Eq. S1})$$

Where R_g is the radius of gyration, and $I(0)$ is the intensity at zero scattering angle ($q = 0$). The exponential term in the Guinier approximation can be determined by linear fitting the plot of $\ln(I)$ versus q^2 . This plot is known as the Guinier plot. The region selected for linear fitting is termed the “Guinier region.” Guinier fitting provides information about the overall molecular size and the quality of the data.

3.2 Porod's Equation

The high- q region of WAXS typically follows Porod's equation[2] (Eq. S2):

$$I(q) \propto q^{-\alpha} \quad (\text{Eq. S2})$$

$\alpha \approx 4$: Smooth pore interface (classical Porod behavior); $3 < \alpha < 4$: Fractal surface (surface roughness); $\alpha < 3$: Mass fractal (e.g., pores formed by nanoparticle aggregates).

4 Adsorption Model Fitting

4.1 Langmuir model

The Langmuir model is based on single-layer surface adsorption, assuming all adsorption sites are identical and adsorbed particles are completely independent.[3] Its expression is shown in Eq. S3:

$$q_L = q_{\max} \frac{b_L P}{1 + b_L P} \quad (\text{Eq. S3})$$

In the equation: q_L denotes the gas adsorption capacity at different pressures, mmol/g; $q_{\max}$ denotes the maximum adsorption capacity, mmol/g; b_L denotes the adsorption equilibrium constant; p denotes the adsorption pressure, MPa.

4.2 Freundlich model

The Freundlich isothermal adsorption model is one of the most widely applied models for non-ideal adsorption and multilayer adsorption on heterogeneous surfaces, with its expressions given by Eq. S4 and Eq. S5[4]:

$$q_F = k_F P^{1/n_F} \quad (\text{Eq. S4})$$

$$\ln q_F = \ln k_F + \frac{1}{n_F} \ln p \quad (\text{Eq. S5})$$

In the equation: q_F represents the gas adsorption capacity at different pressures, mmol/g; P denotes the adsorption pressure, MPa; k_F is the Freundlich constant, indicating the adsorbent's capacity to adsorb CO_2 [5]. A higher k_F value indicates more effective adsorption. n_F is the adsorption intensity constant; when n_F falls between 1 and 10, it indicates favorable adsorption conditions[6].

4.3 Dubinin-Radushkevich model

The Dubinin-Radushkevich (D-R) isotherm adsorption model was employed to determine adsorption properties [7]. The adsorption isotherm equations are related to Eq. S6, Eq. S7, and Eq. S8:

$$q_D = q_{\max} \cdot e^{(-\beta \cdot \varepsilon^2)} \quad (\text{Eq. S6})$$

$$\varepsilon = RT \ln(1 + Pr) \quad (\text{Eq. S7})$$

$$E = \frac{1}{\sqrt{2\beta}} \quad (\text{Eq. S8})$$

β and ε are adsorption energy parameters and energy parameters, $\text{mmol}^2/\text{kJ}^2$; q_D is the adsorption amount of the nanofluid at different pressures, mmol/g; $q_{\max}$ is the saturated adsorption amount, mmol/g; e is the equilibrium Polanyi potential related to equilibrium concentration, kJ/mol; R is the ideal gas constant, $8.314 \text{ J}/(\text{mol} \cdot \text{K})$; Pr is the relative pressure; T is the adsorption temperature, K; E is the mean free energy of adsorption, kJ/mol [8]. In the D-R model, $E < 8 \text{ kJ/mol}$ indicates physical interactions in the system, $8 \text{ kJ/mol} < E < 16 \text{ kJ/mol}$ indicates ion exchange or surface complexation, and $E > 16 \text{ kJ/mol}$ indicates chemical interactions.

4.4 Temkin model

The Temkin isothermal adsorption model was employed to investigate the interactions between adsorbents and adsorbates [9]. This model assumes that the adsorption heat of all molecules in the layer decreases linearly with coverage due to interactions between adsorbents and adsorbates. The isothermal curve model is expressed as Eq. S9 and Eq. S10.

$$q_T = B_T \ln(k_T P) \quad (\text{Eq. S9})$$

$$B_T = \frac{RT}{b_T} \quad (\text{Eq. S10})$$

In the equation: q_T denotes the adsorption capacity of the nanofluid at different pressures, mmol/g; P represents the relative pressure, bar; R is the gas constant; B_T is the dimensionless constant related to the adsorption heat; b_T is the constant associated with the adsorption heat, kJ/mol; k_T is the constant corresponding to the maximum binding energy [10]. In the Temkin model, if $b_T < 1$ kJ/mol, it indicates an exothermic reaction in the system, where physical adsorption dominates; if $b_T > 1$ kJ/mol, it indicates an endothermic reaction in the system, where chemical adsorption dominates.

5 Data Analysis

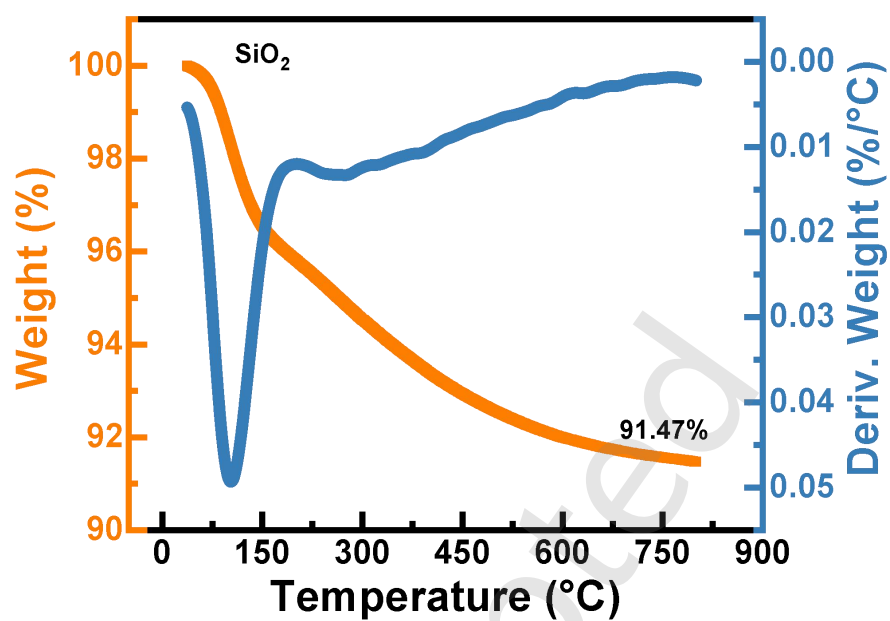


Fig. S1 TG curve of SiO_2 .

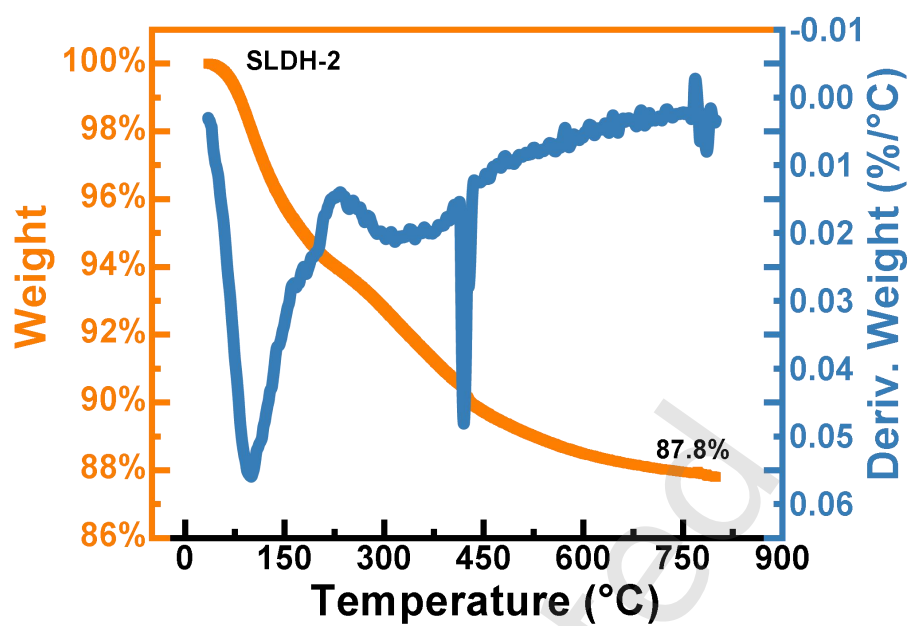


Fig. S2 TG curve of SLDH-2.

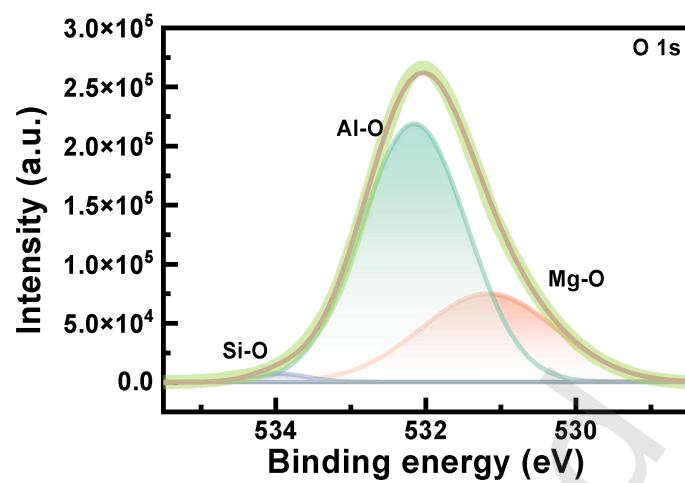


Fig. S3 O 1s high-resolution spectra of SLDH-2-2h.

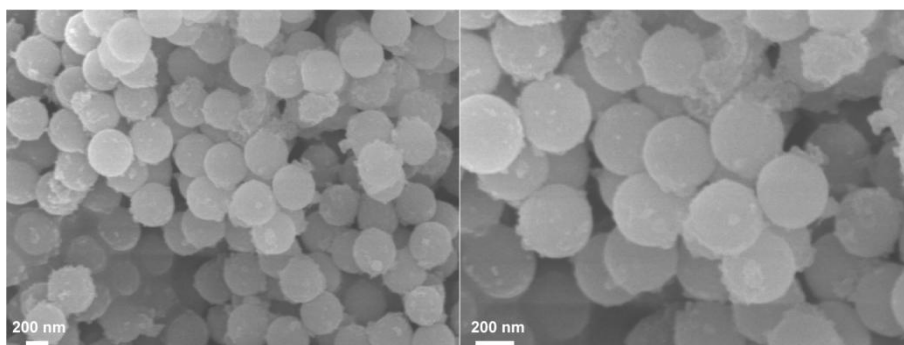


Fig. S4 SEM images of SLDH-2-1h.

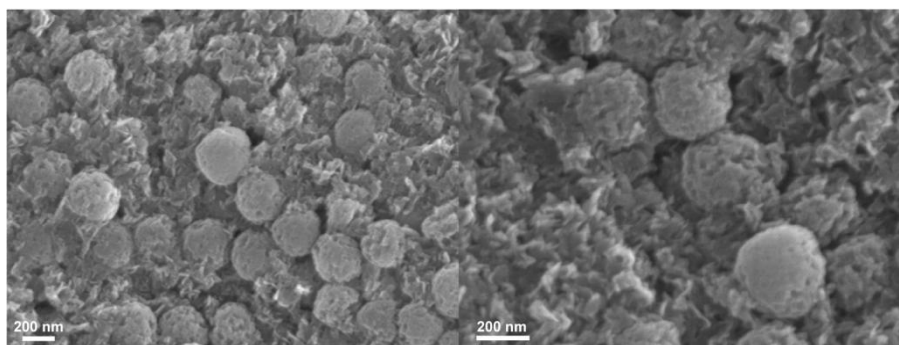


Fig. S5 SEM images of SLDH-2-4h.

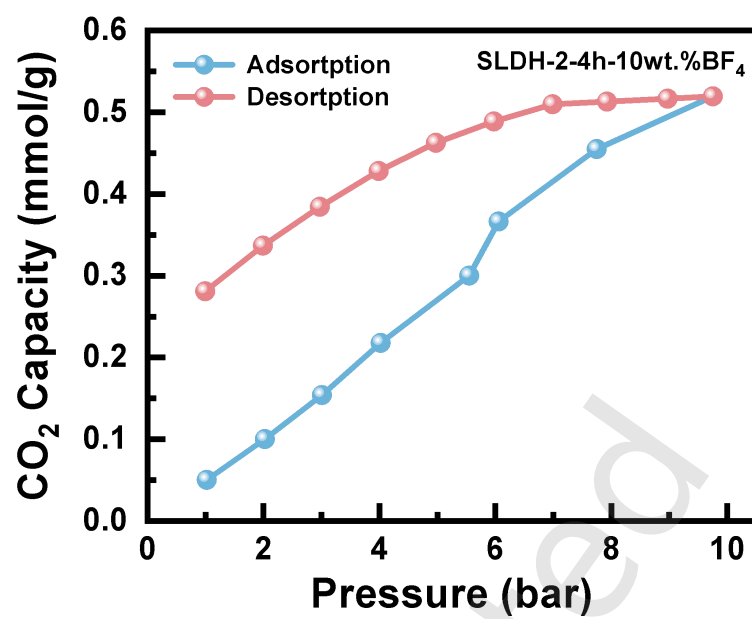


Fig. S6 CO₂ adsorption isotherms of SLDH-2-4h-10wt.%BF₄.

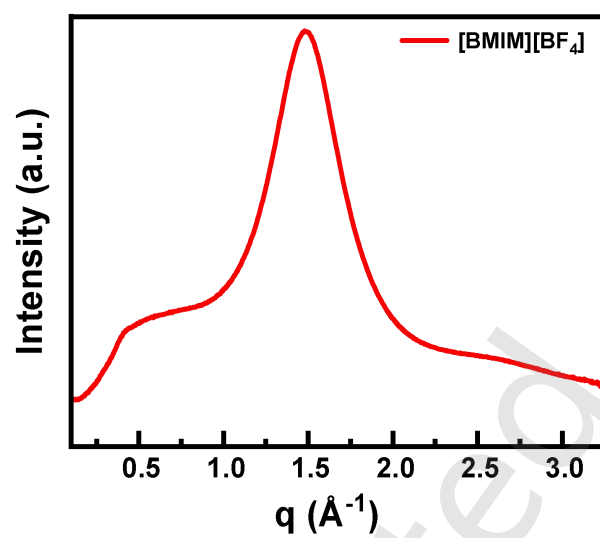


Fig. S7 Scattering curve of wide-angle X-ray scattering for BMIMBF₄.

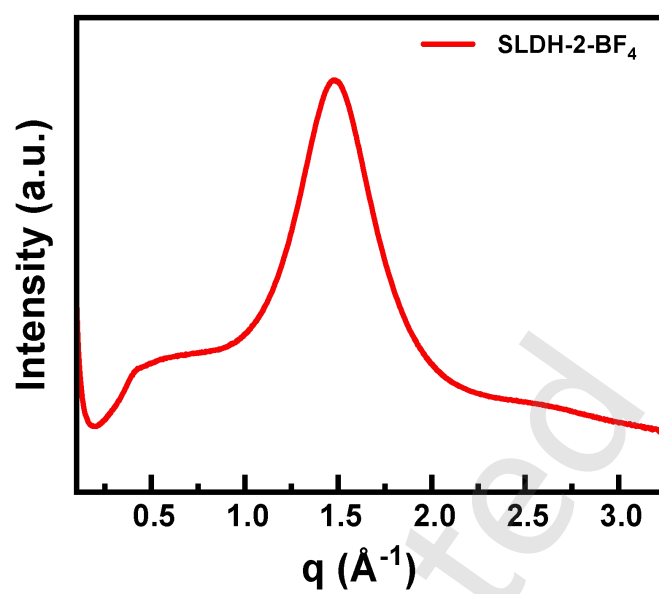


Fig. S8 Scattering curve of wide-angle X-ray scattering for SLDH-2-10%BF₄.

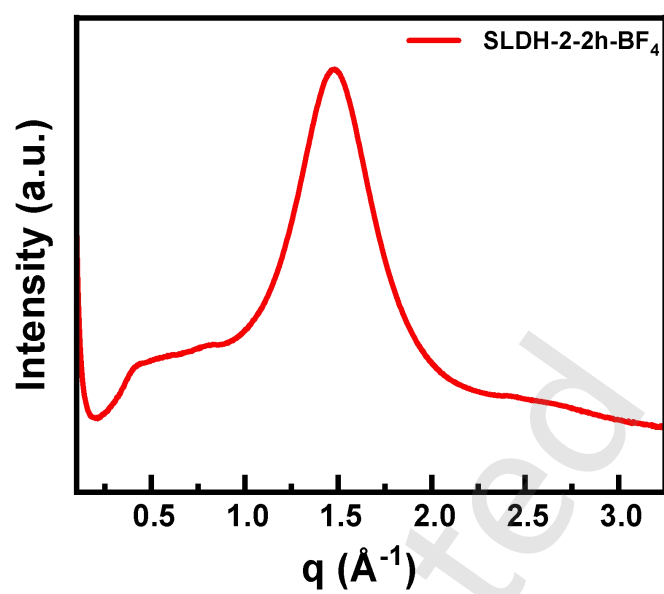


Fig. S9 Scattering curve of wide-angle X-ray scattering for SLDH-2-2h-10%BF₄.

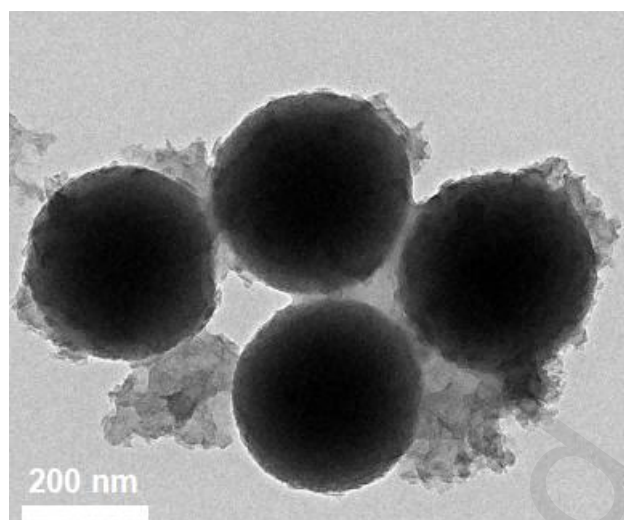


Fig. S10 The TEM image of SLDH-2-4h-10wt.%BF₄ on long-term stability for 6 months.

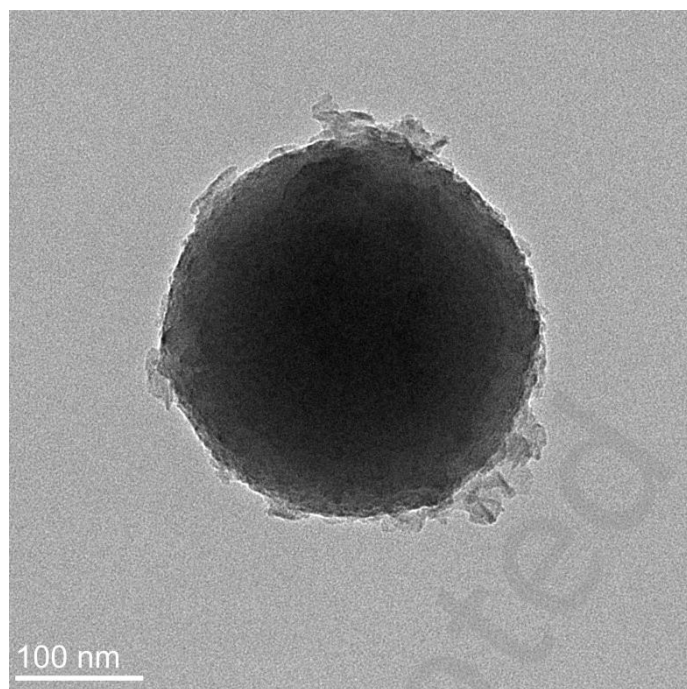


Fig. S11 TEM characterization on SLDH-2-2h-10wt.%BF₄ after ten cycles of CO₂ adsorption

Table S1 The comparison of specific surface area of SiO₂, SLDH-2 和 SLDH-2-2h

Samples	S _{BET} * (m ² /g)	S _{micro} * (m ² /g)	S _{meso} * (m ² /g)
SiO ₂	14.1917	6.5896	12.1948
SLDH-2	22.1280	2.8911	22.9679
SLDH-2-2h	131.9687	19.4652	161.4303

*Note: BET specific surface area (S_{BET}) was calculated by the BET multipoint method (automated range P/P₀ = 0.0380-0.1751); microporous specific surface area (S_{micro}) was calculated by the T-Plot method; mesoporous specific surface area (S_{meso}) was calculated by the BJH method of desorption (1.7-195.6 nm).

Table S2 The comparison of pore volume of SiO₂, SLDH-2 和 SLDH-2-2h

Samples	V _{total} * (m ³ /g)	V _{meso} * (m ³ /g)	V _{micro} * (m ³ /g)
SiO ₂	0.0615	0.0576	0.0028
SLDH-2	0.1090	0.1080	0.0011
SLDH-2-2h	0.7502	0.7414	0.0073

*Note: Calculation conditions for total pore volume (V_{total}) (P/P₀ = 0.990, pore diameter < 195.6 nm); mesopore volume (V_{meso}) was calculated using the BJH method (1.7–195.6 nm); micropore volume (V_{micro}) was calculated using the T-Plot method.

Table S3 The comparison of pore size of SiO₂, SLDH-2 和 SLDH-2-2h

Samples	Average pore size* (nm)	Mesopore size* (nm)	Microporous pore size* (nm)
SiO ₂	18.8933	18.8933	0.8418
SLDH-2	19.7035	18.8089	0.8335
SLDH-2-2h	22.7387	18.3708	0.8767

*Note: The average pore size was calculated using the BET method at 4V/A; the mesopore size (PS_{meso}) was determined from the desorption curve using the BJH method; the micropore size (PS_{micro}) was calculated using the H-K (Original) method.

Table S4 The comparison of viscosities of PLs in previous studies and our work.

PLs	Pore generators	Sterically hindered solvents	Type	Viscosities (Pa·s at 25 °C)	Ref.
POC PLs	CC15-R	Hexachloropropene	III	4.18	[11]
PL-1	M-UX	[Hmim]Br	III	0.58	[12]
P-UiO-66-PL-8	P-UiO-66	PDMS6000	III	0.54	[13]
HS PL	HS	Organosilane and oligomer	I	0.66	[14]
UiO-66-OH PLs	UiO-66-OH	Polyether amine	I	14.0	[15]
MOF liquids	ZIF-4	NA	I	8.0 (567 °C)	[16]
UiO-66 PL	D2000@ UiO-66	[M2070] [IPA]	III	4.0	[17]
ZIF-based PL	ZIF-67	[C ₆ Blm ₂] [NTf ₂]	III	0.54	[18]
UiO-66-liquid-M2070 PL	UiO-66	Organo silane and oligomer	III	4.5	[19]
MOF PL	UiO66@PDMS	PDMS	III	6.0	[20]
Zeolites PL	Zeolites	[P _{6,6,6,14}] [Br]	III	9.55	[21]
MOF PL	ZIF-67	N-heterocyclic carbene ligands	III	0.16	[22]
POC PL	Crown-ether cage	CH ₃ Cl PCP	III	0.02	[23]
ZIF-8/Water PL	ZIF-8	Water	III	0.013	[24]
UiO-66@ZIF-8-PL	UiO-66@ZIF-8	[EMIM][NTf ₂]	III	0.08	[25]
SLDH-2-2h-10 %BF ₄	SLDH-2-2h	BMIMBF ₄	Type III	0.12	This work

Table S5 Comparison of CO₂ Capacity of PLs Reported in the Literature

Materials	Porous sources	Type	Conditions	CO ₂ Uptake (mmol·g ⁻¹)	Ref.
P5-PL-8%	CuMOP	Direct Mixing	298 K, 1 bar	0.13	[26]
ZIF-90-NNa-PL	ZIF-90	Crown ether coating	303.2 K, 1 bar	0.56	[27]
PL-3	UiO-66-NH ₂	Direct Mixing	303 K, 20 bar	25.47 cm ³ ·g ⁻¹	[28]
UiO-66@OS@PEGS	UiO-66	Silanization	298 K, 10 bar	14.3 cm ³ ·g ⁻¹	[29]
ZIF-8 porous liquid (30%)	ZIF-8	Direct Mixing	RT, 10 bar	1.54	[30]
UiO-66@SiO ₂ -B-PL	UiO-66@SiO ₂	Core-shell	303 K, 2.5 bar	0.62	[31]
ZIF-67(S)@ZIF-8-PLs	ZIF-67(S)@ZIF-8	Core-shell	298 K, 10 bar	1.37	[32]
MCM-410-3%	MOF-Derived Carbon	Carburization	298 K, 10 bar	1.89	[33]
Br-ZIF-PL	ZIF-8	Core-shell	298 K, 10 bar	0.715	[34]
PL-UiO@SiO ₂ -2	UiO-66@SiO ₂	Core-shell	298 K, 2.5 bar	0.82	[35]
ZIF-8-PL	ZIF-8	Direct Mixing	298 K, 10 bar	0.81	
Silicalite-1-PL	Silicalite-1	MXene coating	298 K, 10 bar	0.46	[36]
ZSM-5-PL	ZSM-5	Silanization	298 K, 10 bar	0.38	
PL-3	MXene/UiO-66	Direct Mixing	298 K, 10 bar	0.52	[12]
UiO-66-liquid	UiO-66	Silanization	298 K, 30 bar	0.86	[15]
Hollow silica PLs	Hollow silica	Direct Mixing	298 K, 25 bar	2.2182	[37]
PL-2	Hypercross linked polymers	Direct Mixing	298 K, 18 bar	1.6027	[38]
UiO-66-OH (4)-PL	UiO-66-OH	Silanization	298 K, 10 bar	0.32	[39]
SLDH-2-2h-10%BF₄	SLDH-2-2h	Dual functional LDH interface	298 K, 10 bar	1.51	This work

Table S6 Fitting parameters of the Langmuir model

Samples	Langmuir model						R^2
	$1/q_{\max}$	SE	$1/(b_L \cdot q_{\max})$	SE*	bL	$q_{\max}$ (mmol/g)	
SLDH-2-2h-10%BF ₄	-0.384	0.059	0.963	0.036	-2.507	-2.604	0.840
SLDH-2-10%BF ₄	-0.486	0.175	1.243	0.097	-2.556	-2.056	0.563
[BMIM][BF ₄]	-3.366	0.511	5.904	0.310	-1.754	-0.297	0.861

*SE: Standard Error

Table S7 Fitting parameters of the Frundlich model

Samples	Frundlich model						
	1/n _F	n _F	SE	logk	k _F	SE	R ²
SLDH-2-2h-10%BF ₄	1.211	0.825	0.015	0.201	1.589	0.007	0.999
SLDH-2-10%BF ₄	1.215	0.823	0.034	0.090	1.230	0.017	0.995
[BMIM][BF ₄]	1.330	0.752	0.020	-0.483	0.329	0.009	0.998

*SE: Standard Error

Table S8 Fitting parameters of the D-R model

Samples	D-R model						
	β	SE	$\ln q_{\max}$	$Q_{\max}$ (mmol/g)	SE	E (kJ/mol)	R^2
SLDH-2-2h-10%BF ₄	-0.034	0.002	1.149	3.155	0.083	3.839	0.984
SLDH-2-10%BF ₄	-0.034	0.001	0.865	2.375	0.056	3.852	0.995
[BMIM][BF ₄]	-0.037	0.002	-0.375	0.687	0.111	3.680	0.980

*SE: Standard Error

Table S9 Fitting parameters of the Temkin model

Samples	Temkin model						
	B_T	SE	$B_T \cdot \ln K_T$	SE	K_T	b_T (kJ/mol)	R^2
SLDH-2-2h-10%BF ₄	0.643	0.089	-0.176	0.147	0.026	3.857	0.867
SLDH-2-10%BF ₄	0.446	0.058	-0.086	0.089	0.006	5.553	0.909
[BMIM][BF ₄]	0.132	0.021	-0.043	0.035	0.046	18.735	0.845

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