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Experimental Investigations of Physical Adsorption of Carbon Dioxide by Activated Carbon as Adsorbents- A Review

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Abstract

This paper reviews different types of carbonaceous adsorbents used for CO2 capture by physical adsorption. Adsorption is a preferable method due to less energy penalty than absorption process. Paper mainly focuses on carbonaceous adsorbents which include activated carbon (AC), activated carbon fiber's (ACFs) and graphene adsorbent materials. Operating parameters were discussed and experimental investigations for high CO2capture capabilities were stated. Performance parameters such as CO2 uptake, adsorbent surface modification methods, adsorption/regeneration kinetics were discussed. A comprehensive study on carbonaceous adsorbents have been made to update progress in area of CO2 capture. Future research to maximize CO2 capturing using carbonaceous adsorbents is suggested.

Keywords: CO_2 capture; carbonaceous; physical adsorption; activated carbon;

Nomenclature

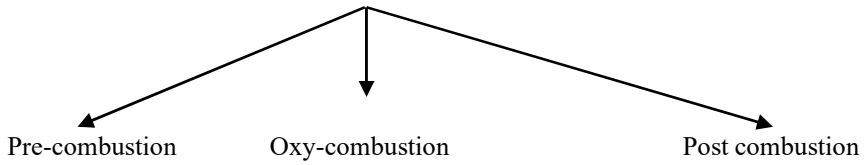
$T_g$feed	Flue gas feed temperature	**acronyms**	
$P_f$feed	flue gas feed temperature	AC	Activated carbon
C_{out}	CO_2concentration at outlet	BET	Brunauer-Emmett-Teller
C_{feed}	CO_2 concentration in feed stream	NAOH	Sodium hydroxide
mmole	millimole	PSA	Pressure swing adsorption
g	grams	TSA	Temperature swing adsorption
KJ/mole	kilojoule per mole	ACFs	Activated carbon fibers
mg	milligram	CNT	Carbon nano tube
lpm	liters per minute	GO	Graphene oxide

1. Introduction

Global warming is the result of increasing greenhousegases, CO2is the major contributor, level in the environment making it a very serious issue for the environmentalist [1][2] (Mondal, Balsora et al. 2012, Lee and Park 2015). Usage of fossil fuels is the influencing source to satisfy energy demand all over the world. Fossil fuel used for energy generation contributes

approximately 40% of CO_2 emissions [3] (Yang, Xu et al. 2008). Use of fossil fuelsmay go for several coming years more even if several other ecofriendly energy producing technologies are emerging. This scenario has created necessity to develop technologies which will be efficient and economical to capture CO_2 gas from the environment[4](Rubin and De Coninck 2005). CO_2 is the major contributor ,about 64%,to enhance greenhouse effect[5](Change 1990).Due to concern of greenhouse effect itis need of an hour to develop technologies for reducing level of CO_2in environment for which carbon capture and storage(CCS) is the main technology to reduce CO_2level. This paper briefly reviews the experimental investigations for CO_2 capture using carbonaceous materials used as adsorbents and focuses on providing a current understanding of the experimental work done so far w ith the use of carbonaceous adsorbent materials.CO_2 capture technology mainly includes physical and chemical absorption,adsorption,cryogenic and membrane process[1](Mondal, Balsora et al. 2012).

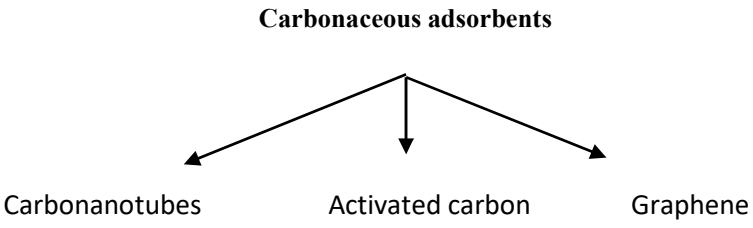
Classification of carbon capture technologies[6](Herzog 2009)



2.Carbonaceous adsorbents

Using carbonaceous adsorbents has various benefits likehigh chemical and physical stability, good heat transfer, physical strength and bio-affinities [7][8][9](Pinto,Goncalves et al. 2013, Rondeau-Gagné and Morin 2014, Zhang, Aboagye et al. 2014). These adsorbents are very effective for adsorbing gases, they are light weight as wellas have more specific surface area and porosity [10][11] (Lee and Park 2010, Gupta and Saleh 2013). Carbonaceous adsorbents have varieties like activated carbon having good porosity ,molecular carbon sieve, carbon nano tubes and graphene. . They are very economical to procure and can be made available from natural resources[12](Abd, Naji et al. 2020).

3.Physical adsorption by using carbonaceous materials

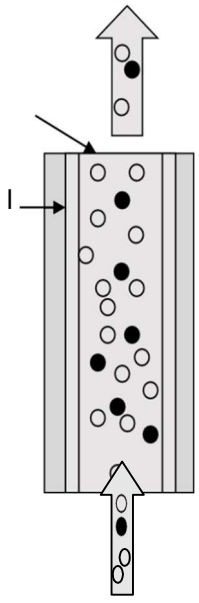


From several reviews on CO_2 capture by adsorption indicates that amount of CO_2 may be reduced from source like flue gases by using carbonaceous adsorbents such as activated carbon, carbon nanotubes, grapheme [13][12](Songolzadeh, Ravanchi et al. 2012, Abd, Naji et al. 2020).Physicaladsorption is advantageous as it is easy for regeneration /desorption of adsorbent material by pressure swing adsorption(PSA) or temperature swing adsorption(TSA)[13](Songolzadeh, Ravanchi et al. 2012), making it an economical process due reduced energy requirement for post combustion carbon capture due to absence of bonding between the sorbate and sorbent[14][13](Ben-Mansour,Habibetal. 2016)(Songolzadeh, Ravanchi et al. 2012).On the other side some adsorbents like activated carbon have low carbondioxide(CO_2) and nitrogen (N_2) selectivity[15](Li, Ma et al. 2011). In most of the experiments carried out for CO_2 capture selective separation of CO_2molecules on gas solid influence is involved[16](Balsamo, Budinova et al. 2013).Development of suitable adsorbent material with high affinity towards CO_2 molecules is a very important in investigating the better resultson CO_2adsorption[14](Ben-Mansour, Habib et al. 2016).

Various laboratory scale experiments conducted on physical adsorption by researchers by using a stream of feed gases(mostly mixture of CO_2 and N_2). As a result of intermolecular attraction between feed gases and adsorbents physical adsorption takes place. This physical adsorption depends uponvarious influencing operating parameters such as temperature, partial pressure, porosity of adsorbing material[17](Meisen and Shuai 1997).

During laboratory scale experiments, column is filled withadsorbents and a mixture of gases (comprising mostly $CO_2$and N_2) is passed through this column. Due to affinity towards CO_2molecules, it gets adhere on surface of adsorbing material till surface area of adsorbents gets saturated i.e no surface areais available for deposition of CO_2gas molecules. Further regeneration process of adsorbent has to be carried out for further cycles of experiment. Fig 1 shows adsorption mechanism.

1. Fig 1-Schematics of adsorption carbon capture process in a cylindrical bed[14](Ben-Mansour, Habib et al. 2016)



4. Carbonaceous materials for physical adsorption

Overview: Due to easy availability, cheap in cost and good thermal stability it is established that carbonaceous materials are preferred over available adsorbents for CO_2 adsorption. These are carbon based material having better properties like affinity towards carbon dioxide molecules, chemical and thermal stability, heat conduction and strength[18][19](Lozano-Castelló , Cazorla-Amorós et al.2002, Bilalis, Katsigiannopoulos et al. 2014). Various forms of carbonaceous adsorbents are available cheaply like porous activated carbon, charcoal coal, molecular carbon sieve, carbon nano tubes and graphene having high specific surface area, high porosity and light weight. Due to its low cost, high

surface area, wide availability, easy surface fictionalizationand easy surface desorption process these carbonaceous material can be used as CO_2 adsorbents[20](Younas, Sohail et al. 2016). Activated carbon and charcoal, due to its high porosity, is capable of capturing CO_2 up to 10 to 15% by weight. Therefore it is suitable in the case when CO_2 purity is below 90% because of low selectivity towards CO_2/N_2[21][22](Radosz, Hu et al. 2008, Levesque and Lamari2009).

Physical adsorption and chemical adsorption are the main CO_2 adsorption processes. Physical adsorption is the common phenomenon and due to weak vander waals forces , binding force between adsorbent and adsorbate can be broken by thermal means[23](Su, Lu et al. 2009). Rise in temperature above 50^0C decreases CO_2 adsorption capacity significantly. Therefore ambient temperature is more suitable for adsorption [24](de Andrés, Orjales et al. 2013).Adsorption capacity is important parameter in determining quantity of absorbent material required and deciding dimensions of adsorption column of experimental set up . Better efficiency of adsorption results in reduced adsorbent requirement and compact experimental set up. Process of adsorption is considered as best technology , when adsorption attains a CO_2loading within range of 3-4mmol/g of adsorbing material[25](Gray, Champagne et al. 2008). Selectivity and adsorption/desorption kinetics are also a important factors responsible for achieving more efficiency of adsorption by adsorbents. CO_2 adsorption kinetics mass transfer through the surface of adsorbent depends upon porosity of adsorbent as well as functional group on the surface of adsorbent material[12](Abd, Naji et al. 2020). Another important and necessary criterion for cheap operational cost is the regeneration/desorption capacity of adsorbent for which heat of adsorption for energy requirement calculation is required. In case of physical adsorption heat of adsorption ranges from -25 to -50 kJ/mol and in chemical adsorption it is -60 to -90 kJ/mol[26](Samanta, Zhao et al. 2012).

5.Experimental investigations on activated carbon (AC) asadsorbent material-

Activated carbon (AC) is suitable because of its capability of giving desired performance at atmospheric pressure and ambient temperature. Due to activated carbon (AC) particles characteristics like large specific surface area and small low- volume pores it has been widely accepted as potential adsorbent. Activated carbon (AC) is hydrophobic in naturedue to which moisture removal process can be avoided before lab scale experimentation on adsorption. Activated carbon (AC) has great affinity towards CO_2 molecules and ease in regeneration of adsorbent at a temperature lower than 373 K. Activated carbon (AC) has high resistivity to acidic and alkali conditions, high porosity, enhance adsorption process and gas diffusion as well. Activated carbon (AC) are easy to modify in its surface chemistry and physiochemical characteristics through modification methods[27](Mukherjee, Okolie et al. 2019). Some

reviewed experimental work by using activated carbon is tabulated in table 1.

(Raganati, Ammendola et al. 2014) conducted CO_2 adsorption experiments with sound assisted fluidized bed using activated carbon (AC) as adsorbent material. An activated carbon DARGO FGD (NORIT) has been investigated at ambient pressure and temperature. Operating conditions of the adsorption experiment like fluidization velocity(0.1,0.25,0.5,0.75,1,1.5,2 cm/s), CO_2 inlet concentration (5,10,15 % vol in N_2), sound pressure level (120,125,135,140 dB) and frequency (20,50,80,120,300 Hz) were varied to test adsorption capacity. Pressure drops and bed expansion curveswere analyzed with application of sound and without sound conditions along with optimum range of frequency for high fluidization.

Absorption test were performed with and without sound assistance to evaluate effect of sound pressure level adsorption capacity forCO_2. Comparative analysis is done in terms of breakthrough curves, moles of CO_2 adsorbed (n_{ads}),breakthrough time (t_b), the fraction of bed utilized at breakpoint(W)[28].

(Raganati, Chirone et al. 2020)in further research work tested activated (AC) carbon as CO_2adsorbent, with and without sound assistance fluidization. Study on sound assisted fluidized bed temperature swing adsorption cycle by modifying through heating and purging strategy were carried out. Adsorption/desorption temperature (25^0-150^0C) and CO_2 partial pressure (0.05-0.2 atm) were experimented and its effects was evaluated. It was observed that by application of heating and purging strategy to a fluidization by sound assistance results in recovery of CO_2up to 80%.This research work concluded that working capacity of CO_2 during temperature swing adsorption processes is affected by operating temperature and partial pressure of CO_2 [29].

(Huang, Cheng et al. 2015)prepared a high-quality activated carbon (AC) from coconut shell. Experimental investigations were done by varying parameters like activation temperature and time to study basic characteristics of activated carbon suchas charcoal yielding, ash, Brunauer-Emmett-Teller specific surface area, pore volume. Increase in activation temperature and time increases the ash content and reduced ash content respectively. Activation done at 100^0C for 120 minutes gains highest BET specific surface area (824 m^2/g) and total pore volume (0.502 ml/g). Analysis shows that coconut shell activated carbon has more adsorption capacity in comparison to commercial activated carbon [30].

(Karbalaei Mohammad, Ghaemi et al. 2020) in his research work make use of activated carbon modified by using hydroxide solution (concentration range of 10-40%) for CO_2 adsorption. For experimentation temperature range of 20^0- 80^0C and pressure range between 2 bars -10 bars were kept forinvestigating kinetics, isotherms and CO_2 adsorption thermodynamics involved. To see the optimal results of CO_2 pressure and temperature on CO_2 adsorption capacity, response surface methodology technique is applied. Optimum results on adsorption capacity (104.32 mg/g) were obtained for30% NAOH concentration at 20^0 temperature and 6 bars. The sips model for CO_2 adsorption isotherms is found to be best. Kinetic study shows that pseudo-second-order model best fitted for experimental data. Thermodynamic parametersindicates that CO_2 adsorption process is exothermic in nature [31].

(Al Mesfer, Danish et al.) experimented CO_2 adsorption process by using activated carbon as adsorbent material ranging from 0.4 cm to 0.8 cm in size. Best results wereobtained at on a set feed rate of $6.67x10^{-5}m^3$/sec and with a carbon dioxide concentration of 0.05 (wt.fraction). Findingobserved that breakthrough times depends upon temperature and feed rate. Maximal breakthrough time and saturation timesof 870sec and 1050 sec were observed. Results were compared with silica gel as adsorbent material. Breakthrough time and saturation times have been compared between activated carbon and silica gel. With increased feed rates ($8.33x\ 10^{-5}$ m^3/sec), maximum capacity of 39.14 g CO_2/kg were achieved. It was found that due to greater sorption capacityand better characteristics parameters activated carbon is more economical option forCO_2/N_2 gases mixture [32].

(Das, Behera et al. 2019) developed four stage counters current multistage fluidized bed adsorber to capture CO_2 from flue gas. Monoethanoamine impregnated activated carbon (coconut shell based) used as adsorbent and optimum operating parameters were described which are responsible for CO_2 removal efficiency. Optimization using response surface methodology along with central composite design(CCD) isused to find out relation between three operating variables(CO_2 inlet concentration ranging from 3000ppm to 20000ppm,impregnation ration ranging from 0.2 to 0.6 and weir/bed height in range 20mm to 60mm) with four stages of fluidized bed column (0.21 m height per stage and 0.095m internal diameter) were fixed together with flange joint. Regression model equation is developed through design expert software. Results after optimization found to be for CO_2 concentration (7312ppm),impregnation ratio (0.31), weir/bed height of 48.65mm and CO_2 removal efficiency of 95.17%.With same operating condition experimental CO_2 removal efficiency was found to be 95.97%,thus found to be in agreement with predicted operating conditions [33].

(Al-Ghurabi, Ajbar et al. 2018) conducted experiment considering four cases of mixing in different proportions of external group B particles in fluidized bed containing activated carbon(500g).The amount of external group B particles added were in proportion of free weight basis 0,5,10 and 30% weight. Results of fluidization hydrodynamics due toaddition of external particles were studied and shows better quality of fluidization, thus better CO_2 removalefficiency.10% by weight showed better results over 5% and 30% by weight mixing. On the other hand, poor quality of fluidization was observed with 5% by weight sand mixing [34].

(Dantas, Amorim et al. 2009) used commercial activated carbon and nitrogen enriched activated carbon by mixing 10g of activated carbon in 500mL of 10^{-1} M 3-chloropropylamine hydrochloride solution as adsorbent material. Some chemical and physical factors were identified that influence CO_2 adsorption capacity. His work concluded that nitrogen enrichment blocked some pores of activated carbon due to which only the large pores is filled by CO_2 resulting in decreased CO_2 adsorption and increased adsorption rate [35].

(Casas, Schell et al. 2012)performed experimentation in a fixed bed packed with adsorbent (commercial activated carbon) with mixture of CO_2/H_2gas as feed mixture. Different temperature (25^0C,45^0C,65^0C and 100^0C) and pressures (1bar,5 bar,10 bar,15 bar,20 bar,25 bar,35 bar) were set for conducting experiments. Adsorption phenomenon ofCO_2/H_2mixture

were analyzed focusing on number of important features having impact on capacity of adsorption in continuous process(PSAor TSA) for separating CO_2 and H_2.Mathematical model has been developed for checking accuracy level depending on operating conditions. Research work done consider it as better, as such no adjustment was done on the adsorption isotherms after measuring them in a independent manner in a gravimetric set-up [36].

(Rashidi, Yusup et al. 2014) developed economical and efficient activated carbon (coconut based) with single stage activation process making it porous. Due to predominantly micro porosity activated carbon help in CO_2 gas adsorption. Experiment was carried out at temperatures (25^0C, 50^0C, 100^0C) using adsorbate purified carbon dioxide and mixture ofbinary gases. Experimental result shows that due to less surface area and micro pore volume, lowerCO_2 adsorptioncapacity on optimized activated carbon occurs. Lower adsorption capacity attributes to reduced CO_2 partial pressure in case of mixture of gases. Kinetic studies were carried out for purified CO_2 adsorption by activated carbon and it results in dominant physisorption mechanism indicating the magnitude of activation energy lesser than 25kJ mol^{-1}. Further concluded that gas adsorptive behavior gets reduced at high temperatures [37].

(Danish, Parthasarthy et al. 2021) carried adsorption test usingsilica gel and date pits-based activated carbon synthesizedusing tubular furnace by physical activation. Rise in temperature is kept at 10^0C/min carbonization of biomass andN_2 flow is monitored continuously for 2h around 600^0C.Important parameters like dependence of temperature, feed flow and initial CO_2 level on the breakthrough and exhaustion periods were varied with operating parameters. Comparison between carbon produced from date and silica gel clearly shows that exhaustion and breakthrough spans were longer for carbon produced from dates. Maximal CO_2 adsorption of 73.1 mg/g was achieved at 5 lpm of feed gas at temperature of 298 K andCO_2 proportion of 5% in feed gases.[38]

(Ibrahim and Al-Meshragi 2019) conductedCO_2 adsorption test on activated carbon produced from olive trees in fixed bed apparatus. In order to investigate kinetic and thermodynamic parameters, adsorption and breakthrough curves weredetermined at various temperatures (30^0C, 50^0C, 70^0C and90^0C). Experimental result shows that the CO_2 adsorption on olive tree activated carbon depicts physisorption behavior and adsorption capacity decreases as temperature increases. MaximumCO_2 sorption capacity on activated carbon ranged from 109.5 mg CO_2 /g to 35.46mg CO_2 /g and from 129.65mg CO_2 /g to 35.55 mg CO_2 /g. of activated carbon for initial concentrations of 10% and 13.725 % vol respectively.[39]

(Al-Janabi, Vakili et al. 2018) conducted combined experimental and numerical study ofCO_2/N_2 adsorption in fixed bed with three different adsorbents one of which is bulk activated carbon. Effect of adsorbable gas (CO_2) inlet concentration on CO_2breakthrough curves were studied by varying N_2 flow rate based on a constant CO_2 flow rate of 5cm^3/min to avoid fast saturation of adsorption bed by CO_2. Experiment with different adsorbents was conducted at constant temperature of 50^0C. His work further concluded that percentage shift in breakthrough curve is practically independent of adsorbent used or total flow rate. His work is helpful in predicting the performance of materials under flow conditions.[40]

(Zulkurnai, Ali et al. 2017) prepared activated carbon using sea mango functionalized with

deep eutectic solvent DES which is a compose of choline and glycerol. Impregnationwith DES ratio for preparation of activated carbon was kept at 1:2 precursor -to- activate ratio. The results show optimum CO2 adsorption capacity of the activated carbon by using CO2 gas treatment method (9.851 mgCO2/gsol), followed by the absence of gases (9.685 mgCO2/gsol), steam (9.636 mgCO2/gsol), and N2 (9.536 mgCO2/gsol).Experimental investigation reveals about higher CO2 adsorption capacity of DES- functionalized activated carbon than non-functionalized activated carbon despite reduction in surface area of functionalized activated carbon about 50% than non- functionalized activated carbon. Elemental N on the surface of the activated carbon plays a significant role in CO2 adsorption.[41]

(Boonpoke, Chiarakorn et al. 2012) synthesizes collected sugar bagasse, washed to remove dust and oven dried(378Kfor 24 h) to make activated carbon. Activated carbon wasactivated by ZnCl2 maintaining weight ratio of bagasse toZnCl2 of 1:1 at ambient temperature for 1h.Surface modification was done on a bagasse based activated carbon bythree different amines:Polyethlenimine(PEI),monoethylenimine(MEI) andaniline (ANL) with loading ratio of 5%,25% and 50% of weight. Desirable characteristics like high carbon content, high surface area and porosity were obtained on this synthesized activated carbon. High CO2 adsorption efficiency was observed decreasing with rise in temperature but keeping temperature above 323K and a concentration of CO2lowerthan 30% v/v., bagasse based activated carbon modified with PEI at 5 and 25% weight results in higher adsorption capacity.[42]

(Dantas, Luna et al. 2011) conducted study on fixed bed adsorption of carbon dioxide with CO2/N2mixture on activatedcarbon (NORIT R2030). Experimentation was done at various temperatures and adsorption dynamics were analyzed for different effect caused by N2 adsorption and desorption. Breakthrough curved were obtained at these temperature (301K-306K,323K,373K and 423K) with CO2/N2mixtures. Study of adsorption kinetics of CO2and N2were carried out by fitting linear driving force (LDF) based model taking intoaccount energy and momentum balance.[43]

Source of activated carbon	Surface area (m^2/g)	Pore volume (cm^3/g)	Pore diameter		Gas composition	Temperature	Ref
DARGO FGD (NORIT	1060	-	<2nm	0.31(mmol/g) ordinary condition 0.37(mmol/g) (sound assited)	5% CO_2 in N_2 10% CO_2 in N_2 15% CO_2 in N_2	Ambient	28
Activated carbon (coconut shell)	824	0.502	30 mesh	14.40 mg/L	100% CO_2	Ambient	30
Activated carbon modified by sodium hydroxide solution	483.91	0.2744	2.2678nm	104.319 mg/g	100% CO_2	20^0C -80^0C	31
Commercial activated carbon	858	0.425	0.4 -0.8 cm	39.14 g CO_2/Kg sorbent	0.05 (vol. %)	298K	32
Amine impregnated activated carbon	572.27	0.259	100 mesh	-	3000 ppm to 20,000 ppm	Ambient	33
Activated carbon with mixed external particle of Geldart group B classification	941.51	0.22973	1.7–300.0 nm	Better with mixed external particle of Geldart group B classification	0–2 SLPM for N_2 0–200 CCPM for CO_2	Ambient	34
Nitrogen-Enriched Activated Carbon	1053.0	0.0972	0.0038 m		10% and 20% CO_2 in helium	301K,323 K, 373 K, and 423K.	35
Commercial activated carbon	8.5 × 108 [m2/m3] Specific surface area	-	0.003m	higher with lowering temperature	CO_2/H_2 mixtures (25%/75%, 50%/50% and 75%/25%)	25 °C, 45 °C, 65 °C and 100	36
Activated carbon(agriculture residue)	370.71(optimized) 717.22 (commercial)	0.15 optimized 0.29 commercial	1.63nm (optimized) 1.64nm (commercial)	1.79 mmol /g (optimized)	20 % CO_2 and 80 % nitrogen (N_2)	25 °C, 50 °C, 100 °C	37
Activated carbon (date pits based)	848.3	0.45	2.26nm	73.1 mg/g	CO_2/N_2	20 °C, 25 °C, 40 °C	38
Activated carbon(from olive trees)	602	0.61	5mm	109.5 to 35.46 and from 129.65 to 35.55 mg CO_2/g	10 and 13.725% vol.,	30°C, 50°C, 70°C, and 90°C	39
Bulk Activated carbon(sigma Aldrich)	-	-	0.15mm	0.242mmol/g	15–25% CO_2 in CO_2/N_2 mixtures	50^0C	40
Activated carbon(sea mango based)	882.71	0.4696	-	by using CO_2 gas treatment method (9.851 mgCO_2/gsol),	15% CO_2 in N_2	ambient	41

Bagasse-based activated carbon	923.39	0.5330	2.01nm	0.20 mmol/g	<30% CO_2 v/v	323K	42
Commercial activated carbon (NORIT R2030)	1053 m2 /g	-	0.0038 m	-	10% CO_2/N_2 v/v	301–306, 323, 373 and 423 K	43

Table 1- **Reviewed physically and chemically activated carbon dioxide adsorption**

6.Conclusion

Physical adsorption of carbon dioxide using carbonaceous adsorbent is efficient due to less energy requirement for desorption process as compared to other capturing technologies. This paper concludes about state of knowledge about physical adsorption by carbonaceous adsorbents for CO_2 capture. Modification by functionalization of adsorbent material enhances CO_2 adsorption process. Other activated carbon materials like activated carbon fibres and graphene due to high porosity are promising for CO_2 capture.Properties like high thermal stability, mechanical strength, high CO_2 selectivity, low preparation cost of adsorbent material and high resistance to moisture are the main criteria for comparison with other adsorbent materials. Compared with other materials carbonaceous material possess high stability and are cheaper.Influential operating parameters like temperature and pressure has greater effect on physical adsorption. With the rise in temperature,adsorption capacity gets reduced. To overcome this difficulty efforts are required towards modification of carbonaceous material by suitable activation of adsorbent material. Research regarding preparation of low cost, sustainable and easy to regenerate/recycle carbonaceous material should be carried out. Eco friendly raw material for producing adsorbent should be focused. There is scope of research for CO_2 capture by using ACF and graphene.

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