High-Throughput Discovery of Ni(IN)$_2$ for Ethane/Ethylene Separation

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Although ethylene (C$_2$H$_4$) is one of the most critical chemicals used as a feedstock in artificial plastic chemistry fields, it is challenging to obtain high-purity C$_2$H$_4$ gas without any trace ethane (C$_2$H$_6$) by the oil cracking process. Adsorptive separation using C$_2$H$_6$-selective adsorbents is beneficial because it directly produces high-purity C$_2$H$_4$ in a single step. Herein, Ni(IN)$_2$ (HIN = isonicotinic acid) is computationally discovered as a promising adsorbent with the assistance of the multiscale high-throughput computational screening workflow and Computation-Ready, Experimental (CoRE) metal–organic framework (MOF) 2019 database. Ni(IN)$_2$ is subsequently synthesized and tested to show the ideal adsorbed solution theory (IAST) selectivity of 2.45 at 1 bar for a C$_2$H$_6$/C$_2$H$_4$ mixture (1:15), which is one of the top-performing selectivity values reported for C$_2$H$_6$-selective MOFs as well as excellent recyclability, suggesting that this material is a promising C$_2$H$_6$-selective adsorbent. Process-level simulation results based on experimental isotherms demonstrate that the material is one of the top materials reported to date for ethane/ethylene separation under the conditions considered in this work.

Introduction

C$_2$H$_4$ (ethylene) is one of the most important olefins for the industrial mass production of commercial polymer materials and high valued chemicals.\(^1\) C$_2$H$_4$ is produced through a steam cracking process, where C$_2$H$_6$ (ethane) is the primary feedstock for the reaction. The effluent of the steam cracking process contains 5–10% of unreacted C$_2$H$_6$, which must be separated to obtain high-purity C$_2$H$_4$ for the polymerization reaction. However, the similarity in molecular size and volatility of C$_2$H$_6$ (kinetic diameter 4.16 Å; boiling point 169.42 K) and C$_2$H$_4$ (kinetic diameter 4.44 Å; boiling point 184.55 K) makes the separation of the two molecules a challenge.\(^2\) To overcome the limitation of the technology, researchers have highly sought and intensively investigated effective separation methods, such as adsorption separation using zeolites and activated carbons, at near-ambient temperature and pressure during the past decades.\(^3\) Metal–organic frameworks (MOFs) have attracted much attention for the separation of hydrocarbons because of their high pore volumes and designable pore properties.\(^1b,4\) For ethane/ethylene separation, most of the reported MOFs were C$_2$H$_4$ selective, where the separation mechanism typically involves the selective interaction of C$_2$H$_4$ with open metal sites or highly polar groups in the framework.\(^5\) However, these C$_2$H$_4$-selective MOFs are not ideal because they demand an additional desorption procedure to afford the C$_2$H$_4$-rich product stream (≥99.95%) and the harsh desorption conditions owing to their strong interactions.\(^5c,6\) In contrast, C$_2$H$_6$-selective MOFs are more energy efficient because high-purity C$_2$H$_4$ can be directly obtained using only a simple adsorption process to adsorb a trace amount of C$_2$H$_6$ molecules. Despite these advantages, this type of adsorption behavior has been reported in a few MOFs and hydrogen-bonded organic frameworks so far, displaying relatively low C$_2$H$_6$ selectivity owing to the similar polarity of C$_2$H$_6$ and C$_2$H$_4$.\(^7\)

The interaction between C$_2$H$_6$ molecules and a framework material can be tuned in MOFs by controlling the pore surface chemistry based on topology and organic ligands. Because C$_2$H$_4$ has a larger quadrupole moment ($1.50 \times 10^{-26}$ esu cm$^2$) and a smaller polarization ($42.52 \times 10^{-25}$ cm$^3$) than those of C$_2$H$_6$ ($0.65 \times 10^{-26}$ esu cm$^2$ and $44.7 \times 10^{-25}$ cm$^3$, respectively),\(^7\) the pore surface chemistry of MOFs can be fine-tuned to enable the selective adsorption of C$_2$H$_6$ over C$_2$H$_4$. Separation of C$_2$H$_6$/C$_2$H$_4$ using cracking process, where C$_2$H$_6$ (ethane) is the primary feedstock for the reaction. The effluent of the steam cracking process contains 5–10% of unreacted C$_2$H$_6$, which must be separated to obtain high-purity C$_2$H$_4$ for the polymerization reaction. However, the similarity in molecular size and volatility of C$_2$H$_6$ (kinetic diameter 4.16 Å; boiling point 169.42 K) and C$_2$H$_4$ (kinetic diameter 4.44 Å; boiling point 184.55 K) makes the separation of the two molecules a challenge.\(^2\) To overcome the limitation of the technology, researchers have highly sought and intensively investigated effective separation methods, such as adsorption separation using zeolites and activated carbons, at near-ambient temperature and pressure during the past decades.\(^3\) Metal–organic frameworks (MOFs) have attracted much attention for the separation of hydrocarbons because of their high pore volumes and designable pore properties.\(^1b,4\) For ethane/ethylene separation, most of the reported MOFs were C$_2$H$_4$ selective, where the separation mechanism typically involves the selective interaction of C$_2$H$_4$ with open metal sites or highly polar groups in the framework.\(^5\) However, these C$_2$H$_4$-selective MOFs are not ideal because they demand an additional desorption procedure to afford the C$_2$H$_4$-rich product stream (≥99.95%) and the harsh desorption conditions owing to their strong interactions.\(^5c,6\) In contrast, C$_2$H$_6$-selective MOFs are more energy efficient because high-purity C$_2$H$_4$ can be directly obtained using only a simple adsorption process to adsorb a trace amount of C$_2$H$_6$ molecules. Despite these advantages, this type of adsorption behavior has been reported in a few MOFs and hydrogen-bonded organic frameworks so far, displaying relatively low C$_2$H$_6$ selectivity owing to the similar polarity of C$_2$H$_6$ and C$_2$H$_4$.\(^7\)

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MOFs with a suitable pore size is an effective strategy; however, discovering high-performing MOFs with specific pore sizes is a formidable challenge because of the large number of reported MOFs in the literature. High-throughput computational screening can supplement the experimental efforts to discover MOFs for C$_2$H$_6$/C$_2$H$_4$ separation. Keskin et al. computationally screened 278 MOFs using molecular simulation and compared the adsorption selectivities and working capacities of MOFs with those of zeolites. Jiang et al. applied computational screening to a large set (12020) of MOFs and found 16 top-performing MOFs for C$_2$H$_6$/C$_2$H$_4$ separation. On the basis of the extensive data obtained from the screening, they established the relationships between performance metrics such as working capacity, selectivity, and structural features such as pore size and surface area. However, there is a growing body of evidence indicating that the performance metrics commonly used in the literature, such as working capacity and selectivity, are inadequate to correctly predict the material’s performance in actual process settings, such as in pressure vacuum-swing adsorptions. Recent studies suggest purity and recovery as more meaningful indicators of adsorbent materials for separation. For instance, Rodrigues et al. employed purity and recovery to assess the C$_2$H$_6$/C$_2$H$_4$ separation performance. They evaluated the C$_2$H$_6$/C$_2$H$_4$ separation capabilities of Cu-BTC (BTC = benzene-1,3,5-tricarboxylate) and ZIF-8 (ZIF = zeolitic imidazolate framework) for pressure-swing adsorption and simulated moving bed using purity, recovery, and productivity. Although process-level evaluation is a more appropriate method for assessing material’s performance for adsorption-based separation, computational, or experimental evaluation of all the available MOFs at the process level is not practical due to the high cost and time involved with the task. Therefore, an alternative strategy is needed to discover MOFs with high ethane-selective MOFs.

In this work, we combine high-throughput computational screening with process simulation to computationally screen the Computation-Ready, Experimental (CoRE) MOF 2019 database for the selective adsorption of C$_2$H$_6$ over C$_2$H$_4$. Through computational screening, we found a top-performing MOF, UFATEA01[Ni(IN)$_2$], which we synthesized in the laboratory for testing. Ni(IN)$_2$ (HIN = isonicotinic acid) is a nickel isonicotinate-based ultra-microporous MOF with high stability under humid conditions. In addition, Ni(IN)$_2$ can be synthesized on a gram scale. The experimental data of Ni(IN)$_2$ and other adsorbent data from recently reported studies were compared according to the ideal vacuum swing adsorption (VSA) process modeling. The results of process-level simulations suggest that Ni(IN)$_2$ is the top-performing material used in computational screening and is superior to many of the adsorbent materials reported to date for C$_2$H$_6$/C$_2$H$_4$ separation.

**Results and Discussion**

As shown in Figure 1a, the high-performing MOFs (CEPUT [Co(IN)$_2$], UFATEA01[Ni(IN)$_2$], and CEPUT01[Co(IN)$_2$]) were selected according to the screening procedure consisting of three different filters. In the first filter, we excluded MOFs with a pore limiting diameter (PLD) smaller than 3.75 Å, which corresponds to the diameter of a methyl group of C$_2$H$_6$ from the TraPPE model. Because C$_2$H$_6$ molecules are difficult to access in the pores of MOFs with a PLD smaller than 3.75 Å, materials with smaller pore sizes are screened out. Consecutively, we excluded MOFs containing expensive metal atoms (Au, Ag, Dy, Eu, Ga, Gd, Hf, In, Ir, La, Mo, Nd, Pd, Pt, Pr, Pt, Rh, Ru, Se, Sm, Tb, Te, Tm, U, and Y). This exclusion was employed as the second part of the first filter. Grand canonical Monte Carlo (GCMC) simulations were performed for the remaining 6830 MOFs to evaluate their performances. These simulations computed the binary component (50:50) adsorption uptake for C$_2$H$_6$ and C$_2$H$_4$ in the 6830 MOFs. More detailed methods are discussed in Section S1 in the Supporting Information. Figure 1b presents the distribution of both C$_2$H$_6$ uptake and selectivity (C$_2$H$_6$ uptake/C$_2$H$_4$ uptake) at 1 bar obtained from the GCMC simulations. From the computational screening data, we selected MOFs with a selectivity larger than 3 and a C$_2$H$_6$ uptake larger than 2.5 mmol g$^{-1}$ because selectivity and uptake have been considered as critical metrics in many previous studies. This step reduced the number of target MOFs from 6830 to 10, resulting in a significant reduction in the computational cost for the subsequent process modeling. Detailed discussion on the selection strategy is discussed in Section S6 in the Supporting Information.

Recent discussions in the literature show that the widely popular adsorbent’s performance metrics, such as selectivity and working capacity, do not accurately represent the materials’ process-level performance. For the process-level performance, product (i.e., C$_2$H$_4$) purity and recovery are two main performance metrics. In this work, the ethylene recovery, which is directly related to the amount of ethylene produced from the process, was calculated based on the ideal VSA simulation for the selected ten MOFs to evaluate their process-level performance. Several modifications to the original method were made to enable the process-level performance evaluation for the C$_2$H$_6$/C$_2$H$_4$ separation case, and the details are discussed in Section S1 in the Supporting Information. Figure 1d shows the ideal VSA simulation results for the ten selected MOFs. The results show that the top three MOFs (CEPUT [Co(IN)$_2$], CEPUT01, UFATEA01 [Ni(IN)$_2$]) are the same MOFs but synthesized with different metals, suggesting that the material’s performance depends on the identity of the ligand that forms the favorable pore environment for selective C$_2$H$_6$ adsorption.

To validate the performance of Co(IN)$_2$ and Ni(IN)$_2$, we synthesized and tested the adsorption performance of the two MOFs. Co(IN)$_2$ and Ni(IN)$_2$ were prepared according to the procedure reported in the literature and characterized by powder X-ray diffraction (PXRD) patterns (Figures S3 and S4, Supporting Information). The PXRD patterns of the synthesized MOFs matched well with those of the simulated structures, confirming their high phase purity. Before the gas isotherm measurements, the MOFs were degassed at 160 °C for 12 h under a vacuum to remove the guest molecules from the pores. However, the peaks of Co(IN)$_2$, almost disappeared in the PXRD pattern, indicating a structural collapse during the degassing process (Figure S3, Supporting Information). On the other hand, Ni(IN)$_2$ retained its structure after not only the degassing process but also the various gas adsorption measurements (Figure S4, Supporting Information). Because structural stability is vital in industrial applications, Ni(IN)$_2$ was chosen as a target adsorbent among potential adsorbent candidates.
As shown in Figure 2a, b, Ni(IN)₂ has a 1D channel with a size of 5.06 × 4.37 Å (excluding the van der Waals radii) along the a-axis. The framework is composed of linking isolated Ni octahedra by IN⁻ ligands, affording a square lattice coordination network. The synthesized Ni(IN)₂ was characterized by several experimental methods (Figure 2 and Figures S2, S4, and S5–S7, Supporting Information). The PXRD pattern was well matched with the simulated one, confirming that the MOF was successfully prepared with high purity (Figure 2c). The nitrogen isotherm of Ni(IN)₂ was collected at 77 K (Figure 2d). The isotherm with a typical type I behavior indicates the dominance of microporosity. The Brunauer–Emmett–Teller surface area and the total pore volume of Ni(IN)₂ were calculated to be 520 m² g⁻¹ and 0.305 cm³ g⁻¹, respectively. Furthermore, from the result of the density functional theory analysis, the pore size distribution was in the range 5–12 Å and mainly peaked at 6 Å, which is consistent with the pore size estimated from the crystal structure (Figure 2b).

The accessible surface of the channels is surrounded by pyridine rings of the IN⁻ ligands, which is expected to be a favorable structure for the framework to have host–guest interactions with incoming gas molecules. Single-component C₂H₆ and C₂H₄ isotherms of Ni(IN)₂ were recorded at 298 K and different temperatures after being fully activated at 160 °C under vacuum for 12 h to remove the guest molecules from the pores. These isotherms exhibited a type I behavior with a steep adsorption trend from 0 to 0.1 bar in Figure 3a. This indicates that C₂H₆ and C₂H₄ have strong host-guest interactions with the backbone of Ni(IN)₂. As the adsorption temperature increased from 273 to 323 K, the pressure of the steep adsorption events also increased (Figures S8 and S9, Supporting Information). The C₂H₆ uptake capacity of Ni(IN)₂ at 1 bar (68.36 cm³ g⁻¹) is notably superior to that of the benchmark adsorbents MAF-49 (38.1 cm³ g⁻¹) (MAF = metal-azolate framework), Cu(Qc)₂ (41.4 cm³ g⁻¹), and ZIF-7 (41.1 cm³ g⁻¹).[7e,k]

Ideal adsorbed solution theory (IAST) calculations were performed using the IAST++ program to predict the C₂H₆/C₂H₄ selectivity[17]. The experimental adsorption isotherms of Ni(IN)₂ were fitted with the dual-site Langmuir–Freundlich model and the adsorption selectivity of the binary mixture C₂H₆/C₂H₄ at ratios of 1:1 v/v and 1:15 v/v at 298 K was calculated from the
Figure 2. a) A 3D porous framework with a Connolly surface representation of Ni(II)₂ with microporous 1D channels along the a-axis. b) Pore window based on the distances between the centroids of the pyridine rings in the channel from the crystal structure. c) PXRD patterns of the simulated and as-synthesized Ni(II)₂. d) N₂ adsorption isotherm of Ni(II)₂ at 77 K. The inset indicates the pore size distribution in the range of 5–12 Å.

Figure 3. a) C₂H₆ and C₂H₄ adsorption isotherms of Ni(II)₂ at 298 K. The inset graph is plotted on a log scale. b) Adsorption isotherms and selectivity of Ni(II)₂ predicted by the IAST model for a gas mixture of C₂H₆/C₂H₄ (1:1, v/v) at 298 K. c) Plots of isosteric heat of adsorption of C₂H₆ and C₂H₄ for Ni(II)₂. d) Cycling breakthrough curves of the C₂H₆/C₂H₄ mixture (1:15, v/v) in a fixed bed packed with Ni(II)₂ at 298 K and 1 bar.
C2H6/C2H4 mixture (1:15, v/v). After the first cycle, the adsorption performance remained almost unchanged over five continuous cycles, suggesting that it has excellent recyclability for separation (Figure 3d). Additional experimental and simulation data, such as N2 isotherms and C2H6/C2H4 isotherms with molecular dynamics (MD) simulation results, are available in Figures S13–S15 and Tables S2 and S3 in the Supporting Information. Simulated results for N2, C2H6, and C2H4 indicate type I isotherms and Tables S2 and S3 in the Supporting Information. The adsorption selectivities of the simulated structures calculated from the single-component isotherms (17.6 and 2.88 cm3 g−1, respectively). The red dashed lines indicate C–H···𝜋 interactions. d) Adsorption energy distribution of a single C2H6 or C2H4 molecule inside Ni(IN)2 obtained from force field-based calculations (solid lines). The dashed vertical lines are the binding energy of C2H6 and C2H4 in Ni(IN)2 obtained from DFT calculations.

The estimated selectivities of Ni(IN)2 for C2H6/C2H4 at 298 K and 1 bar were 2.44 and 2.45 at ratios of 1:1 and 1:15, respectively (Figure 3b and Figure S10, Supporting Information). Among the reported MOFs, the selectivity of Ni(IN)2 at 298 K and 1 bar at a ratio of 1:1 outperformed that of most C2H6-selectivity MOFs such as MUF-15 (1.96) (MUF = Massey university framework), MIL-53(Al)-FA (1.9) (MIL = Matériaux de l’Institut Lavoisier; FA = fumurate), Ni(TMDBC)(DABCO)0.5 (1.98) (H2TMDBC = 2,3,5,6-tetramethylylerythphalic acid; DABCO = 1,4-diazabicyclo[2.2.2]octane), and ZIF-7 (1.5).[5a,7g–i] The adsorption selectivities of the simulated structures calculated from binary component GCMC results were 3.65 and 2.91 for the pristine and relaxed structures, respectively (Table S4, Supporting Information). Using the Clausius–Clapeyron formula, we experimentally determined the isosteric heats of adsorption (−Qst) of Ni(IN)2 for C2H6 and C2H4 from adsorption isotherms measured at 273, 298, and 323 K (Figure 3c and Figure S11 and Table S1, Supporting Information). The obtained −Qst for C2H6 at zero coverage is 34.5 kJ mol−1, which is greater than that of 33.3 kJ mol−1 for C2H4. A similar tendency is observed for a higher coverage.

To explore the dynamic separation capability of the C2H6/C2H4 mixture, we performed a breakthrough experiment using a packed column containing ≈1 g of activated Ni(IN)2. As shown in Figure S12 in the Supporting Information, it is clear that Ni(IN)2 can effectively separate a mixture of C2H6/C2H4 (1:1, v/v). C2H4 was initially passed through the column at 7.74 min g−1 and it reached saturation, while C2H6 was first detected at 11.79 min g−1. In the case of a different composition of C2H6/C2H4 (1:15, v/v), Ni(IN)2 can also separate the mixed gas at 17.03 min g−1 for C2H6 and at 21.94 min g−1 for C2H4 (Figure S12, Supporting Information). The results showed that C2H4 is more efficiently adsorbed into the Ni(IN)2 bed. The saturation uptakes of C2H6 and C2H4 on the fixed bed corresponded to 23.9 and 3.16 cm3 g−1, respectively, which is consistent with the expected amount based on the single-component isotherms (17.6 and 2.88 cm3 g−1, respectively). The calculated selectivity from the breakthrough curves is ≈1.98, which is slightly less than that from IAST (2.44). Subsequently, we repeated the breakthrough tests five times for the C2H6/C2H4 mixture (1:15, v/v). After the first cycle, the adsorbent was regenerated completely within 2 h at 160 °C. The results of the recycling experiments showed that the breakthrough performance remained almost unchanged over five continuous cycles, suggesting that it has excellent recyclability for separation (Figure 3d). Additional experimental and simulation data, such as N2 isotherms and C2H6/C2H4 isotherms with molecular dynamics (MD) simulation results, are available in Figures S13–S15 and Tables S2 and S3 in the Supporting Information. Simulated results for N2, C2H6, and C2H4 indicate type I isotherms generated from the structure (Figures S14 and S15, Supporting Information). To reflect more realistic conditions, the relaxed Ni(IN)2 structure was obtained from the pristine Ni(IN)2 structure by MD simulation. The simulated isotherms from the relaxed Ni(IN)2 are more similar to the experimental data than from the pristine Ni(IN)2.

The result of the potential energy surface analysis shows the unique pore environment of Ni(IN)2, which leads to the selective adsorption of C2H6 over C2H4. Figure 4a shows the strong adsorption pockets within Ni(IN)2 created by the ligand along the pore wall and that the size of each pocket can only accommodate a single C2H4 molecule. Periodic density functional theory (DFT) calculations were carried out to investigate the selective adsorption mechanism of C2H6 over C2H4. Figure 4b,c shows the optimized configurations of C2H6 and C2H4 in Ni(IN)2 from DFT calculations. For visualization purposes, C2H6 and C2H4 molecules were represented by green and orange colors, respectively. The red dashed lines indicate C–H···𝜋 interactions. The lack of C–H···𝜋 interactions between the pore wall and C2H4 molecule results in the lower DFT binding energy of C2H4 (−49.9 kJ mol−1) than C2H6 (−43.9 kJ mol−1). Figure 4d shows the adsorption energy distribution of C2H6 and C2H4 in the pores of Ni(IN)2 obtained from Monte Carlo sampling of the pore with a single C2H6 molecule. The simulation data suggest that there is only one dominant adsorption energy peak for each molecule. This is consistent with the result of the potential energy surface analysis.
Figure 5. Results of the adsorbent evaluation (in terms of C$_2$H$_6$ recovery) based on the ideal VSA process simulations where 0.01 mbar desorption is used for the volumetric C$_2$H$_6$/C$_2$H$_4$ feed ratio of 1:15 at 298 K and 1 bar. a) Based on the molecular simulation results of the top adsorbent (Ni(IN)$_2$) obtained from Figure 1d screening of the CoRE MOF 2019 database (red), of the other top candidates in Figure 1d screening work (blue), and of the recently reported adsorbents (black). b) Based on the experimental isotherm data of Ni(IN)$_2$ (red) and of the reported adsorbents (black).

shown in Figure 4a, which shows a single strong adsorption site within the pores of Ni(IN)$_2$. The adsorption energy of C$_2$H$_6$ obtained from the simulation shows that the dominant adsorption energy peak for C$_2$H$_6$ is $-39.5$ kJ mol$^{-1}$, while that for C$_2$H$_4$ is $-35.3$ kJ mol$^{-1}$, providing quantitative evidence for the origin of the C$_2$H$_6$-selective nature of the material.

The ideal VSA process simulations were performed to evaluate the performance of Ni(IN)$_2$ in a practical setting. Figure 5a shows the results from the VSA process simulations based on the pure component adsorption isotherms obtained from the GCMC simulations. Besides the MOFs listed in Figure 1d (colored in blue both in Figure 1d and in Figure 5a), we also evaluated additional high-performing materials for ethane/ethylene separation reported in the literature for comparison with Ni(IN)$_2$. The results show Ni(IN)$_2$ is the best performing MOFs in terms of ethylene recovery. Recovery is a more realistic performance metric for the adsorbent process than productivity, a commonly used performance metric in recent literature. Additional results and discussions related to productivity and recovery metrics are provided in Section S8 of the Supporting Information. Furthermore, the experimental isotherms of the high-performing MOFs reported in the literature were also used to carry out the ideal VSA process simulations (Figure 5b; colored in black). We found that the Ni(IN)$_2$ is among the top-performing adsorbent materials for ethane/ethylene separation. Additional results and discussions related to different operating conditions are provided in Tables S4–S6 and Figures S16–S18 in Supporting Information.

**Conclusion**

In conclusion, we carried out high-throughput computational screening and process-level evaluation to discover C$_2$H$_6$-selective MOFs from the CoRE MOF 2019 database. We explored a large number of MOFs to search for the most effective adsorbent for C$_2$H$_6$/C$_2$H$_4$ separation and successfully found an adsorbent, Ni(IN)$_2$, that exhibits high performance in the separation process. Ni(IN)$_2$ with a well-matched pore environment can selectively adsorb C$_2$H$_6$ over C$_2$H$_4$ with a high experimental C$_2$H$_6$/C$_2$H$_4$ selectivity of 2.45. The results of the breakthrough experiments with a mixture of C$_2$H$_6$/C$_2$H$_4$ (1:1 and 1:15, v/v) revealed that C$_2$H$_6$ was selectively separated from the gas mixture. The separation performance was well maintained over five repeated cycles. Using the process simulation, we compared the process-level performance of Ni(IN)$_2$ with those of the recently reported MOFs. The result of this comparative evaluation shows that Ni(IN)$_2$ outperforms many of the adsorbents and ranks third place among the ten high-performing MOFs. On the basis of these results, this work suggests that the MOF has a strong potential as a solid adsorbent for the separation of olefin/paraffin in industrial environments.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

Research data are not shared.
