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Rates of Adsorption on Hydrotalcite Pellets with Respect to Modifications of Area for Removal of CO_2

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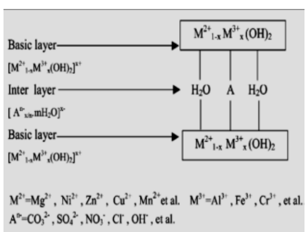
Graphical abstract



Abstract

Hydrotalcite (HTC) has recently attracted the attention of researches in CO_2 removal technology because of its ability to adsorb appreciable amounts of CO_2 compared with many other adsorbents. It is known that pelletized forms of HTC could meet commercial applications which can be easily handled, transported and are of market appeal. The objective of this research is to study the rates of adsorption of CO_2 which is an important aspect in the determination of the viability of the removal of CO_2 with respect to the modifications of HTC's surface area that have been encountered. Some modifications of HTC morphology and surface structures with adsorption of CO_2 was observed based on Scanning Electron Microscopy, Thermal Gravimetric Analyzer, X-Ray Diffraction Analyzer and analysis of adsorption area of HTC towards CO_2. The modifications of area could be contributed by the disintegration and/or agglomeration effects. A model based on the Langmuir rate model was developed in order to investigate the interesting behavior of HTC area towards the adsorption of CO_2. Based on a cross sectional area of a mole of CO_2, which is 136.4 m^2/mmol as reported by Aylmore *et al*. [31], the monolayer areas of HTC after adsorption of CO_2 is calculated to be 177.32 m^2/g. This value when being compared with the Brunauer Emmett and Teller (BET) value of commercial HTC which is 110.98 m^2/g, it was found that there is an increase in equivalent areas of HTC after the adsorption of CO_2.

Keywords: CO_2; hydrotalcite; pellets; rates of adsorption; adsorption area

1.0 INTRODUCTION

Separation, capture and storage of carbon dioxide have received significant attention in recent years due to its adverse contribution to the global warming. There are a number of separation technologies that has been applied to CO_2 capture. These involved processes like absorption, membrane and adsorption. Large scale removal of CO_2 from natural gas, flue gas, synthetic gas and other industrial gases is commonly accomplished using amine based absorption which suffers from inherent regeneration cost and inefficiency. Adsorption is considered to be competitive and viable method for removal of CO_2 in comparison with other technologies [1-2].

A few inorganic materials like zeolite and activated carbons have been found to have good adsorption capacities for CO_2. However, they are not attracted to high temperature applications because CO_2 adsorption capacities of these materials decreased drastically with high temperatures [3]. Therefore a trend with the applications of HTC has emerged because the HTC materials exhibit high selectivities and good adsorption capacities for CO_2 at high temperature.

HTC possesses capabilities to separate carbon dioxide under difficult conditions because of high abrasion resistance, high thermal stability and small micropore diameter which results in higher exposed surface area and hence high capacity of adsorption and stable interdispersion of the active species with high reproducibility [4-5] for adsorption of CO_2. Due to the homogenous interdispersion of the constituting elements in the HTC matrix, the mixed oxides in HTC formed upon the thermal decomposition of anionic clays possess unique properties [6]. Their most important applications are due to their permanent anion-exchange and adsorption capacity, the mobility of their interlayer anions and water molecules and the stability and homogeneity of the materials formed by their thermal decomposition [7].

HTC is a natural layered mineral or anionic clay, constitute a class of lamellar ionic compound. Layered double hydroxides (LDH) also called hydrotalcite like compounds is synthetically prepared [8-9]. It contains a positively charged (cations) hydroxide layer or brucite sheet and charge-balancing anions which is carbonate in the interlamellar space besides water molecule as shown in Figure 1. HTC like compounds are represented by the general formula,

$$\left[M^{2+}_{1-x}M^{3+}_{x}(OH)_2\right]^{x-}\left[A^{n-}_{x/n}\right]mH_2O$$

where, M^{2+} and M^{3+} are bi and tri-valent metal cations, respectively and A is an interlamellar anion with charge n- [10].

HTC have promoted much interest over the last two decades because of the versatile properties that they show in various fields like post combustion capture applications, purification of natural gas and many more. These properties are mainly related to the structural features of the compounds [11]. HTC has the ability to reconstruct their structure when exposed to water and CO_2 as thermal decomposition occurred. Therefore, these materials have the potential applications for adsorption of CO_2 at high temperature [12].

It was reported that HTC produced by calcinations has potential for CO_2 adsorption [13]. Previous investigations revealed that HTC undergoes interlayer water dehydration, dehydroxylation of layered hydroxyl, OH^- groups and the release of interlayer CO_3^{2-} groups in various temperature regimes, finally leading to the formation of amorphous Mg/Al mixed solid oxides with a larger surface area and good stability at high temperatures which makes the mixed oxide a viable material for CO_2 adsorption [2, 14]. However, at low calcinations temperatures which in the range of 100-300°C, HTC loses interlayer water. The mixed oxides obtained exhibited peculiar properties such as high surface areas and narrow pore size distribution [15].

Many researchers have presented their work on the absorption and adsorption of CO_2 on HTC. These mainly involved adsorption batch studies on HTC powders [8-9, 16] and HTC membranes based on sol gel preparations [18]. Several studies have presented adsorption isotherms for HTCs [2, 13, 16, 19] and HTC like compounds with additives [7, 13, 20-22]. Industrial applications of powders are known to be cumbersome due to the handling difficulties and environmental issues related to dust. Under such circumstances pelletized forms of HTCs could be appealing for industrial usage. However, studies on adsorption of CO_2 on HTC in pellet form have not yet been thoroughly understood.

The objective of this research is to study the rates of adsorption of CO_2 which is an important aspect in the determination of the viability of the removal of CO_2 with respect to the modifications of HTC's surface area. The modifications of HTC morphology and surface structures with adsorption of CO_2 will be observed based on Scanning Electron Microscopy (SEM), Thermal Gravimetric Analyzer, X-Ray Diffraction (XRD) Analyzer and analysis of adsorption area of HTC towards CO_2. Details of the work are presented in Section 2.0 and 3.0.

2.0 EXPERIMENTAL

2.1 Materials and Equipment

HTC powders were purchased from Tomita Pharmaceuticals, Japan. Table 1 shows the properties of commercial HTC and the chemicals used.

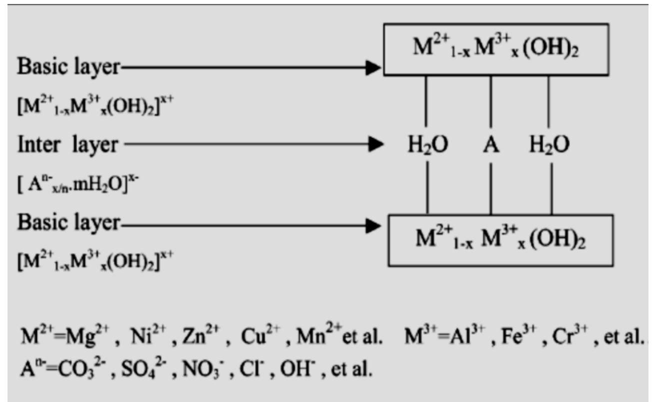


Figure 1 2-D structure models for hydrotalcite [37, 38]

Table 1 Properties of HTC (Tomita AD500)

Properties	Content
Aluminium oxide	15.8%
Magnesium oxide	37.3%
Carbon dioxide	8.1%
Chloride	0.3% max
Sulfate	0.5% max
pH	9.0 %
Loss on drying	5. 7%
Apparent volume of material	28 mL/10g
Mean particle diameter	12.55μm *
Pore diameter	2.35nm *

*In situ measurement

A hot plate magnetic stirrer, an oven and a carbolite furnace were used in this study. Measuring equipments used were SEM, XRD and BET analyzers. An experimental rig consisting of a pressure gauge, a flow meter, a tubular batch reactor and a tube carbolite furnace was utilized in order to determine the rates of adsorption before it can be applied into the standard models of adsorption.

Figure 2 shows the schematic diagram of the experimental setup. A stainless steel tubular reactor loaded with HTC samples is connected to a 5% CO_2 cylinder gas, a pressure gauge and valves. The pressure gauge is used to monitor the pressure during the adsorption process. The line had a flow meter facility in order to monitor the flow of CO_2. The reactor is positioned vertically inside the tube carbolite furnace. A digital thermocouple was attached to the reactor in order to monitor the temperature of the furnace. A vacuum pump was used whenever necessary to evacuate gases from the reactor.

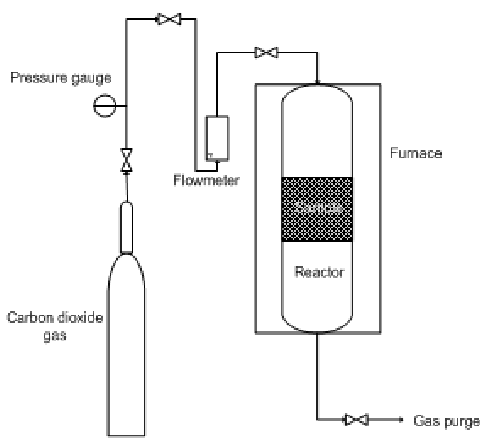


Figure 2 Schematic diagram of experimental setup

2.2 Methods

Initially, samples of raw HTC powder were investigated using SEM, XRD and BET analyzers. The SEM, XRD and BET studies were carried out on HTC samples after initially heating to 32°C, 300°C and 550°C and cooled to normal temperatures thereafter.

2.2.1 Preparation of HTC Pellet

HTC powder was pressed using tablet press machine into the required size (8 mm, 15 mm and 20 mm) in a mould with pressure loading of 1000psi. The weight of the pellet is determined by preliminary tests in order to obtain the thickness of the pellet of 0.20 cm. Later, the pellets were dried in an oven at 100°C for 8 hours.

2.2.2 Determination of Rate of Adsorption

Leak tests were conducted initially in order to check the possible leakages from connectors in the reactor. The unloaded reactor was filled with pressurized N_2 gas mixture until 1.5bar. Then, the pressure drops were checked and connectors were tightened until no pressure drop was indicated. Soap solution method was used to check leaks from joints.

The HTC pellets were loaded in the reactor and the lid is tightly closed. Leaks were checked again with N_2 gas. The vacuum pump is turned on until the vacuum pressure reached 0.01bar. The furnace was set to a desired temperature and the temperature of sample was allowed to reach the set point (300°C or 550°C). The furnace was not used whenever experiments were carried out for room temperature condition. CO_2 gas was allowed briskly to enter the reactor under pressure. After the pressure reached an optimum condition, all valves were closed. The pressure readings were collected every 10 minutes until the pressure did not indicate any further drop. At the end of the experiment, the furnace was switched off and the reactor was allowed to cool. The reactor was pressurized again and the leak tests were carried out again to ascertain that there have been no leaks during the experiment.

2.2 Preliminary Studies

Preliminary studies which consist of characterization processes were examination results from BET analysis using the Micromeritics ASAP 2020 analyzer and XRD analysis. HTC (Tomita AD500) was selected based on the surface area and adsorption capacity. Further characterization such as SEM, XRD and TGA were carried out on the HTC pellets.

2.3 Theoretical Studies : Extended Model of Langmuir with Surface Modifications

An extended model of Langmuir which is based on the Langmuir rate model shown in Equation 1 was developed in order to investigate the behaviour of modifications of HTC morphology and surface structures with adsorption of CO_2.

$$\frac{dQ}{dt} = k_1 Q_v p - k_{-1} Q \quad (1)$$

where Q_v is the vacant site of adsorption (g_{CO2}/g_{HT}), Q is the total CO_2 adsorbed within a time t of the experiment (g_{CO2}/g_{HT}), k_1 is the rate constant of adsorption (min^{-1}), k_{-1} is the rate constant of desorption (min^{-1}) and p is the partial pressure of CO_2 within the reactor at time t (bar).

The total pressure (P) within the reactor versus time (t) was used to estimate the total amount of CO_2 adsorbed within a time (t). The values of Q for different t were used in order to estimate the dQ/dt for different values of P as expressed in Equation 2.

$$\frac{dQ}{dt} = \frac{-44\,(V - v)}{RT}\frac{dP}{dt} \quad (2)$$

As been observed in the preliminary studies which are explained in Section 3.1, this model assumes that with CO_2 adsorption in HTC, changes in the internal surfaces of HTC were discovered with the extent of adsorption of CO_2. Therefore, it is assumed that there is a direct relationship between the increments (dQ_v) of the vacant sites with the increment (dQ) of adsorption of CO_2 (Equation 3). Equation 4 shows the relationship between Equation 1 and the integration between dQ_v and dQ. Equation 5 shows the simplified version of Equation 4.

$$dQ_v \propto dQ \quad (3)$$

$$\frac{dQ}{dt} = K_1 Q p + K_2 p - k_{-1} Q \quad (4)$$

$$\frac{1}{Q}\left(\frac{dQ}{dt} - K_2 p\right) = K_1 p - k_{-1} \quad (5)$$

where K_1 is $k_1\alpha$ (m^2/min), K_2 is $k_1\beta$ ($g_{CO2}/g_{HT}/min^2$), α is the proportional constant and β is the constant in the integration step of Equation 3. Equation 5 was used as an alternative model equation for the analysis.

Hence, a plot of $(1/Q)(dQ/dt - K_2 p)$ versus p should give a straight line with gradient K_1 and intercept $-k_{-1}$.

3.0 RESULTS AND DISCUSSION

Many HTC available were prepared by various methods such as co-precipitation, sol gel and hydrothermal. Table 2 presents the data obtained from the BET analyses of HTC from various origins. It can be seen that the commercial HTCs usually have BET surface areas of average values of 110.98±0.07 m^2/g whereas the laboratory prepared HTCs were 50.58±0.22 m^2/g [23-25]. The variation of BET areas of between these two samples was observed to be around 22% which is regarded as too high for dependence on the research investigations. HTC (Tomita-AD500) was studied using XRD analyzer to support the selection source.

Table 2 BET analyses of HTC from various origins

Hydrotalcite	BET surface area (m^2/g)	Reference
Hydrotalcite (Laboratory prepared)	50.58	This study
HTcp (Laboratory prepared)	51.00	Othman *et al*. 2006
HTsg (Laboratory prepared)	121.80	Othman *et al*. 2006
Hydrotalcite (Laboratory prepared)	153.95	Yang and Kim, 2006
Hydrotalcite (Laboratory prepared)	80.00-90.00	Kovanda *et al*. 2005
Hydrotalcite (Tomita-AD500)	110.98	This study
Hydrotalcite (PURAL MG30)	110.00	Yong *et al*. 2001
Hydrotalcite (PURAL MG50)	90.00	Yong *et al*. 2001
Hydrotalcite (PURAL MG70)	120.00	Yong *et al*. 2001

3.1 Preliminary Results

In order to understand the changes of chemical behavior of HTC with heat treatment, several samples of HTC were initially heat treated at 300°C and 700°C. Figures 4-6 show the XRD patterns for samples of the respective heat treated hydrotalcite samples. These figures were compared with the XRD patterns of hydrotalcite at 32°C shown in Figure 3.

It was observed from Figure 4 that the HTC phase starts to decrease at heat treatment temperature around 300°C. It was reported that the decrease of HTC phase could be due to dehydration of interlayer water molecules and dehydroxylation [15, 23-25]. Heating beyond 450°C seems to lead to dehydroxylation along with decarbonation which results in the destruction of HTC and formation of other mixed oxides such as periclase [12, 23-25]. HTC phase seems showing dominance of MgO phase at temperature 700°C. It was observed that HTC undergone chemical rearrangements when heat treated from 32°C up to 700°C.

In order to investigate the changes in physical structure of HTC with CO_2 adsorption and heat treatment, SEM images of HTC were examined. Figure 7 represents the SEM image of fresh HTC (Tomita AD500) at 32°C. Figure 8 shows the structure after reaction of the same sample with CO_2 at 32°C. It was observed that the particles were in rearranged structure with modified surfaces after reaction with CO_2 at 32°C. Similar findings for heat treated HTC at 450°C and 550°C prior and after reaction with CO_2 (Figures 9-12). The morphology shown in Figures 7-12 seem to be compatible with the structure of HTC (MG30-K) studied by Oliveira *et al*. [22] with HTC structure in agglomerates formation. Based on the above studies, it was indicated that the HTC particles are not morphologically stable and undergo changes in particle structures and at all temperature when CO_2 is adsorbed.

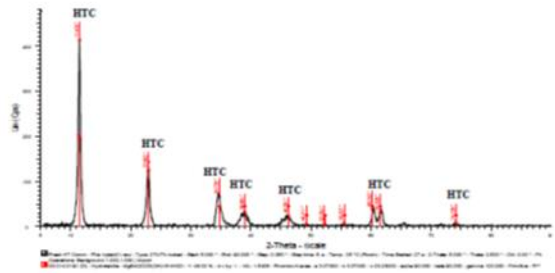


Figure 3 XRD pattern of HTC at 32°C (Tomita-AD500)

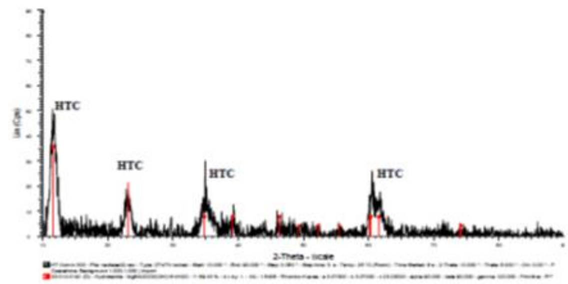


Figure 4 XRD pattern of HTC (Tomita-AD500) after subsequent heat treatment at 300°C

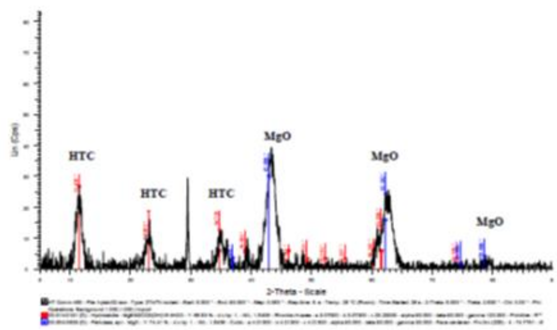


Figure 5 XRD pattern of HTC (Tomita-AD500) after subsequent heat treatment at 450°C

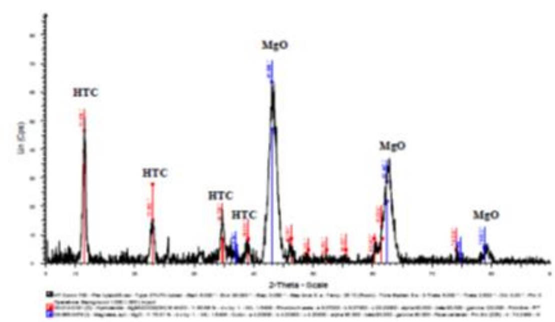


Figure 6 XRD pattern of HTC (Tomita-AD500) after subsequent heat treatment at 700°C

Figure 7 SEM image of HTC (Tomita AD500) prior to reaction at 32ºC

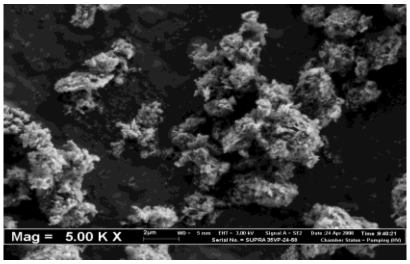


Figure 8 SEM image of HTC (Tomita AD500) after reaction with CO_2 at 32ºC

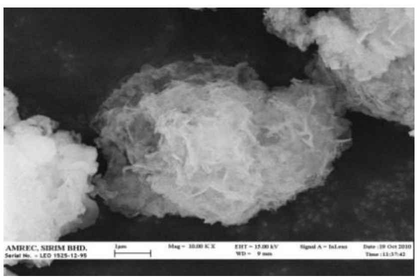


Figure 9 SEM image of HTC (Tomita AD500) prior to reaction at 450ºC

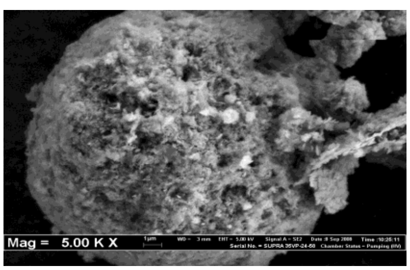


Figure 10 SEM image of HTC (Tomita AD500) after reaction with CO_2 and heat treatment at 450ºC

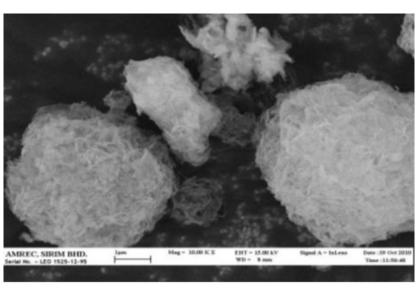


Figure 11 SEM image of HTC (Tomita AD500) prior to reaction at 550ºC

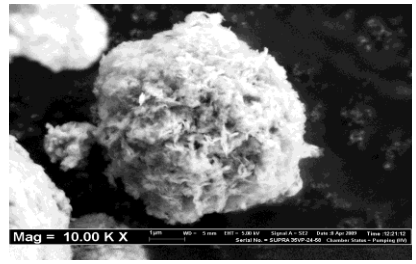


Figure 12 SEM image of HTC (Tomita AD500) after reaction with CO_2 and heat treatment at 550ºC

3.2 Analysis of Adsorption Area of HTC Towards CO_2

In order to investigate the effect of morphological changes on the effective surface area of HTC with temperatures and the adsorption of CO_2 as observed in SEM analysis on the effective surface area of HTC, analysis of adsorption capacity of HTC were carried out. Table 3 shows the maximum adsorption capacities of CO_2 of different HTCs that have been reported by previous studies as well as observed in this study. It can be seen that HTC sample gave the adsorption capacity of 1.3mmol/g which is comparable with the data from HTC sources.

Table 3 Maximum adsorption capacity of CO_2 on HTC

Hydrotalcite	Adsorption capacity (mmol/g)	Estimated equivalent monolayer area (m^2/g)	Reference
Hydrotalcite (Tomita AD500)	1.3	177.32	This study
HTlc	1.79	244.16	Hutson and Attwood, 2008
HTcp (600)	0.635	86.60	Othman *et al*., 2006

Based on a cross sectional area of a mole of CO_2 which is 136.4m^2/mmol as reported by Aylmore *et al*. [31], the monolayer areas of HTC after adsorption of CO_2 is calculated to be 177.32m^2/g. This value when compared with the BET value of 110.98m^2/g obtained for HTC (Tomita AD500), it can be observed that there is an increase in equivalent areas of the HTC after the adsorption of CO_2. This observation was compatible with the data for HTCs reported in Hutson and Attwood [30] and Othman *et al*. [23-25] as shown in Table 3. The same observation was also observed where the equivalent areas of HTC were increased by using the cross sectional area of a mole of CO_2 as been reported in Martin-Aranda *et al*. [32]. Heat treatments of HTC have also been reported to increase the surface areas by several authors such as Soares *et al*. [33-35] and Winter [36].

From the above analyses and from the observations of previous researchers [23-25, 36], it was found that the heat treatment of HTC leads to surface modification with increase in adsorption areas. This could be the result of chemical transformation of HTC on heating as observed in the XRD studies. Also, SEM studies have confirmed this effect. Furthermore, samples of HTC already saturated with CO_2 and subsequently heat treated showed different morphologies with different heat treatment conditions supporting the above observations. Also adsorption of CO_2 on HTC have also shown changes in morphology after adsorption showing that both heat treatment as well as adsorption lead to changes in morphology and chemical structures thereby leading to changes in available areas for adsorption of CO_2.

3.3 Analysis of Rates of Adsorption of CO_2 towards Hydrotalcite Pellets

Figure 13 shows HTC pellet with 20mm diameter has higher rate of CO_2 adsorption compared to HTC pellet with 8mm diameter. High exposed area of larger diameter of pellets could contribute to high mass transfer rates of gas on the surface of the pellets. Thus resulting high exposure areas that could tend to increase the rate of adsorption of CO_2.

A close examination of the morphology of HTC after adsorption of CO_2 has shown changes in the structure and morphology which were somewhat prominent as observed using SEM. Disintegrated or deformed particles were observed in morphologies of HTC samples after adsorption of CO_2 at all temperatures were examined. This behaviour is found to be common because of the HTC structures consist of layered double hydroxides where positively charged (cations) hydroxide layer (brucite sheet) and charged balancing anions exist with interlamellar space. When CO_2 is adsorbed on HTC, the CO_2 seems to break down the layered structures leading to changes in the morphology. This shows that with the progression of adsorption of CO_2, the morphology, the structure and the area could change leading to changes in available equivalent adsorption area for CO_2. This could happen when CO_2 is adsorbed and particles disintegrate and exposed further in the internal surfaces facilitating adsorption. This aspect has not been incorporated in any models studied earlier.

Any effect of modifications of area generated due to effects such disintegration and/or agglomeration is worthwhile to be investigated in the form of a rate model for adsorption. Therefore, an extended model of Langmuir was investigated based on the generation or exposure of new surfaces on adsorption. Figure 14 shows the plot of extended model of Langmuir at temperature of 32ºC for HTC pellets. The correlation coefficients obtained (R^2) for the HTC pellets 8mm and 20mm in diameter were observed to be 0.81 and 0.92 respectively. This shows that the experimental rates of adsorption of CO_2 satisfactorily fit the extended model.

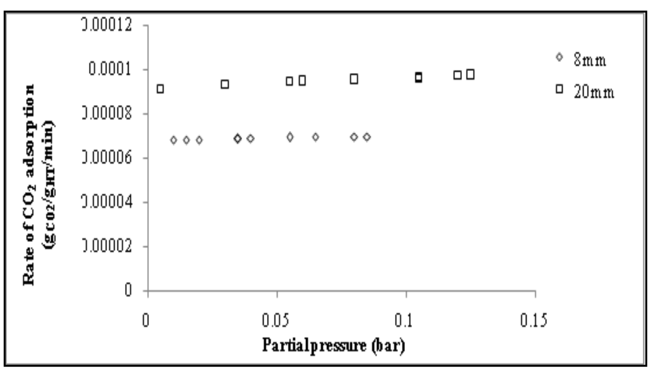


Figure 13 Plots of rates of adsorption versus partial pressure of CO_2 for pelletized HTC (Tomita AD500) at temperature of reaction of 32ºC for 8mm and 20mm diameter pellets

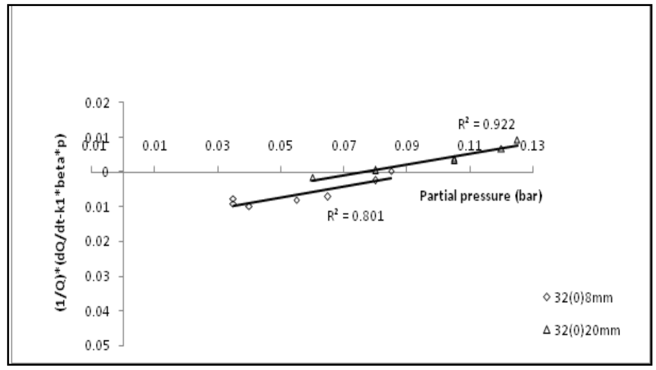


Figure 14 Plots of extended model of Langmuir at temperature of reaction of 32ºC for 8mm and 20mm diameter HTC (Tomita AD500) pellets

4.0 CONCLUSION

HTC has been identified as a good adsorbent for the adsorption of CO_2 because of its stability at high temperature and because of formation of large surface areas conducive to adsorption of CO_2. HTC (Tomita AD500), manufactured by Tomita Pharmaceuticals, Japan were characterized using BET, XRD and SEM analyses in order to investigate the adsorption of CO_2. It was found that HTC (Tomita AD500) has lower variation with only 7% among the values of BET surface area discovered from previous findings. According to XRD analysis, HTC components were observed to have higher compositions at 32ºC which decreased when heat treated to 450ºC. At temperature beyond 450ºC, the components seem to disappear because of the formation of periclase. The changes of HTC structures after subsequent heat treatments were observed according to SEM images. The changes of HTC morphologies and the adsorption of CO_2 have been noticed to lead to increase in surface areas. From the experimental observations, pellets of 20mm diameter used at temperature of 32ºC gave better rates of adsorption compared with smaller size pellets. An extended model of Langmuir which incorporated surface modifications with adsorption was developed based on the Langmuir kinetic principle. The extended model was found to fit the experimental data satisfactorily with good correlation coefficients (R^2) around 0.81 to 0.92.

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