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Carbon Dioxide Removal by Adsorption

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Abstract: Carbon dioxide (CO_2) among other air pollutants is a major culprit to the greenhouse gases that is fueling global warming. To mitigate global warming, Kyoto Protocal urges 37 industrialized nations and European Union to reduce their greenhouse gas emissions to a level of 5.2% on average lower than those of 1990 during the period of 2008-2012. It is therefore essential to develop the CCS technologies to cope with the global demand of CO_2 reduction. In this study the technologies of CO_2 removal are reviewed.

Key words: Carbon capture, adsorption, waste rubber, activated carbon, chemical activation

INTRODUCTION

Increasing CO_2 emissions in environment leads to global warming which is an issue of great concern today. The climate of the earth is varying continuously due to various factors, for example: Change in the Earth's orbit, change in the Sun's intensity, change in ocean currents, volcanic emissions and increase in greenhouse gas (GHG) concentrations. The greenhouse effect is the phenomenon where water vapor, carbon dioxide (CO_2), methane and other atmospheric gases absorb outgoing infrared radiation causing an increase of Earth's temperature. Excessive greenhouse gases in the atmosphere are responsible for various environmental problems like continuous rise of water-level in sea, the increasing number of ocean storms, floods and others (Lu *et al.*, 2009).

Carbon dioxide (CO_2) among other air pollutants is a major culprit to the greenhouse gases that is fueling global warming. Among the greenhouse gases, CO_2 contributes more than 60% to global warming because of its huge emission (Albo *et al.*, 2010).

The fact has inevitably aroused numerous public concerns on excessive and uncontrolled CO_2 emission with respect to its impact on climate and environment change. However, the endeavor to offset its increasing trend is becoming even more challenging. Unless taking effective and proper measures, according to the prediction of Intergovernmental Panel on Climate Change (IPCC) by the year 2100, the atmosphere may contain upto 570 ppmv of CO_2, causing a rise of mean global temperature of around 1.9°C and an increase in mean sea level of 3.8 m. Therefore, the agenda on globe wide cooperation in carbon dioxide emission reduction and mitigation has been explicitly put forward from a standpoint of long term sustainable development (Dinda, 2013).

Copenhagen Accord also requests the global temperature increase be limited to 2°C above the pre-industrial level by 2100. International Energy Agency (IEA) pointed out to achieve the ±2°C goal, CO_2 capture and storage (CCS) technology is required and the contribution would be 19% in 2050. It is therefore essential to develop the CCS technologies to cope with the global demand of CO_2 reduction (Yu *et al.*, 2012).

CARBON CAPTURE AND SEQUESTRATION (CCS)

Carbon Capture and Sequestration (CCS) is widely acknowledged and identified to be the major and crucial approach to limit the CO_2 level in the realm of carbon reduction and its derived technologies. The CCS encompasses the removal and sequestration of CO_2 from industrial low CO_2 level containing streams, the generation of high concentrated CO_2 stream and the transport and storage of compressed CO_2 (Zhao, 2012).

The CCS is an integrated process involving three stages. Firstly, CO_2 is captured from power stations and other large industrial sources. After that, CO_2 is transported to a storage site. Then, CO_2 is stored permanently in deep geological features. The CCS encompasses the removal and sequestration of CO_2 from industrial low CO_2 level containing streams, the generation of high concentrated of CO_2 stream and the

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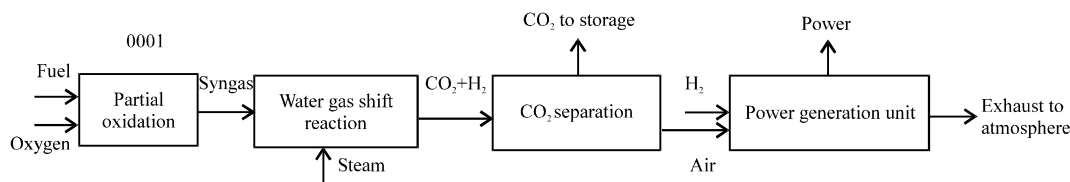Universiti Malaysia Sabah, Kota Kinabalu, 88900, Sabah, Malaysia

Fig. 1: Principle of pre-combustion CO_2 capture (Mondal *et al.*, 2012)

transport and storage of compressed CO_2. Currently, three types of CCS processes for scrubbing exist: Pre-combustion, oxyfuel combustion and post-combustion (Coninck *et al.*, 2009).

Pre combustion process is related to the treatment of synthesis gas (syngas) composed principally of CO and hydrogen. Pre-combustion rate is premised on the production of syngas, removal of CO_2 and combustion of H_2. In water gas shift reaction, the addition of steam and reduction of temperature actually promote the conversion of CO into CO_2 and increase the yield of H_2. From CO_2 and H_2 bearing stream, CO_2 is separated and sent to the compression unit and pure H_2 is further used as an input to a combined cycle to produce electricity (Gibbins and Chalmers, 2008) (Fig. 1).

The technology required for pre-combustion capture is widely applied in fertilizer manufacturing and in hydrogen production. However, the main disadvantage of pre-combustion capture is that it requires a chemical plant in front of the turbine. Complicated chemical processes normally cause extra shut-downs of the plant which can result to a lower power output. Other disadvantages are non-gaseous feed stocks, requirement of the cleaned gas stream and possible expensive scrubbing for high NO_x emission control. Although the initial fuel conversion steps of pre-combustion are more elaborate and costly, the higher concentrations of CO_2 in the gas stream and higher pressure make the separation easier (Kanniche *et al.*, 2010).

It needs high purify oxygen to combust with fuels instead of air. This result in high CO_2 concentrations in the gas stream and, hence, in easier separation of CO_2 and in increased energy requirement in the separation of oxygen from air. Furthermore, material of construction to withstand at high temperature is needed and recycling of flue gases increases the cost of the process (Kanniche *et al.*, 2010).

Post-combustion is used to sequester CO_2 from flue gases produced by the combustion of a primary fuel. Existing power plants use air for combustion and generate a flue gas at atmospheric pressure and typically have a CO_2 concentration of less than 15%. Technologies based on chemical absorption appear to be best adapted to this separation (Kanniche *et al.*, 2010).

Post combustion capture technology has a higher thermal efficiency than pre combustion. Understanding of each capture system or process is great significant in selecting the appropriate approach. Post-combustion capture of CO_2 in power plants is economically feasible for existing power plants because retrofitting existing plants with post-combustion CO_2 capture is expected to be easily conducted and have a relative less cost (Zhao, 2012).

SEARCHING FOR LOW COST ADSORBENTS

Various materials such as zeolites and activated carbons have been investigated as adsorbents for the CO_2 separation and capture by selective adsorption. Among these materials, activated carbon is the most commonly used and most effective modified adsorbent support because of its high specific surface area and big pore volume (Zhao, 2003).

However, the cost of activated carbon is relatively high which limits its application. Researchers are now finding other adsorbents as an alternative to the standard activated carbons which have lower cost and similar efficiency. Natural materials or the waste/by products of industries or synthetically prepared materials which cost less and can be used as such or after some minor treatment as adsorbents are generally called low-cost adsorbents (Mukhlish *et al.*, 2012).

Thus, activated carbon derived from waste tires using a chemical activation method is used as an alternative adsorbent. Activated carbon prepared from rubber tire has been used widely in the adsorption process (Mui *et al.*, 2004).

ACTIVATED CARBON FROM WASTE RUBBER TIRE AS ALTERNATIVE ADSORBENT

Generally tires contain vulcanized rubber plus with the rubberized fabric with reinforced textile cords, steel or fabric belts and steel wire beads (Ko *et al.*, 2004). However, the most commonly used rubber tire is styrene-butadiene copolymer (SBR) that contain 25% styrene. Other rubbers that can be used in tire manufacture include natural rubber, synthetic cis-polyisoprene and cis-polybutadiene. So, rubber tire

usually consists of a mixture of different elastomers such as natural rubber, butadiene rubber and styrene butadiene rubber mixed with other addictives like carbon black, sulfur and zinc chloride (Gupta *et al.*, 2011).

Carbon black usually added to strengthen the rubber and aid abrasion resistance. In addition, extender oil added to soften the rubber for improvement of workability and sulfur added during vulcanization process to cross-link the polymer chains within the rubber and harden and prevent excessive deformation at elevated temperature. Zinc chloride and stearic acid is added to control the vulcanization process and enhance the physical properties of the rubber (Ko *et al.*, 2004). In composition, 32% by weight of the waste tire is mainly constituted of carbon black which contains higher carbon content as 70-75 wt% (Gupta *et al.*, 2011).

However, tires are made of notable durability and almost immune to biological degradation that makes their disposal and reprocessing difficult. Although the waste rubber tires may cause environmental problems such as stockpiling, fire and health hazard but it offers a medium as a cheap source for the preparation of adsorbent materials that can be used in capturing CO_2 from flue gases (Gupta *et al.*, 2011).

CHEMICAL ACTIVATION OF ACTIVATED CARBON

Chemical activation is a single step process that includes the impregnation of the carbonaceous material with dehydrating agent prior to activation (El Qada *et al.*, 2007). Through this chemical activation process, the carbon's surface properties such as carbon structure, pore size and surface area are modified (Quek and Balasubramanian, 2011). The chemical activation involves the reaction of the carbonaceous precursor with chemical activating agents which is later followed by heat treatment stage. Then, in the final stage, the washing step takes place in order to remove the chemical agent and the inorganic reaction products (Linares-Solano *et al.*, 2008).

The impregnation is done purposely to enhance the pore structure of precursor and increases its surface area. The common activating agents used for the chemical activation are potassium hydroxide (KOH), Zinc chloride $(ZnCl_2)$, sodium hydroxide (NaOH), Aluminium chloride $(AlCl_3)$, sulfuric acid and phosphoric acid (El Qada *et al.*, 2007).

The advantages of the chemical activation are single step activation, lower temperatures of activation, low activation time, offer higher yields and a good development of the porous structure. However, the process may involve a complex recovery and recycle of the activating agent that generates liquid discharge and demands of effluent treatment (Yang *et al.*, 2009).

Potassium hydroxide (KOH) was selected as chemical agent in the impregnation procedure because it was found to be more effective than other chemical agents such as $ZnCl_2$ and H_3PO_4 in creating porosity in activated carbons derived from tyres (Amri, 2008). Using activating agent, the impregnation ratio of material to the activating agent is determined. The impregnation with KOH causes degradation of the cellulosic material and on carbonization, dehydration. These processes result in charring and aromatization of the carbon skeleton and creation of the pore structure.

POST COMBUSTION PROCESS

As the name implies, these systems capture CO_2 from the flue gases produced after fossil fuels or other carbonaceous materials (such as biomass) are burned. Combustion-based power plant provides most of the world's electricity today. In a modern coal-fired power plant, Pulverized Coal (PC) is mixed with air and burned in a furnace or boiler. The heat released by combustion generates steam which drives a turbine-generator. The hot combustion gases exiting the boiler mainly consist of nitrogen (from air) plus smaller concentrations of water vapour and CO_2 formed from the hydrogen and carbon in the fuel (Kaithwas *et al.*, 2012).

CO_2 CAPTURE TECHNOLOGIES

There are several technologies available for CO_2 capture such as adsorption, absorption, cryogenic and membrane separation. Adsorption process is the process in which the one or more component of a gas or liquid stream is adsorbed on the surface of a solid adsorbent and a separation is accomplished. The gas is passed through the bed and the solid particles adsorb components from the gas. As the process is an exothermic process, by increasing the temperature, the regeneration of the adsorbents through desorption is able to perform. The advantages of this process are: It required lower energy requirements (Kaithwas *et al.*, 2012), together with operating cost and it is able to remove CO_2 by approximately 90% while the disadvantages included the process configuration is complex (Dinda, 2013).

Absorption process is the process when the two contacting phases are a gas and a liquid and it is based on the reaction between CO_2 and chemical solvent (Mondal *et al.*, 2012). Although the advantages of this process required moderate operating cost and able to handle bulk amount of CO_2 stream together the efficiency

of removing CO_2 approximately 90%, however, corrosion problem and higher energy consumption together with degradation of absorbent are the disadvantages of this absorption process (Dinda, 2013).

Cryogenic separation process commonly used for separation of CO_2 from streams which having high CO_2 concentrations typically more than 50%. The main merits of this process are; no chemical adsorbent is necessary and able to conduct at atmospheric pressure. Limitation of this technologies is problematic to employ for CO_2 capture as CO_2 is develop in conjunction with other gases such as SO_x, NO_x, H_2O which severely interfere with cooling and cause corrosion, fouling and plugging (Mondal *et al.*, 2012).

Membrane separation process, the required energy per unit mass of CO_2 captured is least in the range of 0.5 until 6 MJ kg^{-1} of CO_2 as compared to other existing technologies (Mondal *et al.*, 2012). Besides that, the membrane acts as a semi-permeable barrier and separation occurs by the membrane controlling the rate of movement of various molecules between others phases. The main advantage of this process is that it is highly capable of maintaining product purity even though the capacity is reduced down to 10% of the initial design value. Furthermore, it is also a low cost process for separating gases and extremely reliable with respect to the on-stream factor. Low removal efficiency and low purity of CO_2 make this process inefficient are the limitation of this process (Mondal *et al.*, 2012).

To reduce the amount of CO_2 released into atmosphere, these four main methods are being developed for CO_2 capture and separation (CCS). Among these methods, adsorption with solid adsorbents is one of the most promising options due to its low energy consumption, low equipment costs and easiness for application (Mandal and Bandyopadhay, 2006).

ADSORPTION PROCESS OF CARBON DIOXIDE

Adsorption is a process for removal of one or more components of a mixture with the help of a solid surface. The adsorption processes are based on significant intermolecular forces between gases (including CO_2) and the surfaces of certain solid adsorbents such as waste rubber tire derived activated carbon. Depending on the temperature, pressure and percentage of active loading, single or multiple layers of gases can be adsorbed (Meisen and Shuai, 1997).

In CO_2 capture by adsorption technology, a packed column is mainly filled by spherical adsorbent and CO_2 bearing stream is passed through the column. Carbon dioxide is attracted towards the adsorbent and adheres on the surface of adsorbent. After achieving the equilibrium, desorption to get CO_2 in pure form and regenerated adsorbent can be performed by rising the temperature since this adsorption is an exothermic process. The regenerated adsorbent can be utilized for further cycle. Solid sorbents have the potential for significant energy savings over liquid solvents, in part because they avoid the need for the large quantities of water that must be repeatedly heated and cooled to regenerate the solvent solution. However, adsorption presents lower energy requirements and avoids the shortcomings when compared to absorption (Drage *et al.*, 2009). In post-combustion process, adsorption is recognized to be an attractive process for CO_2 capture from flue gases, due to its lower energy requirements (Aaron and Tsouris, 2005).

CRITERIA FOR SCREENING SORBENTS

A good solid sorbent material for CO_2 removal from flue gas should combine several attributes including high CO_2 adsorption capacity, high CO selectivity, mild regeneration condition and sufficient cyclic stability and mechanical strength (Zhao, 2012).

Adsorption of carbon dioxide by waste rubber tire derived activated carbon was a newly established research. This type of research has not been carried by other scientists. This alternative adsorbent had been tested before for its ability for the adsorption of effluent pollutants such as phenol and mercury, remove methylene blue from aqueous solution and also in gas separation of O_2 and N_2 (Li *et al.*, 2010).

Apart from waste, rubber tire derived activated carbon, many researches had also been carried out before using other materials as alternative adsorbents. Another material that had been tested for its ability to become CO_2 adsorbent is the spent coffee grounds. In this study, Spent Coffee Grounds (SCG), a residue from the soluble coffee industry, was evaluated as an adsorbent for the removal of CO_2 from flue gas. It undergoes chemical activation with CO_2 NCHA29 and NCLK3 adsorption test was measured between 0-120 kPa at 0, 25 and 50°C. The result shows that as pressure increases, adsorption capacity increases. The highest adsorption capacity occurs at lower temperature (Gonzalez *et al.*, 2008).

Based on a previous study of Dreisbach *et al.* (1999) conducted adsorption equilibrium of CO_2 on Norit type activated carbons at high pressure upto 6 MPa at 298 K where they found a fairy high pure CO_2 loading upto 11.27 mmol g^{-1}. This has shown that CO_2 adsorption capacity of activated carbon increases as the temperature drops.

Furthermore, recent research carried out has proved that amine impregnated adsorbent such as MCM-41-PEI with a PEI loading of 75 wt% under atmospheric pressure at 75°C has shown the highest CO_2 adsorption capacity of 3.02 mmol g^{-1}. Increased PEI loadings resulted in higher adsorption of CO_2. The adsorption capacity at low temperature will be even larger than that high temperature if the adsorption time is sufficiently long to overcome the diffusion limitation (Xu *et al.*, 2002).

There is a research conducted by Zhao (2012), using amine impregnated sorbent. He used a series of impregnated SBA-15 sorbent containing 50, 60 and 70 wt% active loading of tetraethylenepentamine (TEPA). There is an increasing trend of CO_2 adsorption capacity from 3.59-4.59 mmol g^{-1}. When the amine loading increases from 50-70 wt% (Zhao, 2012).

Besides that, the adsorption capacity increased with increasing the temperature and reached the maximum CO_2 uptake of 4.52 mmol g^{-1} at about 75°C. The low CO_2 uptake at low temperature could be a result of slow adsorption rate because the overall process is diffusion controlled. Therefore, higher temperature is beneficial to significantly accelerate the apparent adsorption rate by reducing diffusional resistance. For the temperature beyond 75°C, it inhibited the CO_2 adsorption on TEPA impregnated SBA-15 due to its exothermal nature. Therefore, this might be due to the thermodynamic limitation that makes shifting in chemical equilibrium beyond 75°C (Zhao, 2012).

On the other hand, Polyaniline-magnetite nanocapsules based nanocomposite (PANI/MNCs) is one of the solid adsorbent used to adsorb CO_2. The PANI/MNCs nanocomposite has shown a maximum adsorption capacity of 54.21 mmol g^{-1} at 14.5 bars and room temperature (28°C). According to BET theory, at high pressure, more gaseous molecules would be available per unit surface area which results in multilayer adsorption (Tamilarasan and Ramaprabhu, 2012).

Besides that, the amount of adsorbed CO_2 molecules decreases with increase in temperature with the use of PANI/MNCs nanocomposite. This may be attributed to the increase in kinetic energy of CO_2 gas molecules at higher temperatures which provides high velocity to CO_2 molecules to escape from surface active sites and pores. At low temperature, CO_2 molecules interact well with adsorbent surfaces which lead to higher multilayer formation of CO2 gas at pores of nanocomposite as well as a fraction of carbonate and bicarbonate formation (Tamilarasan and Ramaprabhu, 2012).

Activated carbons prepared from hydrothermally carbonized (HTC) waste biomass were studied with respect to the adsorption of carbon dioxide. This HTC biomass is then further activated physically or chemically.

Physically Activated Carbon (PAC) was activated into activated carbons by treating the HTC biomass at an elevated temperature in a stream of CO_2 at 800°C for 0 or 2 h while Chemically Activated Carbon (CAC) were produced by first treating HTC beer waste with H_3PO_4 and by further activating this acid treated HTC biomass in a flow of N_2 at 600°C. The HTC biomass was made from four different biomass: Grass cuttings, horse manure, organic waste from beer production (beer waste) and bio-sludge from a waste water treatment plant of a paper and pulp mill. As temperature are fixed at 0°C, the adsorption capacity increases with pressure. The PAC from HTC grass cuttings also displayed an excellent cyclability for the adsorption of CO_2 (Hao *et al.*, 2013).

In addition, previous research has also proved that *Eucalyptus camaldulensis* wood can be utilized as CO_2 adsorbent. *Eucalyptus camaldulensis* wood is a kind of lignocellulosic material which is obtained from a waste dump of a wood factory. It has a reasonably high content of carbon, utilized also as raw material for activated carbon. It undergoes chemical activation with $ZnCl_2$ at different impregnation ratios as well as by pyrolysis. The amount of CO_2 adsorbed onto the AC samples prepared with $ZnCl_2$ and H_3PO_4 increases as the impregnation ratio of the activating agent to biomass decreases or, in other words, the CO_2 adsorbed onto the AC increases when the BET surface area and the mesoporosity of the AC samples increases (Heidari *et al.*, 2014). The CO_2 adsorption capacity increases when the temperature decreases.

Moreover, there are previous studies using mesoporous activated alumina functionalized with diethanolamine (DAAB) as the adsorbent for selective adsorption of carbon dioxide. During the adsorption process, it varied its flow rate from 90-150 mL min^{-1}. An increase in the flow rate (120-150 mL min^{-1}) led to the reduction of the residence time of the adsorbate in the column leading to poor performance of the system or reduction on the adsorption efficiency. This reflected on the adsorption capacities which were 55.94, 39.36 and 27.03 mg g^{-1} for the feed flow rates of 90, 120 and 150 mL min^{-1}, respectively (Auta and Hameed, 2014).

Furthermore, CO_2 capture was conducted at various column temperatures (35, 45 and 55°C). It was observed that lower temperature has short breakthrough time. Moreover, lower temperature of adsorption promoted intense selective interactions between impregnated basic amine group on the activated alumina (DAAB) and the acidic CO_2 gas resulting to higher adsorption capacity. In a similar manner, carbon adsorbent modified with nitrogen compound showed higher adsorption of CO_2 gas at room temperature than at elevated temperatures (Auta and Hameed, 2014).

CONCLUSION

Although there are many types of activated carbon derived from industrial waste such as coconut and impregnated with different chemicals such as Cu/Ce used to adsorb CO_2 but that the research using waste rubber tire derived activated carbon has not been conducted before by any researchers in capturing CO_2 although some researchers used this type of adsorbent to remove methylene blu, mercury and O_2/N_2 gas separation.

Besides, there were few studies have been carried out before that had investigated the effects of all three parameters-adsorption temperature, adsorption flow rate and impregnation ratio on the adsorption efficiency capacity of CO_2. Literature on CO adsorption by activated carbon from waste rubber tire via chemical activation of KOH is not available.

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