A Process for Carbon Dioxide Capture Using Schiff Bases Containing a Trimethoprim Unit

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Abstract: Environmental problems associated with the growing levels of carbon dioxide in the atmosphere due to the burning of fossil fuels to satisfy the high demand for energy are a pressing concern. Therefore, the design of new materials for carbon dioxide storage has received increasing research attention. In this work, we report the synthesis of three new Schiff bases containing a trimethoprim unit and the investigation of their application as adsorbents for carbon dioxide capture. The reaction of trimethoprim and aromatic aldehydes in acid medium gave the corresponding Schiff bases in 83%–87% yields. The Schiff bases exhibited surface areas ranging from 4.15 to 20.33 m²/g, pore volumes of 0.0036–0.0086 cm³/g, and average pore diameters of 6.64–1.4 nm. An excellent carbon dioxide uptake (27–46 wt%) was achieved at high temperature and pressure (313 K and 40 bar, respectively) using the Schiff bases. The 3-hydroxyphenyl-substituted Schiff base, which exhibited a meta-arrangement, provided the highest carbon dioxide uptake (46 wt%) due to its higher surface area, pore volume, and pore diameter compared with the other two derivatives with a para-arrangement.

Keywords: Schiff bases; carbon storage media; synthesis; trimethoprim unit; surface area; adsorption; meta-substituted arrangement

1. Introduction

The consumption of fuel has increased significantly over the years because of the continuous growth of energy-demanding human activities. The burning of fuels leads to high levels of carbon dioxide (CO₂) in the atmosphere [1], with the contribution of fossil fuels to the emission of CO₂ representing approximately 60% [2]. The main contributors to the increased CO₂ emission into the atmosphere are pharmaceutical and agrochemical plants. Global warming and climate change are associated with high levels of CO₂ emission [3,4]. In particular, these environmental changes have led to an increase in the level of seas and oceans, which can cause flooding, acidic rains, and the collapse of the global economy [5,6]. Unfortunately, the rapid reduction of the level of CO₂ in the atmosphere is hindered by the large-scale consumption of fuels, which is difficult to reduce in the short run. Therefore, to overcome the environmental problems associated with global warming, research has been devoted to the development of strategies for reducing the level of CO₂, such as the design of new materials for efficient CO₂ uptake [7–10].

Various processes have been established for the capture and storage of CO₂, resulting in the reduction of its concentration in the atmosphere [11–16]. The capture technique
requires the separation of CO₂ from other gases at a high pressure followed by its adsorption [17]. The design of new materials for CO₂ capture and storage has attracted increasing research attention [18–20]. The most common commercial absorbents for CO₂ are bases such as potassium carbonate, aqueous ammonia, and ethanolamine [21–23]. The process using ammonia, in which ammonium carbonate is produced, is simple. However, the use of ammonia or ethanolamine is not convenient because of their volatility, high energy demand, and high operational cost [24]. Therefore, alternative absorbents with specific characteristics such as nonvolatility, recyclability, reusability, high adsorption capacity, and long life are still needed [25–27]. In addition, the application of such materials should be cost- and energy-effective.

Metal-containing adsorbents (e.g., metal oxides) have ionic and basic properties and can thus be used to capture CO₂. The exothermic process of CO₂ adsorption using metal oxides leads to the formation of metal carbonates [28,29]. However, metal oxides exhibit limited CO₂ selectivity and adsorption capacity. Other materials with high chemical and thermal stability and large surface area and pore volume have been used for CO₂ capture, including zeolites [30] and activated carbons [31], among others [32]. However, the use of these materials for CO₂ capture is accompanied by various disadvantages related to their poor selectivity or hydrophilic properties, which render them unsuitable to be used with flue gases. The CO₂ adsorption capacity of carbon-containing materials can be enhanced using a base. For example, the combination of polyacrylonitrile and potassium hydroxide was proved to be a good medium for CO₂ capture under mild conditions (25 °C and 1 bar) [33]. In addition, the CO₂ uptake capacity of a resorcinol–formaldehyde matrix was high when potassium carbonate was used [34]. In this context, metal–organic frameworks (MOFs) can act as CO₂ adsorbents because of their high surface area, low adsorption heat [35], and formation of strong interactions with CO₂ through hydrogen bonding [36]. Similarly, porous organic polymers (POPs) are suitable for CO₂ capture because they are low-density, stable materials with tunable structures. The skeleton of POPs can accommodate various polar groups, organic units, and heteroatoms that can increase their adsorption capacity toward CO₂ [37] by facilitating its coordination. However, the use of metals in the production of POPs causes environmental problems that require attention [38]. Therefore, the development of new materials for CO₂ adsorption is still an active area of research. Several other useful strategies have been used to reduce the level of CO₂. For example, the CO₂ mitigation through the direct photoreduction by sunlight and the catalytic hydrogenation to hydrocarbons can be efficient [39]. Some progress has been made using such an approach.

In the quest for new adsorbents for CO₂ capture, we turned our attention to heterocyclic compounds containing a 2,4-diaminopyrimidine moiety, because they are useful as biologically active compounds [40]. In particular, trimethoprim, which is used as an antibiotic to treat various infections [41], contains two different aromatic rings and is characterized by high nitrogen (19.3%) and oxygen (16.5%) contents. Considering that these characteristics could be beneficial for CO₂ adsorption, we attempted the synthesis of new trimethoprim Schiff bases and investigated their application in CO₂ capture. Recently, a range of materials has been designed, synthesized, and used to capture CO₂ [42–49].

2. Materials and Methods

2.1. Materials

Chemicals were obtained from Merck (Schnelldorf, Germany).

2.2. Instrumentations

Fourier transform infrared (FTIR) spectra were recorded on an FTIR 8300 Shimadzu spectrophotometer (Tokyo, Japan) using KBr pellets. The carbon, hydrogen, and nitrogen contents of the Schiff bases were determined using a Vario EL III elemental analyzer (Elementar Americas Inc., Ronkonkoma, NY, USA). Scanning electron microscopy (SEM; 25 kV) images were captured using a KYKY-EM3200 microscope (Ontario, CA, USA).
Nitrogen (N\textsubscript{2}) adsorption–desorption isotherms (77 K) were performed on a Quantachrome analyzer (Quantachrome Instruments, Boynton Beach, FL, USA). Before the measurements, the samples were dried (120 °C) under a flow of dry N\textsubscript{2} (2 h) using a vacuum oven (Cascade TEK, Cornelius, OR, USA). The Brunauer–Emmett–Teller (BET) equation was used to calculate the specific surface area (relative pressure (\(P/P_0\)) = 0.98). The pore size distribution was determined using the Barrett–Joyner–Halenda (BJH) method.

2.3. Adsorption Experiment

The CO\textsubscript{2} uptake (313 K and 40 bar) experiments were performed on an H-sorb 2600 high-pressure volumetric adsorption analyzer (Gold APP Instrument Corporation, Beijing, China) equipped with two degassing and analyzing ports that worked simultaneously. Optimization of the pressure condition required repeating the CO\textsubscript{2} uptake experiment more than 10 times. To the measuring tube containing a sample of the Schiff base, a known quantity of CO\textsubscript{2} was injected until equilibrium between the adsorbed gas and the Schiff base was reached. The final equilibrium pressure was recorded automatically using. The adsorbed quantity of CO\textsubscript{2} was calculated from the generated data.

2.4. Synthesis of Schiff Bases 1–3

A mixture of trimethoprim (2.9 g, 10 mmol) and 3-hydroxybenzaldehyde, 4-anisaldehyde, or 4-(dimethylamino)benzaldehyde (10 mmol) in boiling methanol (MeOH; 25 mL) containing glacial acetic acid (AcOH; 0.5 mL) was refluxed while being continuously stirred for 6 h. The solid obtained on cooling to room temperature was collected through filtration, washed with MeOH (10 mL), and dried to give the corresponding Schiff base 1, 2, or 3 (their FTIR spectrum in Supplementary Materials Figures S1–S3).

3. Results and Discussion

3.1. Synthesis of Schiff Bases 1–3

The condensation of equimolar equivalents of trimethoprim and an aromatic aldehyde (3-hydroxybenzaldehyde, 4-anisaldehyde, or 4-(dimethylamino)benzaldehyde) in boiling MeOH in an acidic medium for 6 h gave the corresponding Schiff bases 1–3 (Scheme 1) in 83%–87% yields (Table 1). The reaction accommodates different substituents (hydroxy, methoxy, and dimethylamino) at different positions (meta and para) of the aryl ring.

![Scheme 1. Synthesis of Schiff bases 1–3.](image)

**Table 1.** Characteristic physical properties of Schiff bases 1–3.

| Schiff Base | Color         | Ar               | Yield (%) | Melting Point (°C) | Calculated (Found; %) |
|------------|---------------|------------------|-----------|--------------------|-----------------------|
|            |               | 3-HOC\textsubscript{6}H\textsubscript{4} | 85        | 179–181            | 63.95 (64.09) 5.62 (5.69) 14.20 (14.28) |
| 1          | Yellow        | 4-MeOC\textsubscript{6}H\textsubscript{4} | 83        | 188–190            | 64.69 (64.72) 5.92 (5.95) 13.72 (13.77) |
| 2          | White         | 4-MeN\textsubscript{2}C\textsubscript{6}H\textsubscript{4} | 87        | 76–79              | 65.54 (65.55) 6.46 (6.48) 16.62 (16.68) |

The FTIR spectra of 1–3 showed characteristic absorption bands in the 1655–1657 cm\textsuperscript{-1} region due to the vibrations of the CH=\textsubscript{N} bonds (Table 2). In addition, absorption bands corresponding to the vibrations of the NH\textsubscript{2}, CH, C=\textsubscript{N}, and C=C groups in the aromatic systems were observed in the regions of 3446–3447, 3109–3127, 1589–1591, and 1506–1543 cm\textsuperscript{-1}, re-
spectively. Moreover, the FTIR spectrum of 1 (Supplementary Materials Figure S1) showed a broad absorption band at 3317 cm\(^{-1}\) attributable to the vibration of the OH group.

**Table 2.** Fourier transform infrared (\(\nu\), cm\(^{-1}\)) absorption bands of Schiff bases 1–3.

| Schiff Base | C=C | C=\(\text{N (Ar)}\) | C=\(\text{N}\) | CH (Ar) | OH | NH\(_2\) |
|------------|-----|----------------|-------------|--------|----|--------|
| 1          | 1543| 1589          | 1657        | 3109   | 3317| 3447   |
| 2          | 1506| 1591          | 1655        | 3121   |    | 3446   |
| 3          | 1506| 1589          | 1657        | 3127   |    | 3447   |

3.2. **SEM of 1–3**

SEM was used to analyze the surface morphology of 1–3. This technique can detect the presence of impurities and provides information about the homogeneity of the sample and the size and shape of the particles [50]. The SEM images (10 \(\mu\)m) of 1–3 (Figures 1–3) indicated the presence of a relatively uniform and amorphous surface with particles of different shapes and sizes. The surface morphology of the Schiff bases was similar in terms of pore size and shape. The pore dimension was found to be in the range of 33.6–41.7 nm for 1, 29.5–39.0 nm for 2, and 24.9–53.7 nm for 3, which are comparable with those of telmisartan tin complexes (20–50 nm) [43] and fusidate metal complexes (29–50 nm) [45] and smaller than those of carvedilol metal complexes (51–394 nm) [42], melamine Schiff bases (20–392 nm) [46], polysilicates (35–208 nm) [47], and polyphosphates (28–981 nm) [48,49].

![Figure 1. The SEM for the surface of Schiff base 1.](image-url)
Figure 2. The SEM for the surface of Schiff base 2.

Figure 3. The SEM for the surface of Schiff base 3.
3.3. \( \text{N}_2 \) Isotherms, Surface Area and Pore Size Distribution of 1–3

The quantity of gas uptake, measured using a gravimetric tool, controls the shape of corresponding physisorption isotherms [51] and provides information about the strength of the interactions between the gas and the adsorbent. The \( \text{N}_2 \) isotherms for both adsorption and desorption processes using Schiff bases 1, 2, and 3 are presented in Figures 4–6, respectively. Type III isotherms were observed in all cases, indicating the absence of monolayers and the presence of relatively weak interactions between the gas and adsorbents. The isotherms started at the origin, suggesting that the heat of adsorption was similar to the heat of condensation. The adsorption of gas on the surface of the Schiff bases was favorable, leading to a sharp increase in the adsorption as the pressure increased [52].

![Figure 4. \( \text{N}_2 \) adsorption (ADS) and desorption (DES) isotherms of Schiff base 1.](image)

![Figure 5. \( \text{N}_2 \) adsorption (ADS) and desorption (DES) isotherms of Schiff base 2.](image)
The porosity of 1–3, which provides information about their interactions with the adsorbed gas, was determined at 77 K from the N$_2$ adsorption–desorption isotherms using the BJH method [53], and the specific surface area was calculated using the BET method [54]. The pore size distributions of 1–3 are illustrated in Figure 7, and the porosity values are summarized in Table 3. As can be extracted from these results, the Schiff bases had mesoporous structures with average pore diameters ranging from 6.6 to 11.4 nm which is consistent with many of our previous work. Schiff base 1 had the largest pore volume (0.0086 cm$^3$/g) and diameter (11.4 nm). Materials with pores diameter of 10 to 15 nm have been proven to be optimal for adsorption [55]. The specific surface areas of 1–3 were low, with Schiff base 1 with a meta arrangement (Figure 8) exhibiting the highest surface area (20.3 m$^2$/g). The surface areas of trimethoprim Schiff bases (4.2–20.3 m$^2$/g) were relatively larger than those of carvedilol metal complexes (6.1–9.0 m$^2$/g) [42], melamine Schiff bases (5.2–11.6 m$^2$/g) [46], and polysilicates (8.2–18.0 m$^2$/g) [47], albeit smaller than those of telmisartan tin complexes (32.4–130.4 m$^2$/g) [43], valsartan metal complexes (16.0–22.8 m$^2$/g) [44], fusidate metal complexes (31.2–46.9 m$^2$/g) [45], and polyphosphates (24.8–213.5 m$^2$/g) [48,49].
Figure 8. *meta*-arrangement in 1 and *para*-arrangement in 2 and 3.

Table 3. Porosity of Schiff bases 1–3.

| Schiff Base | Specific Surface Area (m²/g) | Pore Volume (cm³/g) | Average Pore Diameter (nm) |
|-------------|------------------------------|---------------------|---------------------------|
| 1           | 20.3                         | 0.0086              | 11.4                      |
| 2           | 19.8                         | 0.0067              | 8.3                       |
| 3           | 4.2                          | 0.0036              | 6.6                       |

3.4. CO₂ Uptake of 1–3

Various factors influence the adsorption of CO₂ over an absorbent, including pressure, temperature, pore volume and size, and surface area of the adsorbent materials. In addition, the magnitude of the interaction between CO₂ and the absorbent is highly important [56,57]. A series of experiments were performed to optimize the pressure condition (1–40 bar). The CO₂ adsorption isotherms of 1–3 at 313 K and 40 bar are shown in Figure 9, and the CO₂ uptake results are listed in Table 4. Excellent CO₂ uptake (up to 46 wt%) was achieved over the present Schiff bases. This high CO₂ adsorption capacity can be attributed to the appropriate pore size distribution (shape, volume, and diameter) and the strong van der Waals interaction between adsorbents 1–3 and CO₂. Moreover, the adsorption of gas could be further enhanced by the possible formation of hydrogen bonding between CO₂ and the –HC=N–, OH, and NH₂ moieties of the Schiff bases. The polar nature of the heteroatoms (N and O) contained in the Schiff bases could contribute to the interaction between CO₂ and the adsorbents. In addition, the strong Lewis base nature of 1–3 could be conducive to the CO₂ capture. In fact, porous materials containing heteroatoms such as N, O, Si, and P have been used as CO₂ adsorbents [47–49].

Table 4. CO₂ uptake capacity of Schiff bases 1–3ᵃ.

| Schiff Base | CO₂ Uptake (cm³/g) | CO₂ Uptake (mmol/g) | CO₂ Uptake (wt%) |
|-------------|--------------------|---------------------|------------------|
| 1           | 230.2              | 10.3                | 46               |
| 2           | 214.6              | 9.6                 | 43               |
| 3           | 133.7              | 6.0                 | 27               |

ᵃ Measurements were performed at 313 K and 40 bar.
Figure 9. CO\textsubscript{2} adsorption isotherms of Schiff bases 1–3.

Schiff base 1, with a \textit{meta}-arrangement, exhibited the highest CO\textsubscript{2} uptake (46 wt\%), followed by 2 (43 wt\%) and 3 (27 wt\%), which have a \textit{para}-arrangement (Figure 8). Both 1 and 2 have a higher surface area (20.3 and 20.8 m\textsuperscript{2}/g, respectively) than that of 3 (20.8 m\textsuperscript{2}/g). In addition, 1 and 2 have larger pore volume and diameter than those of 3. The present results indicate that not only the surface area of the adsorbents but also their porosity affected the adsorption capacity, being the effect of the latter particularly important. Indeed, although the surface areas of 1–3 were low, their CO\textsubscript{2} uptake was remarkable. Clearly, the high CO\textsubscript{2} adsorption capacity over Schiff base 1 can be due to a combination of various factors including pore volume and diameter and a strong interaction with CO\textsubscript{2}.

The CO\textsubscript{2} capture over 1–3 was much better than over other absorbents under similar conditions (high pressures and temperatures) [42–49]. For instance, CO\textsubscript{2} adsorption capacity over metal complexes containing a carvedilol moiety was very limited (2.1–3.5 wt\%) [42]. The efficiency of nickel (3.5 wt\%) and copper (3.3 wt\%) complexes was higher than that of a cobalt complex (2.1 wt\%). Organotin complexes bearing a telmisartan unit afforded CO\textsubscript{2} uptake of 7.1 wt\% [43], and that over valsartan metal complexes was as low as 6.8 wt\% [44]. The CO\textsubscript{2} uptake over a manganese fusidate complex, which has a relatively high surface area, was 7.2 wt\% [45]. The CO\textsubscript{2} uptake over copper (6.7 wt\%) and zinc fusidate (6.3 wt\%) complexes was lower than that over a manganese-containing adsorbent. Better CO\textsubscript{2} uptake (10.0 wt\%) was observed over Schiff bases containing a melamine unit, which have very small surface areas [46]. CO\textsubscript{2} uptake of 6.0 wt\% was achieved over polysilicates with a \textit{meta}-arrangement [47], and highly aromatic polyphosphates achieved much better CO\textsubscript{2} uptake (14.0 wt\%) [48]. By contrast, other polyphosphates with a high surface area led to low CO\textsubscript{2} uptake (6.0 wt\%) [49]. Other materials with high surface areas, such as nanocarbons, afforded reasonable CO\textsubscript{2} uptake under mild pressure and temperature conditions [31–33].

4. Conclusions

An efficient and simple process is developed for the synthesis of three new Schiff bases containing a trimethoprim moiety. The Schiff bases, which have mesoporous structures with a small surface area, act as strong Lewis bases and promote electrostatic interactions with CO\textsubscript{2}, exhibiting a high CO\textsubscript{2} adsorption capacity. The 3-hydroxyphenyl-substituted derivative with a \textit{meta}-arrangement has the highest surface area and pore size distribution and serves as an efficient CO\textsubscript{2} storage material compared with its counterparts exhibiting a \textit{para}-arrangement. The results presented herein demonstrate that the newly synthesized
Schiff bases are more efficient as adsorbents for CO$_2$ capture than other reported porous materials under similar conditions.

**Supplementary Materials:** The following are available online at https://www.mdpi.com/article/10.3390/pr9040707/s1, Figure S1: FTIR spectrum of 1, Figure S2: FTIR spectrum of 2, and Figure S3: FTIR spectrum of 3.

**Author Contributions:** Conceptualization and experimental design: E.T.B.-T., G.A.E.-H., D.S.A., M.A.B., M.H.A.-M., and E.Y.; experimental work and data analysis: A.A.Y.; writing—original draft preparation: G.A.E.-H., D.S.A., and E.Y.; and writing—review and editing: G.A.E.-H., D.S.A., and E.Y. All authors have read and agreed to the published version of the manuscript.

**Funding:** The authors are grateful to the Deanship of Scientific Research, King Saud University for funding through Vice Deanship of Scientific Research Chairs.

**Institutional Review Board Statement:** Not applicable.

**Informed Consent Statement:** Not applicable.

**Data Availability Statement:** Data are contained within the article.

**Acknowledgments:** We thank Tikrit and Al-Nahrain Universities for technical support.

**Conflicts of Interest:** The authors declare no conflict of interest.

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