



Recent Updates in Using Metal-Organic Frameworks (MOFs) for Nitrogen Compounds Removal from Liquid Fuels



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Abstract

The increasing demand for ultra-clean fuels has driven extensive research into efficient methods for removing nitrogen-containing compounds (NCCs) from liquid fuels. This review focuses on enhancing the adsorption capacity and reusability of MOFs and their composites, especially those with functionalized sites, show promise in effectively removing nitrogenous compounds like indole and quinoline. Studies highlight the role of hydrogen bonding and acid-base interactions in the adsorption process, and strategies like functionalizing MOF linkers and metal sites are being explored to improve performance. MOF composites, like ZIF-67(x) @H₂N-MIL-125, have demonstrated remarkable adsorption capacities for indole removal from fuel. Modifying MOFs with functional groups, like -COOH, on both the linker and metal sites can significantly improve the removal of nitrogen-containing compounds, especially neutral ones. MOFs with open metal sites, like MIL-101(Cr), are also being investigated for their potential in adsorptive denitrogenation. The adsorption of nitrogenous substances depends critically on hydrogen bonding, especially when MOFs are functionalized with amino groups. Acid-base interactions between the MOF and nitrogen-containing compounds can also contribute to the adsorption process. MOFs can be reused through solvent washing, making them a sustainable option for fuel purification. The recyclability of MOF materials, like BITSH-1, has been demonstrated for up to four cycles in thiophene removal. In summary, recent advancements in MOF technology, particularly the use of MOF composites and functionalized materials, offer promising solutions for the effective and sustainable removal of N-compounds from liquid fuels. Finally, the review outlines current challenges, such as scalability and structural durability, and proposes future directions toward the design of robust, cost-effective, and sustainable MOF-based materials for industrial fuel purification.

Keywords: Metal-Organic Frameworks; Adsorptive Denitrogenation; Nitrogen Compounds; Liquid Fuels Purification; Functionalized Composites.

Abbreviations

MOFs refers to Metal-Organic Frameworks; TGA is thermogravimetric analysis; ICP-OES is inductively coupled plasma-optical emission spectroscopy; PXRD is X-ray diffraction; MMOFs means magnetic metal organic frameworks; PSM means Post synthetic modification; MMMs refers to mixed-matrix membranes; ADN is adsorptive denitrogenating; NCCs is nitrogen-containing compounds; IND means indole and QUI is quinoline.

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1. Introduction

Metal-organic frameworks (MOFs) have attracted great attention as next-generation adsorbents and catalysts for ADN due to their extraordinarily high surface area, tunable pore architecture, and adjustable chemical functionality. Their modular structure-composed of metal nodes and organic linkers-allows fine control over pore size, coordination environment, and surface chemistry, enabling selective interactions with target nitrogen species. In particular, functionalization with amino, carboxyl, or sulfonic groups enhances adsorption via hydrogen bonding and acid-base interactions, while open metal sites and π - π interactions further improve selectivity and capacity. Metal-organic frameworks (MOFs) are crystalline, porous hybrid materials made up of metal ions or metal-oxo units connected by organic ligands that donate electrons. MOFs are categorized by functional groups based on structural characteristics[1].

These porous frameworks are sturdy and stable. You can use these MOFs for molecular sieving[2]. When guest particles are inserted or removed, flexible frameworks exhibit the greatest change in shape. They are also impacted by outside variables like temperature and pressure[3]. When solvent molecules are removed, dynamic frameworks alter, but when gas molecules are adsorbed under high pressure, they maintain their porous structure[4]. The existence of an open metal site can improve MOF performance. The ability of open metal site MOFs to extract CO₂ is improved when water is present [5]. Grafting functional groups onto the surface of MOFs can improve their adsorption capacity[6]. Recently, the importance of MOFs in adsorption process is investigated by many authors [7-20]

One synthesis approach for creating single crystals of MOFs is the hydrothermal process, which involves combining metal salts with organic ligands in a water media[21]. High-performance MOFs and their composites with adjustable size and structure have been created using this hydrothermal synthesis's straightforward and secure benefits[21]. In the ionothermal and solvothermal synthesis techniques, separated metal ions and organic ligands work together to create crystalline MOFs, which facilitates the controlled production of MOFs similar to the hydrothermal technique[22]. This procedure is part of a special class of solvothermal techniques in which organic ligands react with isolated metal ions to produce or crystallize MOFs without the need for heating[22]. One of the primary benefits of microwave (MW)-assisted synthesis is its rapid heating time, which also saves energy and contributes to a higher yield and higher purity than conventional procedures[23].

The electrochemical approach, which offers a rapid synthesis and mild reaction conditions, has been used to create MOFs. Making MOFs using mechanochemical synthesis has a lot of promise to be beneficial for the environment and the economy[24]. When compared to the solvothermal method, sonochemical synthesis can significantly shorten the crystallization period while also accelerating homogenous nucleation and the production of smaller particle sizes[25]. In order to increase the range and adaptability of functional groups integrated into parent MOFs, postsynthetic modification (PSM) has gained widespread acceptance[26]. A technique for chemical synthesis called microfluidics uses chips to regulate fluid flows at the micro and nanoscale[27]. Limitations of metal-organic frameworks (MOFs) include poor stability in extreme conditions, potential toxicity and biocompatibility issues, high fabrication costs, challenges with regeneration and recycling[28], low electrical conductivity, and difficulties in achieving high selectivity and targeted absorption[29]. These drawbacks present significant hurdles to their widespread commercial application and require ongoing research to develop solutions. In contrast, the present review provides a comprehensive and critical synthesis of recent progress in MOF-based nitrogen compound removal, integrating adsorption performance data from multiple MOF families and correlating these results with structural and chemical modifications. Unlike prior reviews, it emphasizes structure-property relationships, adsorption mechanisms, recyclability, and design strategies for practical applications. Furthermore, this work compiles quantitative comparisons of adsorption capacities, regeneration cycles, and interaction mechanisms (hydrogen bonding, acid-base, π - π , and cation- π) to identify performance trends across various MOF systems. By bridging fundamental understanding and application-oriented analysis, this review aims to clarify the current challenges and guide future research toward the development of robust, scalable, and sustainable MOF-based materials for industrial fuel purification.

2. Applications of MOFs

MOF characterization employs techniques like X-ray diffraction (PXRD, SCXRD) for structure and crystallinity, adsorption of gas for pore characteristics, thermogravimetric analysis (TGA) for thermal stability and composition, spectroscopic methods (FTIR, UV-Vis DRS) for functional groups and optical properties, microscopy (SEM, TEM) for morphology, and elemental analysis (ICP-OES) for composition. These methods reveal structural, textural, elemental, optical, and thermal properties crucial for tailoring MOFs to specific applications like gas adsorption, catalysis, and sensing. Metal-Organic Frameworks (MOFs) have diverse applications because of their huge surface area, adjustable structures, and high porosity, including gas adsorption and storage (e.g., hydrogen, CO₂), catalysis for chemical synthesis and degradation, biomedicine for drug delivery and biosensing, energy conversion and storage in batteries and fuel cells, environmental remediation through pollutant removal, and advanced sensors for detecting various substances[30, 31]. The transportation, storage, and conversion of gaseous fuels present the biggest obstacles to their storage and separation. These typically demand demanding operating conditions and expensive energy. The creation of porous materials for adsorption-based gas storage systems in moderate climates presents a viable substitute. As pure or post-synthetically modified heterogeneous catalysts, MOFs are frequently used in catalysts. If open coordination sites are accessible, using their metal nodes is the simplest method. Biological and medical applications, MOFs are extensively used in biological and medical applications such as biosensing, drug release, biomimetic catalysis, etc. For these applications, MOFs with excellent chemical stability are a must. Because the aromatic units of linkers in the majority of MOFs produce excitation by absorbing UV-visible light and producing luminescence, a significant number of MOFs have been observed to be photoluminescent[32, 33]. Applications of magnetism with MOFs with magnetic properties are known as magnetic metal organic frameworks (MMOFs), and they exhibit magnetism when paramagnetic 3D transition metal nodes are utilized in conjunction with appropriate diamagnetic organic linkers.

3. MOF in fuel purification

Metal-Organic Frameworks (MOFs) are highly porous materials used in fuel purification by acting as selective adsorbents for removing impurities such as nitrogenous compounds (NCs) and sulfur compounds from fuels or for separating hydrocarbon mixtures. Their special qualities, such as their large surface area, adjustable functionality, and regulated pore diameters, enable the selective adsorption of particular molecules and provide an energy-efficient substitute for conventional separation techniques[34]. MOFs can be incorporated into mixed-matrix membranes (MMMs) or used as standalone adsorbents for applications like natural gas purification and deep denitrogenation. MOFs selectively adsorb impurities from fuel streams based on differences in affinity between the MOF surface and the impurities. MOFs can be integrated into membranes to create mixed-matrix membranes (MMMs) that leverage the MOF's selective adsorption and permeation properties for more efficient separations. By adding open metal sites or functional groups, MOFs can be chemically altered (functionalized) to improve their adsorption ability and selectivity for particular pollutants.

3.1. MIL derivatives

A metal-organic framework (MOF), MIL-100(Fe), was impregnated with a Lewis acidic salt, AlCl_3 , in order to prepare an acidic adsorbent. The adsorbent was used in adsorptive denitrogenation (ADN). Despite an 8% decrease in surface area when loading the MOF with AlCl_3 , the maximum adsorption capacity of $\text{AlCl}_3/\text{MIL-100(Fe)}$ for QUI was 17% more than that of the pristine MIL-100(Fe). On the other hands the amount of adsorbed IND decreased with increasing AlCl_3 loading, because this does not have any basicity[35]. Initially, Scandium-triflate (Sc(OTf)_3) was deposited onto porous materials including activated carbon and MOFs. When comparing $\text{Sc(OTf)}_3/\text{CuBTC}$ to virgin CuBTC, a notable increase in the adsorption capacity for SCCs was noted (in the case of BT, about 65% augmentation based on the weight of adsorbents; 90% based on the surface area of the adsorbents). Additionally, QUI was adsorbed more preferentially than virgin MIL-101 over $\text{Sc(OTf)}_3/\text{QUI}$. On the other hand, supported Sc(OTf)_3 had no positive impact on the adsorptive elimination of neutral NCCs like IND. The favorable acid-base interactions between the acidic Sc(OTf)_3 and basic adsorbates like SCCs and QUI may be the source of the enhanced adsorptive capabilities[36].

Using MIL-101(Cr), often known as M101, a MOF, particularly following functionalization with diethylenetriamine (DETA) or ethylenediamine (ED). The efficiency of the MOF in CBZ adsorption improved with the number of amino groups on the MOF, as seen by **Figure 1**, which also shows that the maximum adsorption capacity (Q_0) of M101-DETA for CBZ (per unit BET surface area) was 1.4 and 2.1 times that of M101-ED and M101, respectively. In order to effectively and selectively remove NCCs from fuels with a high concentration of SCCs, amine-functionalized MOFs like M101-DETA, which have a high maximum adsorption capacity of 277.8 mg of CBZ/g, can be recommended as potential/recyclable materials[37].

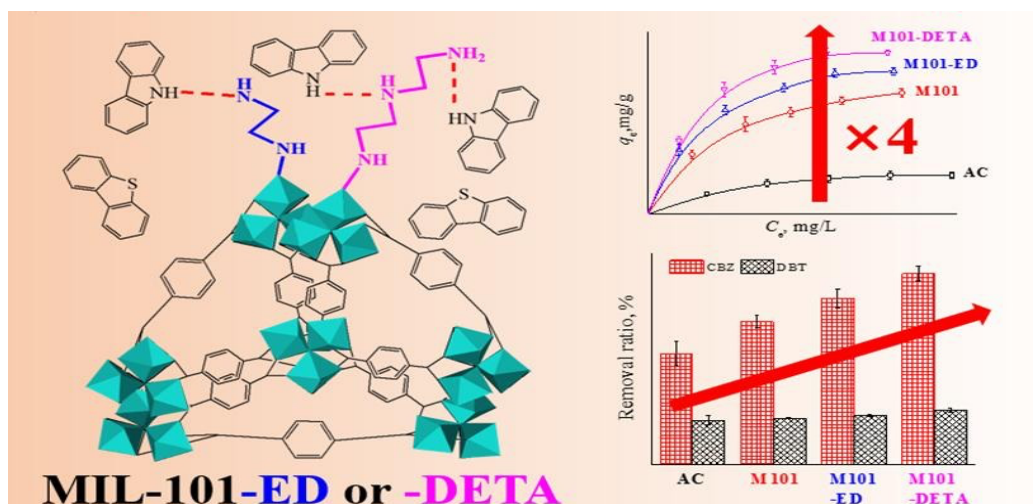


Figure 1: MIL 101-DETA has high adsorption than MIL-ED and MIL101[37].

Initially, PA-encapsulated MIL-101(Cr) adsorbents were made using the simple "incipient wetness impregnation" technique. In comparison to the pristine MIL-101(Cr), the modified MIL-101(Cr)s produced extremely encouraging maximum adsorption capabilities (Q_0) for both the neutral indole and basic quinoline. When PA was encapsulated onto MIL-101(Cr), the adsorption of IND and QUI increased by 86% and 91%, respectively, based on the adsorbent's unit surface area. **Figure 2** demonstrated that PA@MIL-101(Cr)s may be easily regenerated and utilized again in adsorptions for up to multiple cycles. The hydrogen bonding and the acid-base interactions could explain the remarkable adsorptions[38].

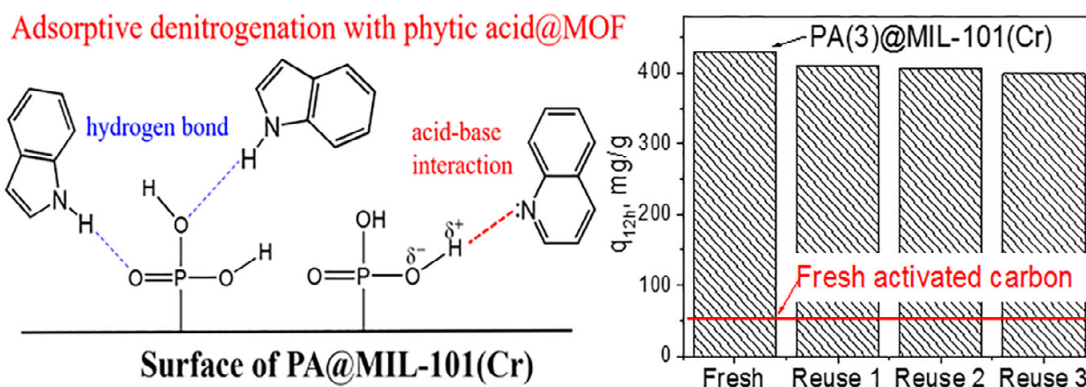


Figure 2: PA@MIL-101(Cr) denitrogenation by hydrogen bonding and the acid-base interactions[19].

Through hydrogenation, grafting, and direct synthesis, five MIL-101 MOFs were produced. When amino functional groups were added to MIL-101, IND's adsorption capacity rose dramatically (up to 1.7 times that of MIL-101). However, due to the lower porosity, the adsorption of QUI decreased when MIL-101 was changed with both amino and butyl groups. The relevance of H-bonds for the adsorption of IND over MIL-101s was demonstrated by the adsorption capacity for IND (based on the unit surface area of MIL-101s), which revealed that MIL-101s with amino groups had an adsorption capacity of approximately 2.3 times that of MIL-101 or MIL-101 with butyl groups[39].

MIL-101(Cr), a very stable MOF, was altered to add amino groups to the ligand and metal sites. The resulting NH₂-MIL-101 then interacted with oxalyl chloride to produce OC-ED-A-M101, a multipurpose adsorbent. According to **Figure 3**, the OC-ED-A-M101's adsorption capacity for quinoline and indole was 9.3 and 11.7 times that of an activated carbon, respectively. Furthermore, when compared to all known MOF-based adsorbents, the novel adsorbent demonstrated the greatest and second-highest adsorption capacities for quinoline and indole, respectively. H-bonding could be used to explain OC-ED-A-M101's notable performances[40].

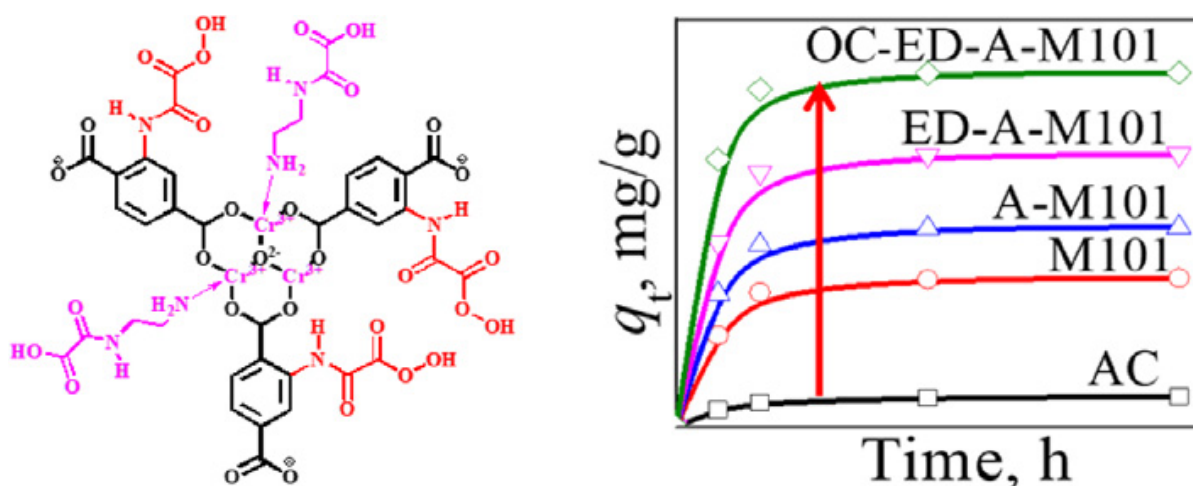


Figure 3: The OC-ED-A-M101 had 11.7 and 9.3 times adsorption capacity from activated carbon and it interpreted with H-bonding[40].

Graphene oxide (GnO), MIL-101 (Cr benzenedicarboxylate), and a highly porous metal-organic framework (MOF) were combined to create a composite GnO/MIL-101. Because of the composite's increased porosity, both IND and QUI demonstrated better adsorption on it as compared to pristine MIL-101 or GnO. The composite adsorbent performed ADN the best, regardless of the solvents under study or the co-presence of IND and QUI. The amount predicted for the porosity was far less than the improvement in IND adsorption. The added mechanism of hydrogen bonding explains this notable improvement[41].

In liquid phase adsorption of nitrogen-containing compounds (NCCs) from a model fuel, phosphotungstic acid (PWA) impregnated metal-organic framework (MOF) MIL-101 was employed as an adsorbent. In MIL-101, basic QUI's adsorption capacity rose by 20% with just 1% PWA impregnation. As seen in **Figure 4**, the minimal interaction between neutral NCCs and acidic adsorbents resulted in a minor reduction in the adsorption of a neutral chemical, IND[42].

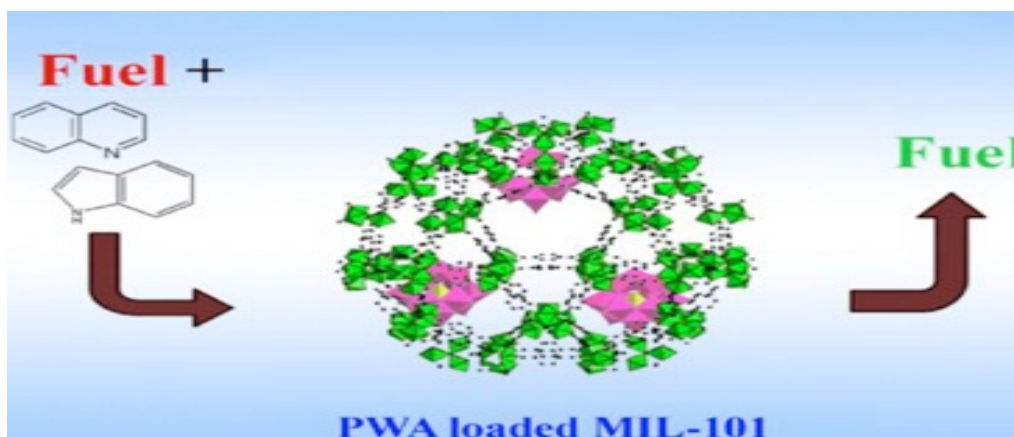


Figure 4: MIL-101 was impregnated with phosphotungstic acid (PWA) adsorption of nitrogen-containing compounds (NCCs) from a model fuel[42].

Using a ship-in-a-bottle approach, polyaniline-encapsulated MOFs (MIL101, Cr-benzenedicarboxylate) were made and used in liquid phase adsorptions. The protonated pANI@MIL101, or P-pANI-5, demonstrated record-high capabilities for the adsorptive removal of both basic and neutral NCCs (**Figure 5**). For instance, the protonated pANI@MIL101's maximal adsorption capabilities (Q_0) for the neutral indole and basic quinoline from n-octane were 602 and 556 mg/g, respectively. Additionally, it is possible to recycle the adsorbents[43].

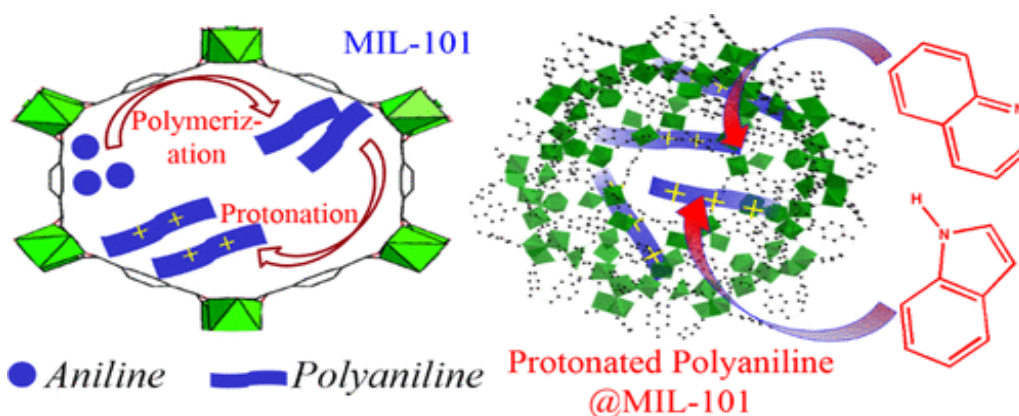


Figure 5: pANI@MIL101 used to removal of both basic and neutral NCCs[43].

MIL-101 was selected as the original MOF, and the method of adding moderate amounts of PMA heteropoly acids to the MOF was tried. The aforementioned tests demonstrate how the performance of the adsorbents can be significantly enhanced by further purification with NH_4F . The best adsorbent was P-M101-F 5%; the adsorption effect of P-MIL101-F x% on QUI increased as PMA increased, and P-M101-F 5% had the best adsorption effect of IND. The fact that the adsorption activity of M101-F/P-M101-Fx% is roughly five to six times that of M101/P-M101 x% is really intriguing[44].

It has been possible to successfully create graphite oxide/metal organic framework (MIL-101) composites. The amount of GO had a significant impact on the composites' surface area, and the MIL-101's surface area was significantly increased by a little amount of GO content (usually 0.25%). The potential uses of the composites in adsorptions were shown by the adsorptive removal of NCCs from model fuels; the adsorption capacity was reliant on the composites' surface area and pore volume. Out of all the adsorbents that have been described thus far, the composite possesses the highest adsorption capacity for NCCs[45].

A simple approach was used to create CuCl impregnated MIL-100(Cr), which was then used for the adsorptive denitrogenation of simulated fossil fuels. Nitrogen-containing compounds' (NCCs') adsorption rose. However, the lower porosity resulted in a decrease in the adsorption of NCCs over $\text{CuCl}_2/\text{MIL-100}(\text{Cr})$, which was prepared similarly but without Na_2SO_3 . Due to the p-complexation effect of the Cu^+ sites of CuCl, the maximum adsorption capacities of $\text{CuCl}/\text{MIL-100}(\text{Cr})$ for quinoline (QUI) and indole (IND) were enhanced by 9% and 15%, respectively, in comparison to pristine MIL100(Cr)[46].

By encasing the phosphomolybdic acid (PMo12) active core in a MIL-100(Fe)-based composite, effective catalytic oxidation was accomplished. Complete desulfurization of a multicomponent model fuel containing the most refractory sulfur and nitrogen compounds was achieved after only 30 min. After this little time, the nitrogen compounds might also be eliminated, mostly

using strategic solvents for extraction. As seen in **Figure 6**, the S/N elimination procedure proceeded more quickly when the costly ionic liquid [BMIM]PF₆ was swapped out for the less expensive ethanol[47].

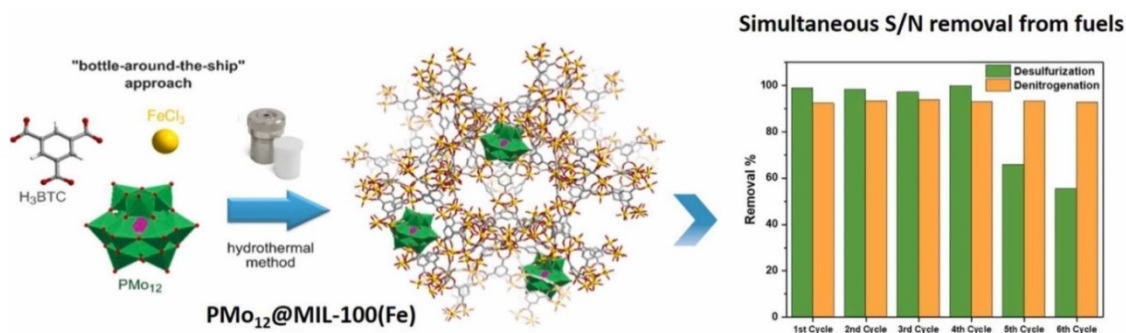


Figure 6: Synthesis of PMo₁₂MIL101(Fe) and shows removal of S/N from fuels[47].

Using modified CuCl and the microwave technique, the MIL-47 was created. For the first time, the use of Central Composite Design (CCD) in the optimization of the ADN process was assessed in this study. For indole (IND), the highest adsorption capacity (Q_0) of CuCl-MIL-47 was 769.2 mg/g. In this case, the π -complexation of IND with Cu produces a synergistic effect that raises Q_0 . Additionally, CuCl-MIL-47's regeneration via ethanol demonstrated its reusability[48].

A stable metal-organic framework (MOF, here, MIL-125) and its amino form (MIL-125-NH₂) were prepared, and the latter one was further modified to get MIL-125 with various functional groups or MIL-125-NHC(O)-C(O)-OH. MIL-125-VFG(VFG means various functional groups) showed the highest adsorption capacity for IND, compared with any adsorbent reported so far. The MOF also showed the second highest uptake of QUI, after the protonated polyaniline(5%)@MIL-101. The remarkable performances of MIL-125-VFG could be interpreted with ample active sites for H-bonding [49].

The adsorption characteristics of N/S-heteroaromatic compounds from model fuels were examined in relation to the functional groups in MIL-101(Cr)-X metal-organic frameworks (MOFs) with X = H, NH₂, and NO₂. When a nitro group is added, the adsorption capabilities of indole and quinoline from isooctane likewise diminish. Even though the porosity is 30% less than that of the parent, the adsorption capacities of quinoline and indole on amino-MIL-101(Cr) rise by roughly 25% and 10%, respectively. The NH₂ group's hydrogen bonding with organonitrogen compounds is responsible for amino-MIL-101(Cr)'s enhanced adsorption capabilities for quinoline and indole[50].

Batch adsorption studies were used to investigate the adsorptive denitrogenation from fuels over three metal-organic frameworks (MIL-96(Al), MIL-53(Al), and MIL-101(Cr)). Lewis's acid sites and a high specific surface area are present in MIL-101(Cr). Acid-base interaction is the primary basis for NCC adsorption. Additionally, pore size and shape are crucial for adsorption over MIL-53(Al) and MIL-96(Al). Furthermore, a straightforward technique can be used to regenerate MIL-101(Cr)[51].

In a metal-organic framework (Cr)-MIL-101-SO₃Ag functionalized with silver ions, an adsorbent with numerous interaction sites has been achieved. Its high adsorption capacity was believed to be caused by the immobilized Ag(I) sites' strong p-complexation interactions with NCCs, as seen in **Figure 7**. The adsorption capacities of QUI and IND in (Cr)-MIL-101-SO₃Ag were approximately 50% and 40% higher in pure n-octane solvent, respectively, than in (Cr)-MIL-101-SO₃H. Additionally, (Cr)-MIL-101-SO₃Ag exhibits excellent NCC selectivity to BT in model fuels[52].

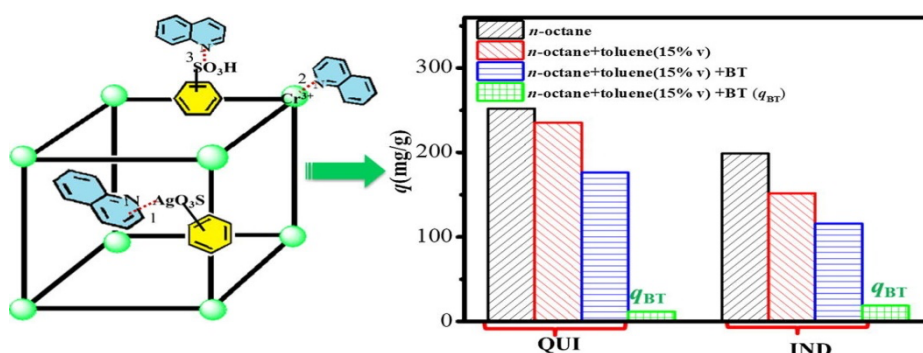


Figure 7: (Cr)-MIL-101-SO₃Ag has high adsorption of QUI and IND in pure n-octan[52].

Due to the locations on MIL-101(Cr) that enable the highest adsorption, MIL-101(Cr) exhibits significant ADN capacities at low concentrations. Because quinoline has a higher adsorption strength (−61.31 kJ/mol) than indole (−38.33 kJ/mol), **Figure 8** illustrates that the adsorption capacity of MIL-101(Cr) is larger for basic quinoline than for nonbasic indole. The sequence of organonitrogen >> organosulfur > naphthalene is followed by the adsorption selectivity of different types of chemicals in fuels, indicating that MIL-101(Cr) is a highly selective adsorbent for ADN[53].

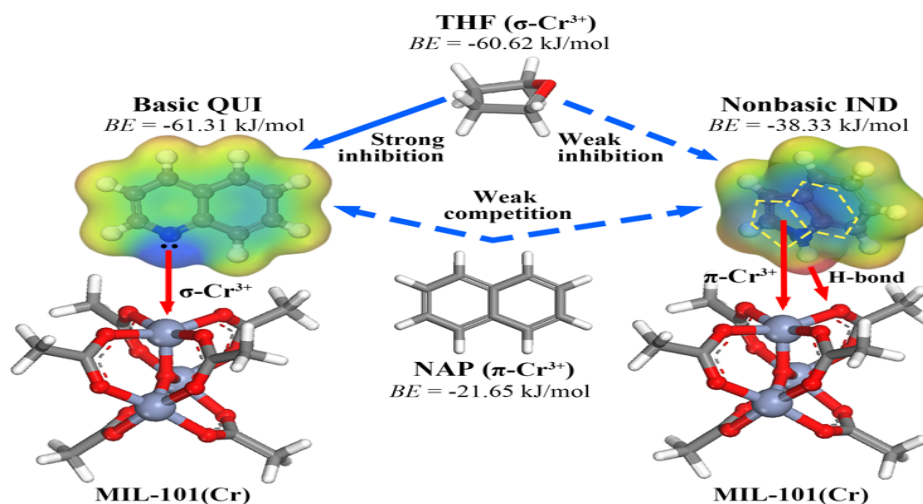


Figure 8: MIL-101(Cr) has high adsorption for QUI than IND[53].

MOF (MIL-100(Cr)) was altered to give the MOFs either basicity or acidity. The modification was done by grafting ethylenediamine and aminomethanesulfonic acid onto coordinatively unsaturated sites of the MOF, MIL-100(Cr). The adsorptive removal of a basic quinoline or benzothiophene can be improved noticeably, especially at low concentrations, with the introduction of an acidic site; however, with a basic adsorbate like quinoline, a basic MOF significantly reduces the adsorptive performance. Because of their reduced porosity, functionalized MOFs exhibit somewhat reduced adsorption for a neutral adsorbate like indole[54].

For the first time, the nucleobase adenine (Ade) was added to a highly porous metal-organic framework (MOF), MIL-101, by grafting onto it. P-Ade-MIL-101 was created by further protonating the Ade-grafted MOF, Ade-MIL-101. P-Ade-MIL-101's adsorption performance was competitive with that of other documented adsorbents; for IND and QUI adsorption, it has a capacity that is 12.0 and 10.8 times that of AC, respectively. Figure 9 demonstrated how Ade-MIL-101 exhibited exceptional IND and QUI adsorption. The method by which P-Ade-MIL-101 removes IND was suggested to be H-bonding in conjunction with cation- π interactions. Furthermore, P-Ade(1)-MIL-101 was proposed to adsorb QUI mostly through acid-base interactions[55].

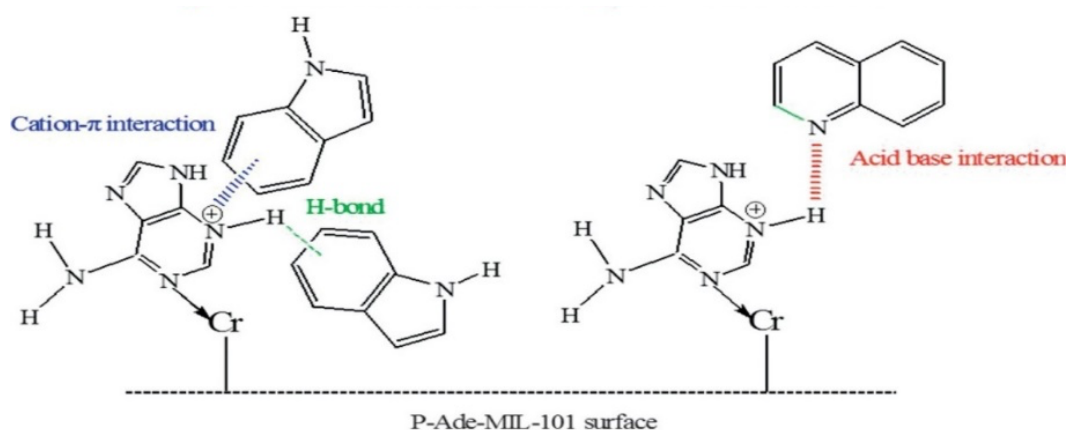


Figure 9: The remarkable adsorption mechanism of IND and QUI by Ade-MIL-101[55].

The composite ZIF-67(x)@H₂N-MIL-125 [Z67(x)@M125] is a metal-organic framework (MOF) was initially created and synthesized. The adsorption efficiency of the ONCs was further improved when Z67(5.0)@M125 was protonated. Importantly, the protonated Z67(5.0)@M125 showed the highest adsorption capacity, especially for IND, compared to any adsorbent that has been previously described based on MOFs or activated carbon. A few mechanisms, such as H-bonding, cation- π , acid-base, and π -complexation, as illustrated in Figure 10, may account for the extraordinary adsorption of the investigated ONCs[56].

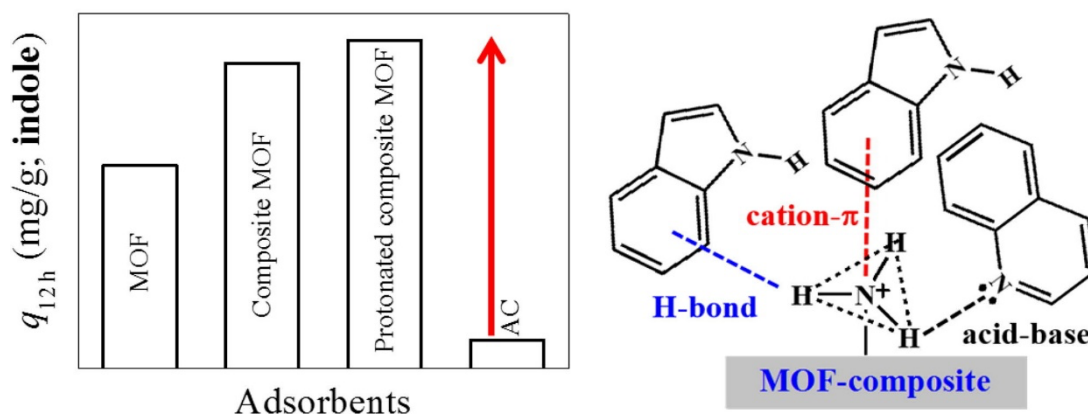


Figure 10: ZIF-67(x)@H₂N-MIL-125 has high adsorption capacity that explained by a few mechanisms including H-bonding, cation- π , acid-base and π -complexation[56].

MIL-100 was produced using various metal oxides, including CrO₃, Mn₂O₃, and V₂O₅. The quinoline (QUI) adsorption capacity was measured to examine these adsorbents' capacity to remove NCCs from fuels. The results showed that MIL-100 (Cr), MIL-100 (V), and MIL-100 (Mn) had maximum adsorption capacities of 49.67, 68.90, and 54.05 (mg N/g adsorbent) at 20°C, respectively. According to experimental results, MIL-100 (V) has the highest quinoline/dibenzothiophene selectivity (18), whereas MIL-100 (Mn) and MIL-100 (Cr) had selectivities of 13 and 12, respectively[57].

For the photocatalytic denitrification of pyridine in fuel, a series of TiO₂@MIL-101(Cr) were synthesized using the solvothermal synthesis approach, which is an efficient way to create composite materials. The results show that the photodenitrogenation performance of MIL-101(Cr) increase greatly owing to composite with TiO₂. 20%TiO₂@MIL-101(Cr) has the highest catalytic activity, and the denitrogenation ratio can reach 70% within 4 h under visible light [58].

Metal-organic framework performanceIn batch and fixed-bed experimental setups, the ability of MIL-101 (Cr) to remove organic nitrogen (N-) compounds through adsorption from straight run gas oil (SRGO) and straight run gas oil/light cycle oil (SRGO/LCO) mixtures was contrasted with that of four commercial adsorbents: silica gel, Selexsorb_ CD, Selexsorb_ CDX, and activated carbon. Batch results at equilibrium at room temperature demonstrated that MIL-101 (Cr) has the highest adsorption capacity by far. Furthermore, MIL-101 (Cr) demonstrated good durability, retaining its initial adsorption capability (from 90 to 85%) after 300 regeneration cycles. The aforementioned indicates that MIL-101 (Cr) has demonstrated promise as a material for the adsorption of N-compounds from middle distillates as a pretreatment step toward the manufacturing of ultra-low sulfur diesel [59].

Aromatic compounds with extremely low acidity or basicity were adsorbed over two highly porous adsorbents, a metal-organic framework (MOF, MIL-101) and activated carbon (AC). This study examined the effects of solvent polarity and the hydrophilicity/hydrophobicity of adsorbents on adsorption. To determine potential uses of the adsorbents in adsorptive desulfurization (ADS), adsorptive denitrogenation (ADN), and water purification, respectively, thiophene, pyrrole, and nitrobenzene were studied in liquid phase adsorptions. The three adsorbates were more efficiently adsorbed by MIL101 when the solvent polarity decreased and by AC when the solvent polarity increased. The hydrophilia of MIL101 and the hydrophobicity of AC account for this phenomenon. Furthermore, it can be said that MOFs, particularly hydrophilic ones, can be employed successfully in non-aqueous phase adsorptions, such as AND and ADS[60].

3.2. UiO derivatives

Compared to the pristine (UiO-66), the UiO-66-NH-SO₃H MOF, which has two functional groups, performed significantly better in removing indole (IND), acidic (UiO-66-SO₃H), and basic (UiO-66-NH₂) MOFs. According to MOF surface area and unit weight, UiO-66-NH-SO₃H's maximum adsorption capabilities for IND were 2.22 and 1.63 times greater than those of UiO-66, respectively. When compared to the other UiO-66 MOFs under investigation, the enhanced performance of UiO-66-NH-SO₃H was ascribed to the material's greater availability of H-bond acceptor sites. Although the adsorbed levels of basic NCCs (such quinoline and QUI) were smaller in UiO-66-NH-SO₃H[61]

Using a continuous tube reactor, aminobenzoate-defected UiO-66-NH₂ adsorbents were successfully produced in 20 minutes of holding time. The UiO-66-NH₂ structure was bound by ABA modulators, which resulted in ligand-missing defects. The QUN and MPR adsorption of the ABA-defected UiO-66-NH₂ was improved. Their NCC removal effectiveness was further dramatically enhanced by protonating the faulty UiO-66-NH₃⁺ framework. The ABA modified UiO-66-NH₃⁺ exhibited the highest absorption capabilities in the batch adsorption mode, with around 275 mg/g for the QUN and 204 mg/g for the MPR. About 28 L and 49 L of liquid fuel containing 1 mg L⁻¹ of quinoline and 2-methyl pyrrole can be treated by 1.0 g of the generated AUio-66-NH₂ pellets under continuous fixed-bed conditions. **Figure 11** illustrates the considerable potential of UiO-66(Zr)-NH₂ for the adsorptive[62].

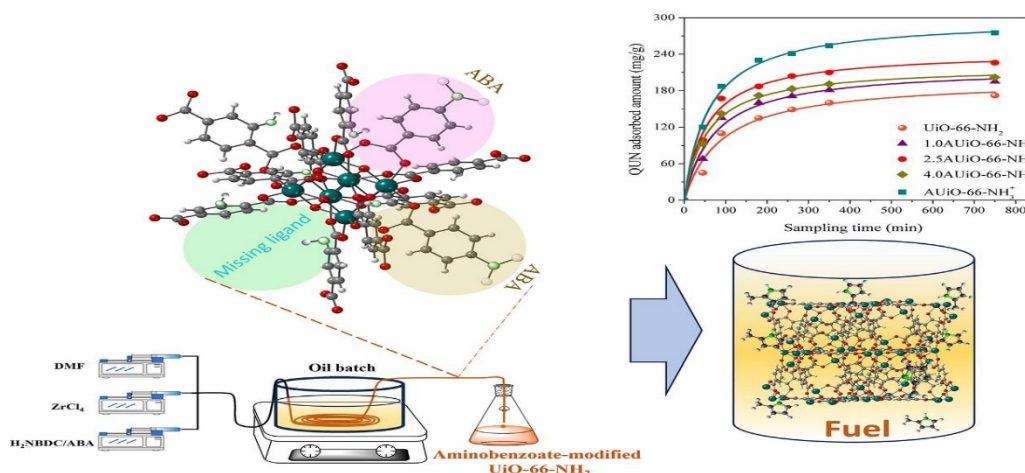


Figure 11: Synthesis of UiO-66-NH₂[62].

To provide a quantitative understanding of the adsorption mechanism, IND and QUI have been adsorbed over eight UiO-66s (seven functionalized UiO-66s and one pure UiO-66). On UiO-66s, the adsorbed amount of IND and QUI rose linearly as the number of H-acceptors and H donors (for the H-bond) increased, respectively. In the IND and QUI adsorption, UiO-66-NH³⁺ and UiO-66-NH₂ had a deviating trend, which might be attributed to base-base repulsion and cation- π interactions, respectively. Of the MOFs under study, UiO-66-NH₃⁺ exhibited the maximum adsorption for both IND and QUI[63].

Using a solvothermal method, MnO₂ NPs were grown on UiO-66 to create a MnO₂/UiO-66 composite. MnO₂/UiO-66 oxidized 2000 ppm of DBT (347 ppm Sulfur) and pyridine (502.8 ppm Nitrogen) in 3 minutes at O/S and O/N of 4, 0.06 g/15 mL catalyst dosage, and 25 °C, according to the catalytic data shown in **Figure 12**. Up to the sixth cycle, a much greater removal of DBT and pyridine (100%) was accomplished under the improved reaction parameters than those of BT, 4,6-DMDBT, indole, and carbazole. An extremely successful, incredibly quick, and economical substitute for the extensive oxidative desulfurization and denitrogenation of fuel oils is MnO₂/UiO-66-NaClO [64].

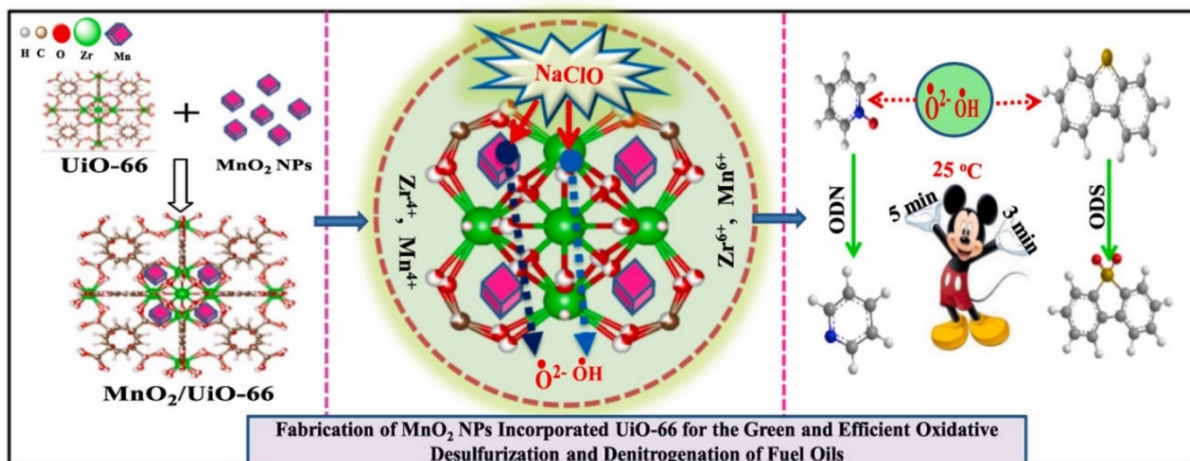


Figure 12: Fabrication of MnO₂NPs incorporated UiO-66 for the green and efficient oxidative desulfurization and denitrogenation of fuel[64].

The CuCl-X/CNT/UiO-66 nanocatalyst was synthesized in two steps and its effectiveness in catalytic oxidative denitrogenation processes was assessed. **Figure 13** displays the produced composites performed exceptionally well in the ODN process because they exhibited better catalytic qualities than UiO-66 and CNT/UiO-66. CuCl₃/CNT/UiO-66 demonstrated 100% catalytic performance for IND, 97.6% for QUI, and 82.5% for CBZ during a 120-minute period under ideal conditions. With its potential for useful applications in fuel treatment procedures, the CuCl-3/CNT/UiO-66 catalyst offers a promising solution for fuel purification and the effective removal of nitrogen-containing chemicals[65].

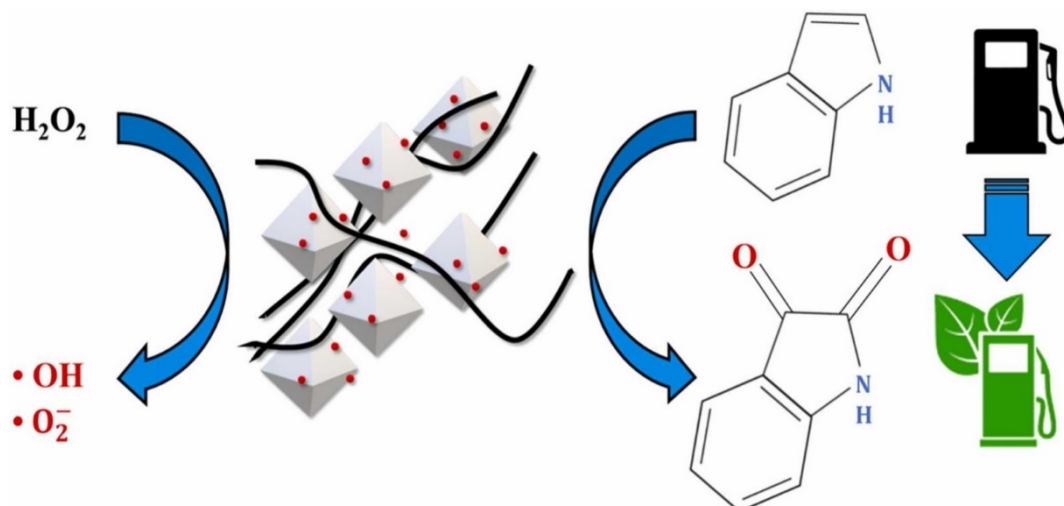


Figure 13: The CuCl-X/CNT/Uio-66 performance in catalytic oxidative denitrogenation[65].

In order to eliminate nitrogen compounds (NCs), a family of UiO-66 MOFs compounds with zirconium and hafnium metal centers was created and employed as adsorptive and oxidative catalytic chemicals. It took several hours before effective adsorptive denitrogenation was discovered, increasing from 19% at 5 h to 79% after 24 h, using UiO-66(Hf). In fact, the UiO-66(Hf) MOF demonstrated high viability for industrial application, creating a denitrogenation system that balances resilience, recyclability, sustainable functionality, and high efficiency, as seen in **Figure 14**[66].

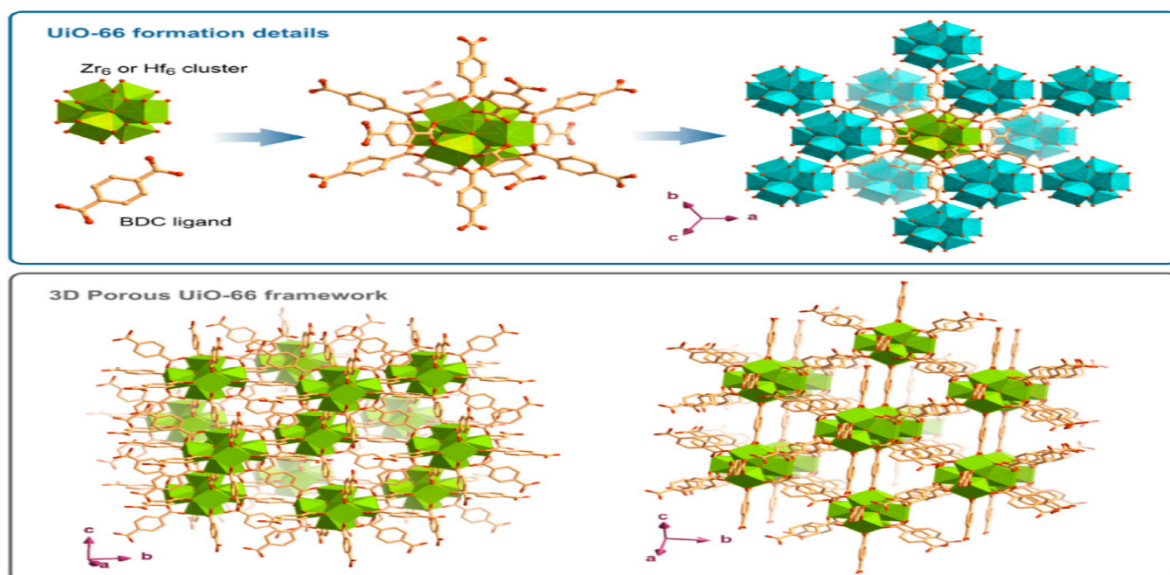


Figure 14: The UiO-66-type metal–organic frameworks (MOF) compounds' structural details: Each metal cluster (light green) coordinates with 12 other clusters (light blue) to form the MOF from the self-assembly of metal clusters and BCD ligands, resulting in an incredibly robust framework (**top**); crystal packing arrangement viewed in two different directions, revealing the porous features of this family of MOFs (**bottom**)[66].

A MOF was made using the functional group COOH (UiO-66-COOH). Although the MOF's porosity was reduced, the presence of this functional group, -COOH, significantly enhanced the adsorption capacity for IND and PYR. Basic NCCs like QUI and PY were also successfully adsorbed by UiO-66-COOH. A PY, PYR, and MPYR adsorption mechanism. Acid-base interactions and H-bonding, respectively[67].

Terephthalate and aminoterephthalate linkers were present in the produced UiO-66 and UiO-66-NH₂, respectively. In spite of the reduced porosity of UiO-66-NH₂, the adsorption capacity of IND was improved up to 46% when compared with pristine UiO-66. The MOF gains greater adsorption capacity due to the additional amino group present. The improved adsorption could be attributed to H-bonding because of the NH₂ group [68].

For the first time, the MOF UiO-66-SO₃H was created and used in the adsorptive removal of NCCs from model fuel. Based on weight and surface area, the adsorption of neutral IND was enhanced by up to 20% and 37% over UiO-66-SO₃H (18) in comparison to the pure MOF. Hydrogen bonding may be the cause of improved adsorption. Complex outcomes in the adsorptive removal of basic QUI were demonstrated by UiO-66-SO₃H. Because of steric hindrance and repulsive interactions, UiO-66-SO₃H demonstrated a greater adsorption capability for QUI (when present at low concentrations)[69].

Due to a favorable hydrogen bonding interaction between Py and the amino group of the MOFs, the adsorption kinetics and the amount of adsorbed Py (both in the vapor and liquid phases) increased as the amino group concentration in UiO-66s rose. This advantageous connection (hydrogen bond) prevented the predicted repulsion between basic Py and basic MOFs with amino groups. As seen in **Figure 15**, the amount of adsorbed Py rose as the temperature rose up to 80 °C, most likely due to the similar sizes of Py and UiO-66's pore window and a slight increase in UiO-66's pore size with temperature[70].

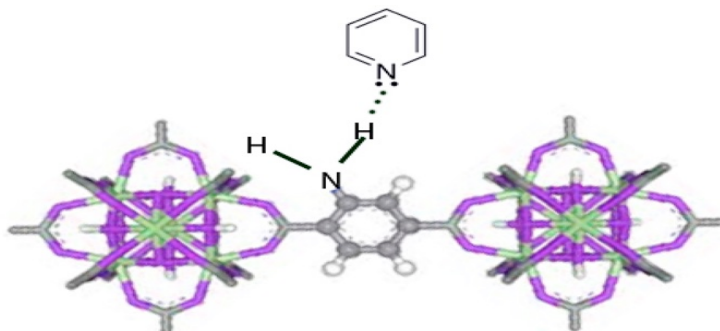


Figure 15: Adsorption of pyridine on amino group of the MOFs in presence of Hydrogen bond[70].

3.3. ZIFs derivatives

An imidazolium-based polyoxometalate (POM) was successfully encapsulated into a ZIF-8 framework to create a heterogeneous catalyst ([BMIM]PMo₁₂@ZIF-8). In order to simultaneously proceed with the oxidative desulfurization and denitrogenation of a multicomponent model fuel containing diverse sulfur and nitrogen molecules, this composite material demonstrated remarkable catalytic performance. All of these chemicals were completely eliminated in just one hour, and the catalyst system could be utilized again for ten cycles in a row without losing its effectiveness, as shown in **Figure 16** [71].

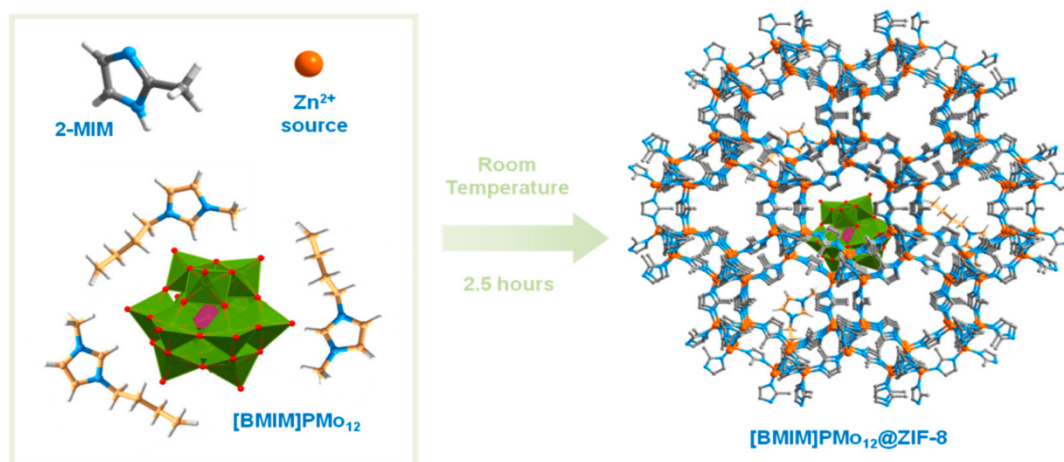


Figure 16: The [BMIM]PMo₁₂@ZIF-8 chemical was prepared using a "bottle around-the-ship" method and a sustainable room temperature synthetic strategy (one-pot and in situ)[71].

3.4. Other types of MOFs

Metal organic framework (MOF) and polysulfone composite films were made. **Figure 17** illustrates how the resulting films were used to remove nitrogenated chemicals from model fuel, specifically indole and quinoline. With 52.6 and 205.3 mg/g MOF, respectively, the maximum adsorption capacities for polysulfone@MOF composite films were much higher than those for polysulfone film, which ranged from 188.0 to 220.1 mg/g[72].

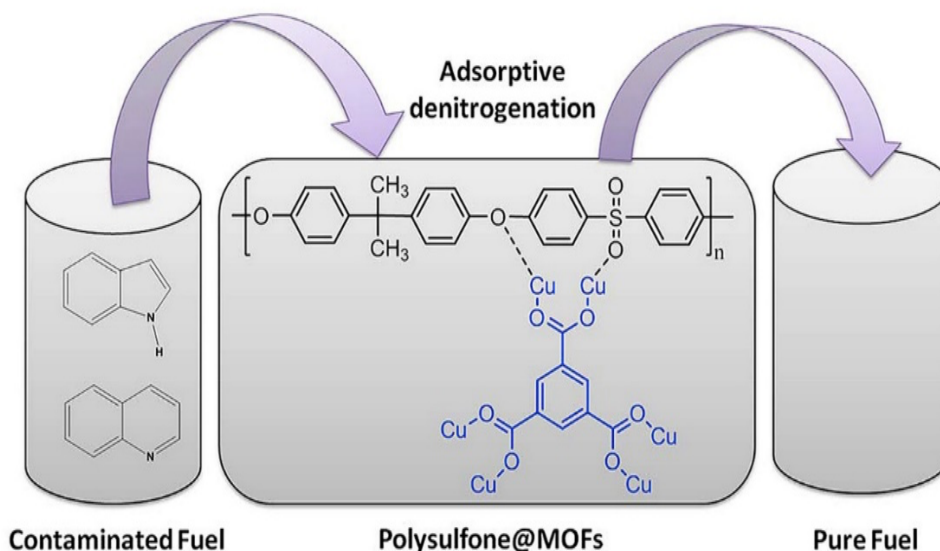


Figure 17: Polysulfone@MOF used in adsorptive denitrogenation from fuel[72].

CuCl_2/ACs was reduced with Na_2SO_3 in ambient circumstances to produce CuCl -loaded activated carbons (ACs). Even though the porosity of AC decreased when CuCl was loaded, the adsorptive capacities of CuCl/AC for QUI and IND were around two or more times those of pure AC. The p-complexation between Cu^+ and NCCs may account for the effective ADN with CuCl/AC . Furthermore, because AC is widely accessible and the preparation may be carried out under ambient circumstances, the CuCl/AC preparation process is an economical and low-energy approach[73].

Fe_3O_4 nanoparticles were encapsulated in an amino-functionalized MOF (TMU-17- NH_2) under ultrasonic irradiation to create a magnetic responsive composite. The composite materials can be reused multiple times and are magnetically detachable. IND and QUI had maximal adsorption capacities of 375.93 and 310.18 mg/g over the produced composite at 25 °C, respectively. IND has a greater adsorption capability than QUI over $\text{Fe}_3\text{O}_4@\text{TMU-17-NH}_2$. Figure 18 illustrated the hydrogen bonding interaction between the amino group in the sorbent and the H atom that is connected to the N atom in the IND structure[74].

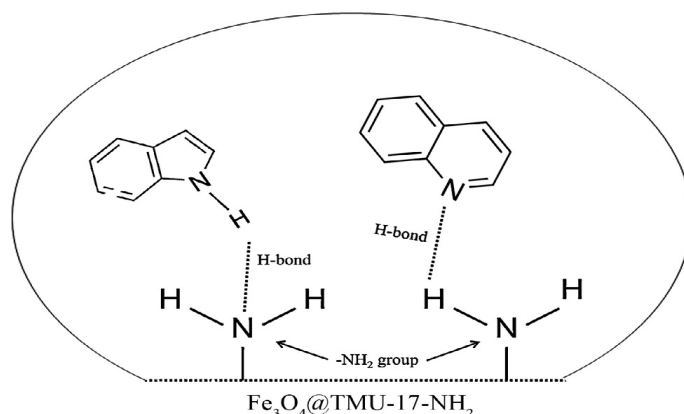


Figure 18: Hydrogen bonding between H atom in $\text{Fe}_3\text{O}_4@\text{TMU-17-NH}_2$, that connected to the amino group in the sorbent and the N atom in the IND structure[74].

Highly porous $\text{Cu}_2\text{O}/\text{MDC-K}$ adsorbents are prepared. The $\text{Cu}_2\text{O}/\text{MDCs}$ showed very efficient adsorptions, by around 213% and 243%, respectively, in contrast to plain MDC that isn't made with KOH. When it came to removing these adsorbates, the novel adsorbent outperformed traditional activated carbon by a factor of five to six. When it came to the adsorbates, the $\text{Cu}_2\text{O}(10)/\text{MDC-K}$ demonstrated superior selectivity over virgin MDCs[75].

$\text{Al}(\text{OH})(1,4\text{-NDC})@-\text{AlOOH}$ is a multiporous (mesoporous) composite that is formed via an in situ green synthesis method. With a strong selectivity for aromatics, Figure 19 demonstrated an active heterogeneous adsorbent for heterocyclic SCCs and NCCs. Two grams of this substance were used to extract 97.4% ($\text{Ci} = 77$ ppmwN) and 58.3% ($\text{Ci} = 350$ ppmwS) of NCCs and

SCCs, respectively, from twenty grams of actual FCC oil in two hours at room temperature. its distinct pore size, structural interactions, and unsaturated Lewis acid metal (Al) sites[76].



Figure 19: Al(OH)(1,4-NDC)/AlOOH is an effective heterogeneous adsorbent for NCCs and heterocyclic SCCs[76].

As seen in **Figure 20**, tungsten nitride-incorporated carbon ($W_2N(x)@C$) was initially created by pyrolyzing a recently created phosphotungstic acid-loaded metal-azolate framework-6, PTA(x)@MAF-6s. Under ultrasound (US) irradiation, the $W_2N(x)@C$ materials—particularly $W_2N(15)@C$ —were discovered to be an extremely effective catalyst for the oxidative elimination of a variety of organo-nitrogen compounds (ONCs), including nitrile, neutral, and basic. Crucially, the $W_2N(15)@C$ had a turnover frequency that was between 80 and 147 times higher than that of catalysts for persistent carbazole oxidation that have been described. Additionally, the catalyst's recyclability in the US irradiative ODN was verified[77].

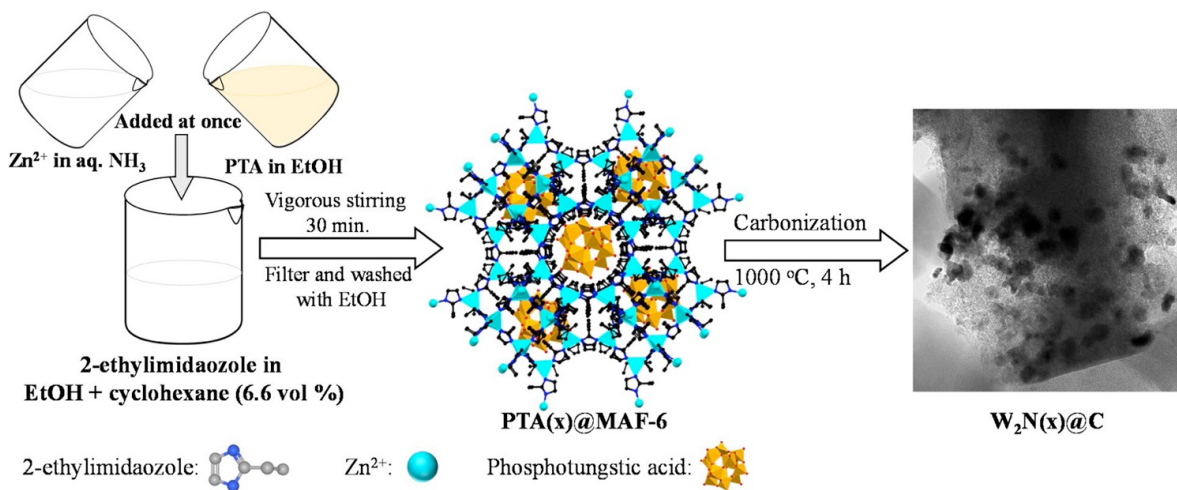


Figure 20: Diagrammatic representation of PTA(x)@MAF-6 and $W_2N(x)@C$ synthesis[77].

The resulting MOF808_EDTA was initially used to remove organics, such as NCCs, from liquids by adsorptive means; it performed remarkably well in this regard. For instance, MOF808_EDTA demonstrated the highest adsorption capacity (Q_0) of 667 and 625 mg/g, respectively, in the removal of quinoline (QUIN) and indole (IND) from fuel in **Figure 21**. When compared to published results, these Q_0 values for QUIN and IND adsorption rank highest and third, respectively[78].

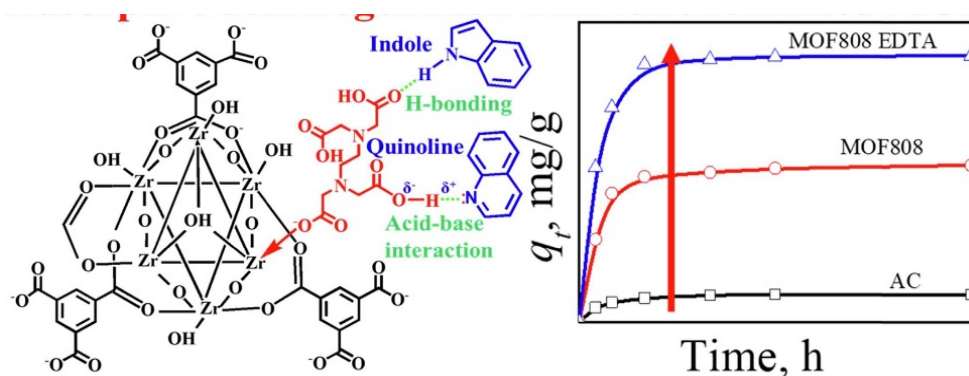


Figure 21: Adsorption of NCCs by MOF808_EDTA using H-bonding and acid-base interaction[78].

There are several different types of metal-organic frameworks (MOF-808s), including pristine MOF-808s, faulty MOF-808s, aminated MOF-808s with defects, and a defective MOF-808 with ammonium sites. First off, some MOF-808s with both a defect and a $-NH_2$ group shown exceptional performance in AND due to the beneficial synergistic contribution of both faulty and amino sites. Second, the experimental and computational results confirm that the H-bonding interactions depicted in **Figure 22** may account for the advantageous adsorption of QUI and IND over M808-y(NH_2/D)s; the acid-base interactions were responsible for the notable QUI adsorption over P-M808-(NH_2/D) [79].

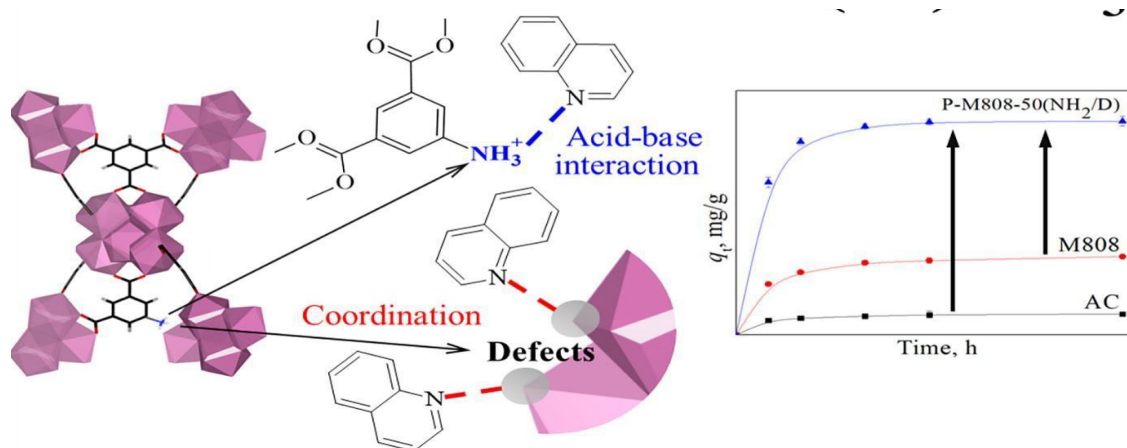


Figure 22: In AND, a few MOF-808s with both defects and the $-NH_2$ group performed exceptionally well[79].

To efficiently extract NCCs from fuel, metal-organic framework (MOF)-derived carbon was oxidatively modified to provide an excellent adsorbent. When compared to activated carbon, OMDC had an adsorption capacity that was four and twenty-six times greater for the removal of CARB and BENZ, respectively. Furthermore, as seen in **Figure 23**, OMDC had the best adsorption capacity for the removal of CARB and BENZ when compared to all other adsorbents that have been reported to date[80].

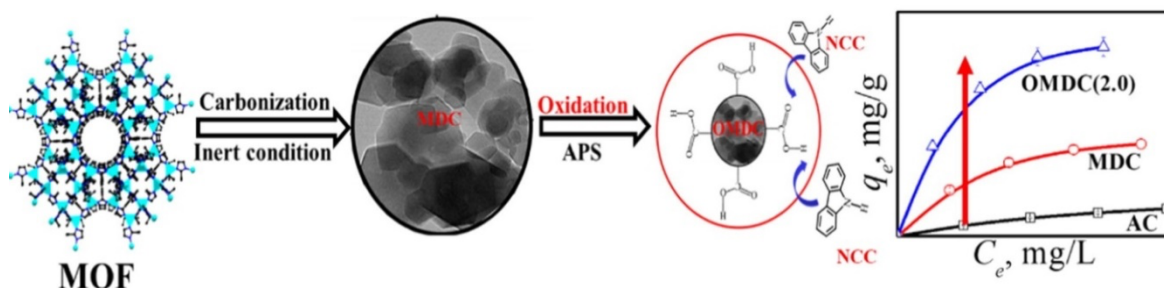


Figure 23: The prepared oxidized MOF-derived carbon (OMDC) and that high adsorption for CARB and BENZ[80].

A Ti-based metal-organic framework (MOF)/melamine combination was carbonized to create it. The produced catalysts performed remarkably well in fuel oxidative denitrogenation (ODN). One created catalyst, TiN@CN-2, for instance, has a low activation energy ($31.0 \text{ kJ}\cdot\text{mol}^{-1}$) and a high turnover frequency (58.1 h^{-1}). A nonradical process involving active Ti-peroxo species may be responsible for the outstanding performance of TiN@CN-2 in ODN, according to theoretical calculations and experimental investigations. As illustrated in **Figure 24**, titanium nitrides are more effective than titanium oxides at activating H_2O_2 and forming Ti-peroxo species[81].

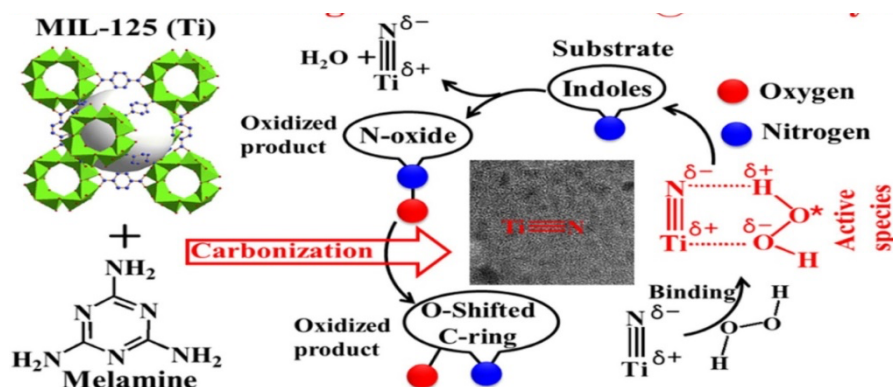


Figure 24: Prepared via carbonization of a Ti- based metal–organic framework (MOF)/melamine mixture [81].

As seen in **Figure 25**, highly porous carbonaceous materials were created by pyrolyzing AIPCP, either with or without loaded IL, and then thoroughly characterized using a variety of methods. Likewise, the effectiveness of CDIL@AIPCP for the model fuel's adsorptive desulfurization and denitrogenation was also apparent. CDIL@AIPCP's exceptional adsorption effectiveness for both aqueous and non-aqueous phases was mostly attributed to H-bonding[82].

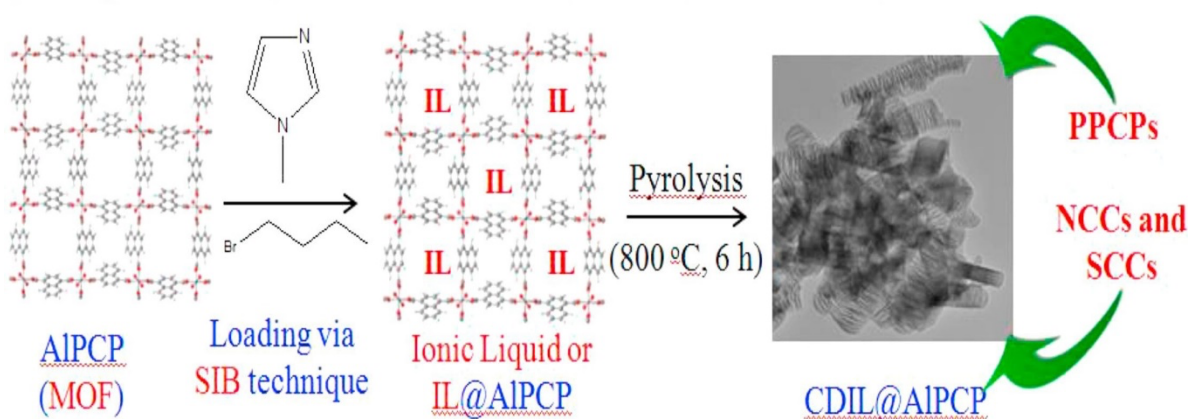


Figure 25: Preparation of CDIL@AIPCP that used in purification of water and fuel[82].

3.5. Quantitative and Comparative Analysis of MOFs Performance in Denitrogenation

A critical evaluation of reported adsorption data reveals that the adsorption capacity of MOF-based adsorbents is highly dependent on the metal node type, pore geometry, and surface functionalization. The quantitative results summarized in Table 1 allow meaningful comparison among representative MOF families and modified derivatives tested for nitrogen-containing compounds (NCCs) such as indole, quinoline, and carbazole.

Table 1: Comparative summary of reported MOF-based adsorbents for nitrogen compound removal from liquid fuels

MOF Type / Composite	Target Nitrogen Compound(s)	Adsorption Capacity (mg g ⁻¹)	Experimental Conditions	Regeneration / Reuse	Reference
MIL-101(Cr)	Indole	99.6	Model fuel (n-octane, 300 ppm N), 298 K	5 cycles, >90% capacity retained	[36]
MIL-100(Fe)	Quinoline	83.4	Model fuel (n-hexane, 250 ppm N), 303 K	4 cycles, 85% retained	[83]
UiO-66(Zr)	Quinoline	62.3	n-Octane, 298 K, 200 ppm	Moderate stability, reduced after 3 cycles	[63]
UiO-66-NH ₂	Indole / Quinoline	115.2 / 97.8	Model fuel, 298 K	6 cycles, >95% retention	[84]
ZIF-8	Carbazole	74.5	n-Octane, 303 K	Slight capacity loss after 5 cycles	[85]
ZIF-67	Indole	68.9	Isopentane, 298 K	Stable for 4 cycles	[56]
NH ₂ -MIL-101(Al)	Quinoline	120.3	n-Octane, 298 K	5 cycles, minor loss	[86]
MIL-53(Fe)/Graphene Oxide Composite	Indole	138.7	Model fuel, 303 K	Excellent recyclability (6 cycles)	[87]
Cu-MOF (HKUST-1)	Quinoline	65.2	Model diesel (200 ppm N), 298 K	Partial decomposition after 3 cycles	[31]
MOF-808(Zr)	Indole	127.4	Model fuel, 298 K	Maintained 90% after 5 cycles	[79]

4. Conclusion

Fuel purification is necessary because of cleaner fuel leads to better combustion, increased engine power, and improved efficiency. Purification is crucial for meeting emission standards and reducing environmental pollution. Fuel purification means removes contaminants like organics based nitrogen, sulfur and oxygen to improve engine performance and longevity by providing cleaner fuel.

Adsorption using special adsorbents can remove the contaminants from liquid fuel. Recently, metal-organic frameworks (MOFs) act as highly porous materials to adsorb (capture) molecules from a liquid or gas onto their surface. This is a highly efficient and versatile method for separating or removing substances, such as pollutants, gases, with other impurities, because of their huge pore volume, broad surface area, and tunable chemistry. MOFs achieve this through various mechanisms like electrostatic forces, hydrogen bonding, and π - π interactions.

MOFs are used to remove nitrogen (N) compounds from fuels through adsorptive denitrogenation (ADN). For ADN, MOFs are functionalized with groups like carboxylic acid (-COOH) or amine (-NH₂) to enhance their adsorption capabilities through mechanisms like hydrogen bonding and acid-base interactions. The N-compounds are then physically adsorbed onto the MOF, and the MOF can be regenerated by washing with a solvent. Combining MOFs with other materials, like polymers or other nanoparticles, can also lead to improved performance.

Challenges for using MOFs in fuel purification include poor stability and issues with scalability. Many MOFs struggle with chemical and thermal stability, particularly in the presence of water, acid, or high temperatures. This is a major limitation for practical use in fuel purification. The traditional solvothermal synthesis methods are often time-consuming, use harsh organic solvents, and can be difficult to scale up for mass production. Future development focuses on improving these aspects through techniques like defect engineering, hybridization with other materials, and developing greener synthesis methods. These developments aim to create more robust and sustainable MOFs for practical applications in fuel purification. For composite materials, such as MOF-based membranes, controlling the interface between the MOF and another material (like a polymer) is crucial but challenging. Understanding the complex relationship between a MOF's structure and its guest adsorption and diffusion properties can be difficult and often requires more advanced characterization techniques. Developing cost-effective, large-scale production methods is an ongoing challenge. While reusable, the process of releasing the captured molecule and preparing the MOF for another cycle needs to be efficient.

5. Future Perspectives

While MOF-based materials have made substantial advances in adsorptive denitrogenation of liquid fuels, there are several promising directions as well as persistent challenges that future research should target to move the field toward practical, industrial-scale application.

1. Enhancing Chemical and Hydrolytic Stability

Many MOFs (especially those with weaker metal-ligand bonds) suffer from degradation in the presence of moisture, acidic or basic species, or under thermal stress. For instance, frameworks based on zirconium or aluminum nodes have shown more resilience under hydrolytic conditions, but even these can lose performance after repeated adsorption-desorption cycles. Future work should prioritize MOFs with inherently robust bonding, or use strategies like protective coatings, hydrophobic linkers, or post-synthetic modifications to improve stability under realistic fuel conditions.

2. Real-Fuel Testing and Complex Mixtures

Most studies to date use model nitrogen compounds (e.g., quinoline, indole) in simplified liquid matrices. However, real fuels contain complex mixtures—sulfur compounds, aromatics, moisture, and trace metals—that can interfere with adsorption and regeneration. Systematic studies using actual fuel samples are needed to understand competing adsorption, poisoning effects, and to evaluate long-term performance.

3. Scalable Synthesis, Shaping, and Engineering

Laboratory-scale MOFs often are powders. For industrial applications, shaping into pellets, monoliths, membranes, or composite beads is necessary, but these processes may reduce surface area, block pores, or damage framework integrity. Moreover, cost and environmental footprint of the synthetic routes (solvents, linkers, and metal precursors) must be optimized. Research should explore greener synthesis (e.g., solvent-free, mechanochemical, room-temperature methods) and scalable fabrication protocols.

4. Regeneration, Durability, and Lifecycle Studies

Reusability is often demonstrated only over a few cycles in lab conditions. To assess commercial viability, studies should extend to many more cycles, simulate harsher regeneration (e.g. thermal, pressure, or chemical desorption), and analyze degradation pathways (structural collapse, loss of functional groups, fouling). Life cycle assessments (LCAs) could help compare MOF-based ADN to conventional HDN and adsorbent alternatives in terms of cost, energy use, and environmental impact.

5. Modular Design and Functional Tuning Based on Mechanistic Insights

Deepening understanding of how specific MOF features (pore size and shape, linker chemistry, open metal sites) influence each mechanism of adsorption (H-bonding, π - π , acid-base, cation- π) will help design more selective and efficient materials. Computational modeling and high-throughput screening can help narrow down promising combinations. Also, defect engineering (controlled introduction or healing of defects) may improve performance if carefully controlled.

6. Standardization of Testing Methods and Performance Metrics

A current hurdle is that different studies often use different conditions (temperature, pressure, solvent, fuel matrix), which makes direct comparison difficult. Establishing standardized protocols for adsorption testing, regeneration, durability, and reporting metrics will enhance reproducibility and allow benchmarking. Also, a shared data repository could help researchers track and compare new materials more transparently.

7. Integration with Other Separation or Treatment Technologies

Rather than relying on adsorption alone, integrating MOFs into hybrid systems—for example combining with membranes, catalytic oxidation, or pretreatment steps—may offer synergistic benefits. MOF-polymer composites or MOF supported on porous substrates could improve mass transfer, mechanical stability, or ease of installation in flow systems.

By focusing research along these lines, the field of MOF-based adsorptive denitrogenation can move beyond promising laboratory results toward real-world fuel purification solutions that are stable, cost-effective, and reliable.

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