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RESEARCH NOTE

Novel Amine Modified Nanoporous SBA-15 Sorbent for the Removal of H_2S from Gas Streams in the Presence of CH_4

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ABSTRACT

A new sorbent has been developed by grafting Hexamethylenetetramine on a mesoporous molecular sieve (SBA-15) by wet impregnation method. This sorbent (Hexamine/SBA-15) has been characterized using N_2 physisorption, X-ray diffraction (XRD) at low angles (2θ°) and FT-IR. To investigate the H_2S removal capacity of this adsorbent from 9000 ppm H_2S balance CH_4-containing gas mixtures, the breakthrough tests have been performed on a fixed-bed flow system. SBA-15 without amine loading showed a very low H_2S adsorption capacity (0.015 mmol-Sulfur/g-sorbent) and saturation capacity (0.032 mmol-Sulfur/g-sorbent). The results show that after the amine loading, hexamine is dispersed inside the mesochannels of mesoporous SBA-15. Effects of the amine loading and sorption temperature on the sorption behavior of this developed sorbent have also been studied. Increase of amine loading enhances the H_2S sorption on this sorbent, When Hexamine was 50wt%, the removal performance was optimum(q_b= 0.9 mmol/g-sorbent).With increase in temperature, both the breakthrough capacity (q_b) and the saturation capacity (q_s) decrease. High sorption capacity is observed at low temperature (T=22°C, q_s= 1.9 mmol/g-sorbent) which provides a promoting effect in removal of H_2S from gas mixtures streams on the developed Hexamine/SBA-15 sorbent. This developed sorbent is regenerated easily at 100°C, and shows very good stability. The breakthrough capacity decreased from 0.9 mmol. g^{-1} for the first cycle to 0.75 mmol g^{-1} for the second cycle and 0.74 mmol g^{-1} for the third cycle. Although the breakthrough capacity decreased about 83 % of the first breakthrough tim, H_2S can be recovered.

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1. INTRODUCTION

Many gas streams, such as biogas and natural gas always contain hydrogen sulfide (H_2S) which is malodorous, corrosive and poisoning for the industrial catalysts [1]. The atmosphere would also be polluted if the used gas contains hydrogen sulfide. The industrial installations and pipe line would be corroded even if the gas streams contain minor amount of hydrogen sulfide. Therefore, removal of hydrogen sulfide less even than ppm level is necessary before gas utilization [2, 3]. Various technologies such as adsorption, chemical absorption (absorption in liquid alakanolamine, ammonia solution and alkaline salt solution), adsorption/oxidation (metal oxide), membrane separation, and biological means have been used for removal of hydrogen sulfide from industrial gas streams [4-7]. Concern about H_2S removal at low temperature is growing because it is a by-product of many of industrial processes [8]. Many metal oxides have been used as sorbents for removing hydrogen sulfide by chemical adsorption/oxidation from gas streams, respectively. However, the adsorbents used for chemical adsorption can be regenerated only at high temperature [9-15].

Therefore, it is necessary to develop a novel solid sorbent that has high capacity and high selectivity for removing acidic gases (H_2S, CO_2). It can also be easily regenerated which allows the sorption and regeneration to be conducted more conveniently. A new concept called 'molecular basket' has been developed more recently for the design of more selective sorbents [15-24]. In the 'molecular basket' sorbent, a polymer or amine molecule with a high density of desired amine groups such as polyethyleneimine (PEI), methyl-

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diethyl-amine (MDEA) are packed into the channels of a mesoporous molecular sieve such as SBA-15, CMK-3, MCM-41 and MCM-48 and are used for removal of pollution [25-30].

In this paper, we report the synthesis and characterization of a novel Hexamine/SBA-15 adsorbent with high sorption capacity for H_2S gas. This new sorbent with better regeneration capability and high stability has been effectively employed in removal of H_2S from gas mixture streams.

2. EXPERIMENTAL

2. 1. Preparation of the Sorbents Mesoporous molecular sieve (SBA-15) was hydrothermally synthesized according to the method reported earlier [6, 8, 12]. Typically, a homogeneous mixture which was composed of 4g pluronic P123 (polyethyleneglycol)-block-poly(propyleneglycol)-block-poly(ethyl-eneglycol) as the template and 9.6ml tetraethyl orthosilicate (TEOS) as the silica source in hydrochloric acid (150ml, 2N) was stirred at 40°C for 24h and further treated at 100 °C for 24h. After the synthesis, the resultant solid was recovered by filtration, washed and dried at 100°C for 24h and finally calcined at the rate of 1°C min^{-1} to 550°C and keeping it 550°C for 6 h to remove the template.

The amine modified SBA-15 sorbents were prepared by a wet impregnation method. In a typical procedure, the desired amount of Hexamine was dissolved in 20 g of methanol (For synthesis of 50wt%Hexamine/SBA-15, 1 g Hexamine was added in 1.0 g calcined SBA-15 in the solution.) under stirring, and then 1.0 g calcined SBA-15 was added to the above solution and further stirred and refluxed for 24h. The slurry was then dried at room temperature. Before sorption studies, the sorbent was pretreated at 100 °C with nitrogen (99.999%) at 50 ml min^{-1} flow for overnight.

2. 2. Characterization of the Sorbents The SBA-15supported Hexamine sorbent was characterized by N_2 physisorption, X-ray diffraction (XRD) and FT-IR. Textural property of the SBA-15 before and after modification with Hexamine was measured by N_2 adsorption using the Micromeritics ASAP 2020 analyzer to examine the porosity and surface area of each sample at 77 K. The relative pressure of N_2 covered from 0.080 to 0.988. The SBA-15 samples were degassed at 363K for 7 h before measuring. XRD was performed with a SWAXS model S3-MICRO X-ray diffractometer with Cu -Kα radiation at low angle range (2θ = 0.01–10°).

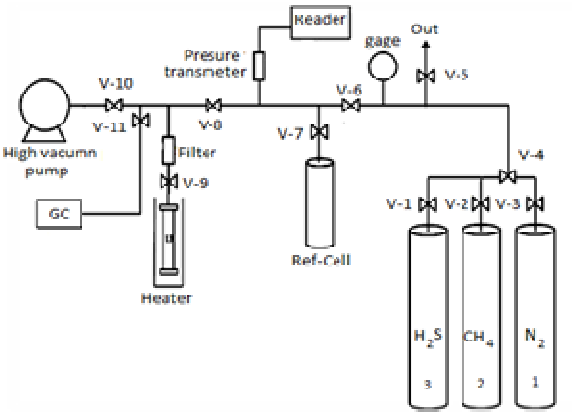

Figure 1. Schematic diagram of the fixed-bed flow system for H_2S adsorption measurements

Infrared spectroscopy (FTIR Shimadzu solution Model 8400) was used to study the characteristic surface species of the SBA-15 before and after modification.

2. 3. H_2S Adsorption Removal Studies to investigate the H_2S removal performance of the adsorbent, the breakthrough tests were performed in a fixed-bed flow system that was assembled in our laboratory (Figure 1).

The sorption tube was made of stainless steel with length of 120mm and inner diameter of 15mm packed with the adsorbent. Before sorption, the sorbent bed was heated to 100 °C under nitrogen (99.999%) at 15 ml min^{-1} flow over night to ensure that it was free of any impurity. When the bed was cooled to the sorption temperature, a model gas that contained 0.9% H_2S in CH_4 was introduced into the sorption bed with flow rate of 62 ml min^{-1}.

All the sorption tests were conducted at atmospheric pressure. H_2S concentration in the exhaust gas was measured by gas chromatograph (Varian CP-3800) equipped with a Thermal conductivity detector (TCD). When the outlet H_2S concentration reached the initial feed concentration, the sorbent was saturated by H_2S. When the sorbent was saturated, sorbent bed temperature was increased to 100°C and held at this temperature to perform the desorption and regeneration steps. The breakthrough capacity (denoted as q_b (mmol-sulfur/g sorbent) was calculated according to the following equation:

$$q_b = \frac{\rho * t * F * AS1 * CI}{AS2 * 10^6 * Mw * W} \quad (1)$$

where, ρ was the pure H_2S density, T was the breakthrough time, the time (min) when the H_2S concentration at outlet reached 1 ppmv, F was the flow rate (ml min^{-1}), AS1 was the GC area from gas sample

injection after adsorption, CI was the initial H_2S concentration (9000ppmv), AS2 was the GC area from gas sample injection before adsorption, Mw was the molecular weight of Sulfur and W was the weight of sample (in gram). Saturation capacity (denoted as q_s,mmol-sulfur/g sorbent) was calculated by integration of the area between the breakthrough curve and the initial H_2S line (9000 ppmv).

3. RESULT AND DISCUSSION

3. 1. Textural Property of the SBA-15 and Modified SBA-15 Material Figure 2 shows the X-ray diffraction patterns of calcined SBA-15 and 50 wt% Hexamine /SBA-15. The XRD reflection peaks are in good agreement with three characteristic peaks (100), (110) and (200) for the 2-D hexagonal structures of SBA-15 [12]. These peaks were still observed in the diffraction pattern of 50 wt% Hexamine/SBA-15. However, the intensity of the diffraction patterns of SBA-15 samples after loading Hexamine decreased which was caused by the pore filling with amine molecules.

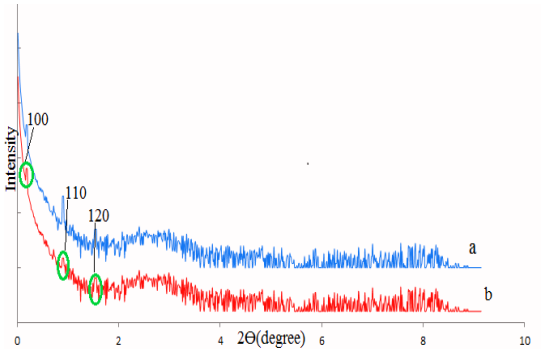

Figure 2. X-ray diffraction patterns of SBA-15(a) and Hexamine (50wt %) /SBA-15(b).

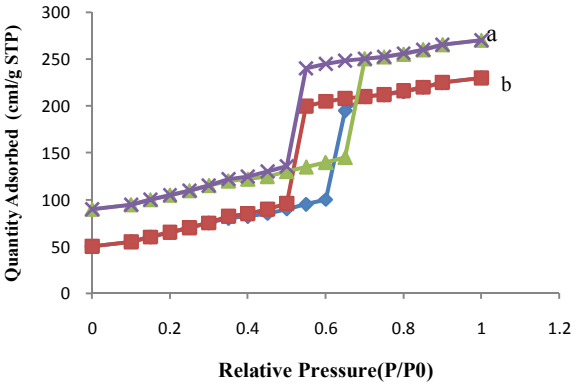

Figure 3. Adsorption isotherms of N_2 on the SBA-15(a) and Hexamine (50wt %) /SBA-15(b)

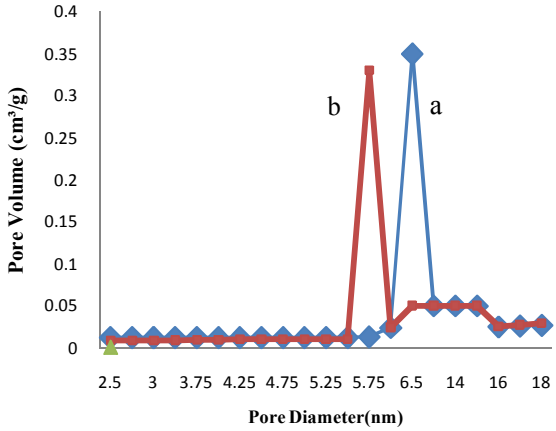

Figure 4. Pore size distribution of SBA-15(a) and Hexamine (50wt %) /SBA-15(b)

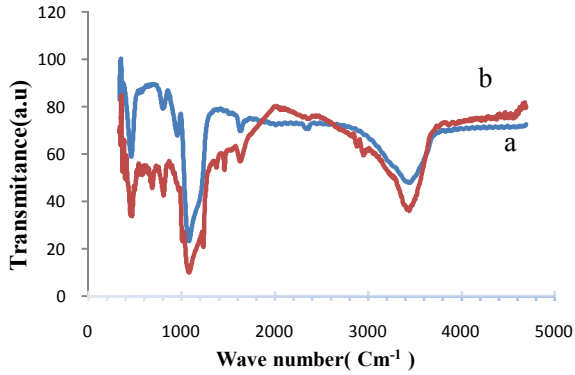

Figure 5. FT-IR spectra of SBA-15(a) and Hexamine (50wt %) /SBA-15(b)

The nitrogen adsorption–desorption isotherms at 77K for the SBA-15 and 50 wt% Hexamine/SBA-15 have been shown in Figure 3.The nitrogen adsorption isotherm of the calcined SBA-15 was a typical reversible type IV adsorption isotherm (S-shape) which is the characteristic of a mesoporous molecular sieve [12]. After Hexamine loading, the S-shaped adsorption isotherm is still preserved.

The pore size distributions evaluated by the BJH model from the adsorption data of nitrogen for SBA-15 and Hexamine/SBA-15 are shown in Figure 4. After amine loading, the pore size, surface area and pore volume decreased (b). The pore size decreased from 6.55 nm to 5.75 nm which could be owing to the formation of an amine layer in the pores. These results were in agreement with the records of XRD characterization. Detailed changes in the BET surface area (S_{BET}), pore size (d) and pore volume (V) were shown in Table 1. Figure 5 shows the FT-IR spectra of calcined SBA-15(a) and that of 50 wt% Hexamine/SBA-15(b) in the scan range of 400 to 4000 cm^{-1}.

Two sharp absorption bands at 1031 and 1630cm^{-1} and a broad band at around 3450cm^{-1} present in the IR spectrum of the calcined SBA-15. These bands area assigned are to single Si–OH and hydrogen-bonded Si–OH groups, respectively [12]. The bands at 2700-3800 cm^{-1} assigned to the CH_2 asymmetric and symmetric stretching modes of the Hexamine modified SBA-15 was attributed to the –NH stretching vibration. The absorbance of the C–N stretching vibration is normally observed around 900–1200cm^{-1}. In the spectrum of Hexamine/SBA-15(b), the bands at 1500-2000 cm^{-1} were attributed –NH vibration, while that emerging at 1660 cm^{-1} was assigned to the bending [–N(R)] in Hexamine.

3. 2. H_2S Sorption Performance

3. 2. 1. Effect of Sorption Temperature The sorption of H_2S over Hexamine (50wt %) /SBA-15 was observed at 22, 55, and 80°C, respectively, under a flow rate of 62 ml min^{-1}. Figure 6 shows the breakthrough curves.

The corresponding sorption capacities were calculated on the basis of the adsorption isotherms and the results are listed in Table 2. It is observed that breakthrough sulfur capacity of Hexamine (50wt%) /SBA-15 decreases from 0.9 to 0.022 mmol/g-sorbent with the increase of adsorption temperature from 22 to 80°C implying that the sorption temperature has a very strong impact on the sorption performance of the amine modified sorbent.

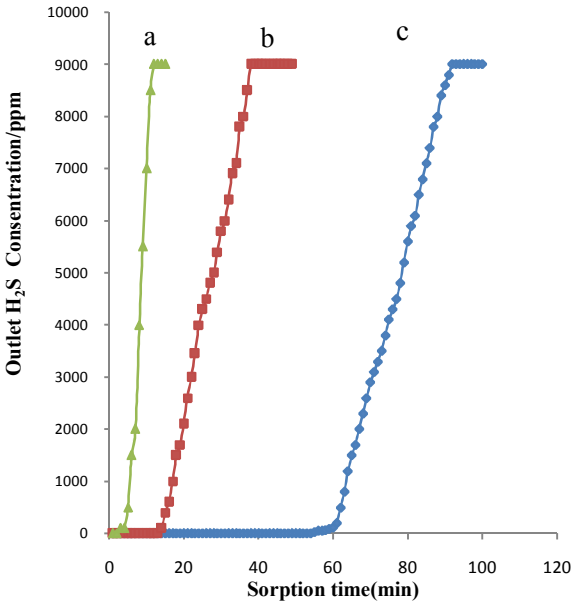

Figure 6. Breakthrough curves of H_2S on Hexamine (50wt %) /SBA-15 at different sorption temperature. (a)80°C, (b)55°C, (c)22°C. Conditions: Flow rate, 62 ml min^{-1}; Gas composition, 9000ppm H_2S in CH_4.

TABLE 1. Properties of SBA-15 material and Hexamine (50 wt%)/ SBA-15.

Sample	S_{BET}(m^2 g^{-1})	d(nm)	V(cm^{3}g^{-1})
SBA-15	937	6.55	1.17
Hexamine(50)/SBA-15	115	5.75	0.34

TABLE 2. Effect of sorption temperature on H_2S removal over Hexamine (50wt%) /SBA-15 sorbent

Temp.(°C)	q_b(mmol/g-sorbent)	q_s(mmol/g-sorbent)
22	0.9	1.9
55	0.32	0.71
80	0.022	0.087

TABLE 3. Effect of Hexamine loading on the H_2S sorption performance of Hexamine/SBA-15 sorbents.

Sample	q_b(mmol/g-sorbent)	q_s(mmol/g-sorbent)
SBA-15	0.015	0.032
Hexamine(30wt%)/SBA-15	0.3	0.65
Hexamine(50wt%)/SBA-15	0.9	1.9
Hexamine(65wt%)/SBA-15	0.7	2.35
Hexamine(80wt%)/SBA-15	0.44	0.87

At 22°C, the breakthrough capacity (q_b) and saturation capacity (q_s) of Hexamine (50wt %) /SBA-15 were 0.9 and 1.9 mmol/g-sorbent, respectively, which are about 41 times as large as those at 80°C (0.022 and 0.087 mmol/g-sorbent, respectively).

This saturation capacity is significantly higher than that reported in literature for the Cobalt-Zinc oxide adsorbent [12] and Amine-Grafted MCM-48 [6]. It is also in the same magnitude as those reported for the Mesoporous-molecular-sieve-supported, polymer Sorbents [1] and mixed metal oxides [9] at the comparable sorption temperature.

3. 2. 3. Effect of Hexamine Loading In order to examine the effect of Hexamine loading on the H_2S sorption performance, sorption of H_2S over Hexamine/SBA-15 sorbents with different loadings (30, 50, 65, 80%wt) was conducted at 295 K. The H_2S breakthrough curves of the SBA-15 samples before and after amine loading are shown in Figure 7 and the corresponding breakthrough adsorption capacity (q_b) and saturation capacity (q_s) are listed in Table 3.

SBA-15 without amine loading showed a very low H_2S adsorption capacity (0.015 mmol/g-sorbent) and saturation capacity (0.032 mmol/g-sorbent). This low sorption capacity might be caused due to the physisorption of H_2S on SBA-15 pore channel.

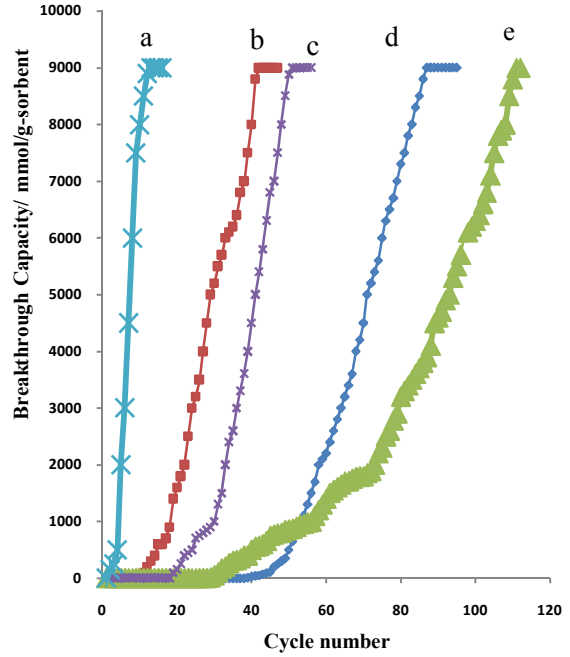

Figure 7. Breakthrough curves of H_2S on the supported Hexamine sorbents. (a) SBA-15, (b) Hexamine (30wt %) /SBA-15, (c) Hexamine (80wt %) /SBA-15, (d) Hexamine (50wt %) /SBA-15, (e) Hexamine (65wt %) /SBA-15.Conditions: Flow rate, 62 ml min^{-1}; T, 22 °C; gas

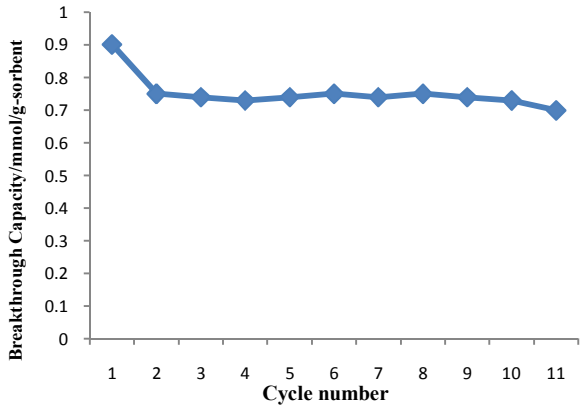

Figure 8. The breakthrough capacity as a function of the number of the Sorption-desorption cycles for Hexamine (50wt%)/SBA-15. Conditions for sorption: flow rate, 62 ml.min^{-1}; T, 22 °C; Gas composition, 9000ppm H_2S in CH_4; and conditions for desorption, N_2, flow

As well known, Hexamine contains numerous amine atom (28.53mmol-N/ Hexamine), which have high affinity with H_2S on the basis of the acid–base interaction. Under dry conditions (without moisture), the main reaction mechanism between H_2S and Hexamine were depicted in Equation (2).

$$H_2S+R_3N \xrightarrow{\nabla} R_3NH–S–HNR_3 \quad (2)$$

when the loading of Hexamine is increased from zero to 50wt%, the breakthrough capacity also increased from 0.017 to 0.9 mmol/g-sorbent, and then, decreased remarkably with further increase in the amine loading. However, the important point is that the maximal breakthrough capacity was at a 50wt% of Hexamine loading, while the maximal saturation capacity was at a 65 wt% of Hexamine loading.

3. 3. Regenerability of the Sorbent To investigate the regeneration and stability of the removal performance, the operations of adsorption cycle were carried out on Hexamine (50wt %) /SBA-15. After saturation, the desorption was conducted at 100°C with pure N_2 purging at 100 mL min^{-1} overnight.

As shown in Figure 8, the breakthrough capacity decreased from 0.9 mmol/g-sorbent for the first cycle to 0.75 mmol g^{-1} for the second cycle and 0.74mmol g^{-1} for the third cycle. Although the breakthrough capacity decreased about 83% of the first breakthrough time, H_2S can be recovered. These results suggest that the adsorbent still has good regenerability and stable sorption performance.

4. CONCLUSIONS

In the present work, we have found that the SBA-15-supported Hexamine sorbent could effectively remove H_2S from gas streams to at least less than 1ppm at ambient conditions.

It is observed that with decrease of the adsorption temperature, both the breakthrough capacity (q_b) and the saturation capacity (q_s) increases. Increase in amount of Hexamine loading on the porous silica SBA-15 has strong influence on the sorption performance of the sorbents. A loading of 50wt % Hexamine gives the largest breakthrough capacity of 0.9 mmol of Sulfur/ (g - sorbent) at 22°C. In addition, prepared sorbent shows good regenerability and stable sorption performance at 100 °C. Due to the large breakthrough capacity and saturation capacity and also excellent removal performance, this can be effectively used for effectively removing H_2S from the gas streams.

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RESEARCH NOTE

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چکیده

در این مطالعه نوع جدیدی از جاذب‌ها به منظور حذف هیدروژن سولفید با استفاده از بارگذاری هگزامتیلن ترا آمین بر روی مزوپروس(SBA-15) سنتز شد. جاذب‌های سنتز شده با استفاده از تکنیک‌های جذب – واجذب نیتروژن، پراش اشعه و امواج مادون قرمز تبدیل فوریه مورد شناسایی قرار داده شد. به منظور بررسی میزان قابلیت جذب هیدروژن‌سولفید از مخلوط گازی متان- هیدروژن‌سولفید که دارای غلظت ۹۰۰ پی پی ام هیدروژن‌سولفید که در تعادل با متان است و همچنین از یک سیستم با بستر ثابت استفاده شد. ترکیب SBA-15 به صورت خالص مقدار جذب بسیار کمی از خود نشان می‌دهد(۰.۰۱۵ میلی مول سولفور بر گرم جاذب برای جذب بیشینه و ۰.۰۳۲ میلی مول سولفور بر گرم جاذب برای جذب اشباع شونده). شواهد نشان می‌دهد که بعد از بارگذاری آمین، هگزامین در درون ساختار کانالی SBA-15 قرار می‌گیرد. تاثیر میزان آمین بارگذاری شده همچنین دمای به کاربرده شده در حین فرایند جذب در حین مطالعه مورد بررسی قرار داده شد. افزایش میزان آمین بارگذاری شده، باعث افزایش مقدار جذب هیدروژن سولفید شد زمانی که مقدار هگزامین بارگذاری شده به ۵۰ درصد وزنی کل جاذب برسد، جذب مقدار بیشینه می باشد (جذب بیشینه= ۰.۹ میلی مول بر گرم جاذب). در حالی که با افزایش دمای جذب، هم ظرفیت با بیشینه جذب و هم ظرفیت اشباع شوندگی هر دو کاهش یافت. جذب بالا برای هیدروژن سولفید در دمای پایین مشاهده شد(دمای جذب=۲۲ درجه سلسیوس، جذب بیشینه= ۱/۹ میلی مول بر گرم جاذب) که نشانگر تاثیر مستقیم دما بر روی میزان جذب هیدروژن‌سولفید به وسیله جاذب بارگذاری شده با هگزامین می‌باشد. جاذب‌های سنتز شده در دمای ۱۰۰ درجه سلسیوس بازیابی بسیار خوبی را نشان دادند. ظرفیت جذبی در مرحله اول جذب از مقدار ۰/۹ میلی مول بر گرم به مقدار ۰/۷۵ میلی مول بر گرم در چرخه دوم و به مقدار ۰/۷٤ میلی مول بر گرم برای چرخه سوم دست یافت. مقدار بازیابی جاذب مقدار ۸۳٪ نسبت به چرخه اول رسید.

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