

## **UNIVERSITI PUTRA MALAYSIA**

SIMULTANEOUS REMOVAL OF CARBON DIOXIDE, SULPHUR DIOXIDE AND NITROGEN OXIDES USING ACTIVATED CARBON MODIFIED WITH METAL OXIDES

# HAZIMAH BINTI MADZAKI

FK 2019 3



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By

HAZIMAH BINTI MADZAKI

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfillment of the Requirements for the Degree of Master of Science

November 2018

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

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# Chair : Assoc. Prof. Ir. Wan Azlina Wan Abdul Karim Ghani, PhD Faculty : Engineering

Greenhouse gases act as a blanket to keep the Earth warm. However, excessive release of greenhouse gases in the atmosphere causes the Earth's temperature to rise above the threshold that led to many environmental problems. Carbon dioxide (CO<sub>2</sub>) is the largest contributor to this problem, but other gases make a significant contribution. Malaysia's greenhouse gas emissions are mainly from power station industry that produces electricity through burning of fossil fuels. Coal-fired power plants emit high concentration of CO<sub>2</sub> as well as sulphur dioxide (SO<sub>2</sub>) and oxides of nitrogen (NO<sub>x</sub>), two of the most widespread and dangerous gases other than CO2. These coal-fired power plants used many technologies to remove these gases from the flue gas stream. However, the technologies used are different for each gases; carbon capture and storage for CO<sub>2</sub> and flue gas desulphurization for SO<sub>2</sub> and low-NO<sub>x</sub> burner for NO<sub>x</sub> control. This multistage and complex removal processes can be very expensive due to high capital and operating cost. Hence, it is important to integrate the multistage process into a single step process for simultaneous removal of CO<sub>2</sub>, SO<sub>2</sub> and NO<sub>x</sub> to reduce the time and cost for flue gas cleaning.

This paper presents a simple way of modification of activated carbon surface using hydrothermal treatment and impregnation with metal oxides. The CO<sub>2</sub> adsorption performance of the modified activated carbon is evaluated by using a CO<sub>2</sub> adsorption unit while the simultaneous adsorption of CO<sub>2</sub>, SO<sub>2</sub> and NO<sub>x</sub> was done using flue gas adsorber unit with actual flue gas stream from combustion of coal in electrical furnace. Two types of metal oxides are used in this study which are cerium oxide (CeO<sub>2</sub>) and copper oxide (CuO). The adsorption study consists of activated carbon hydrothermally treated with metal oxides (ACCe-HT, ACCu-HT and ACCeCu-HT) and also activated carbon

impregnated with metal oxides (ACCe-WI, ACCu-WI and ACCeCu-WI). The objectives of the research are to investigate the physical-chemical properties of modified activated carbon with various metal oxides and to investigate the adsorption capacity in simultaneous removal of carbon dioxide, sulphur dioxides and nitrogen oxides.

In this study, the performance of activated carbon modified by hydrothermal treatment and impregnation techniques was compared. The prepared samples were characterized by different techniques using Brunauer-Emmet-Teller (BET) surface area analysis, Scanning Electron Microscopy (SEM) and X-ray diffraction (XRD). From BET analysis, the surface area and pore volume of raw AC was 1,288 m<sup>2</sup>/g and 0.38 cm<sup>3</sup>/g respectively. After modification, these values decreased with ACCe-HT having the highest surface area among the other modified sample which was 1,169 m<sup>2</sup>/g. This result corresponds to the SEM images that the surface of ACs were covered with metal oxides and peaks of metal oxides appear in XRD spectrum. The adsorption of CO<sub>2</sub> was investigated using a CO<sub>2</sub> adsorption unit, whereby 10% of CO<sub>2</sub> gas was passed through the samples until a breakthrough point was achieved. In the adsorption study, it was found that ACCe-HT had the highest adsorption capacity of 0.86 mmol/g with a breakthrough time of 19.33 min while ACCe-WI, ACCu-WI, ACCeCu-WI, ACCu-HT and ACCeCu-HT had the adsorption capacity of 0.12, 0.52, 0.06, 0.59 and 0.56 mmol/g respectively. In isotherm study using three isotherm models; Freundlich, Sips and Toth, it was also found that Toth Isotherm model shows good fit to the experimental data with correlation coefficient of 0.9910. In simultaneous adsorption from flue gas study using hydrothermal treated samples the adsorption capacity for ACCe-HT, ACCu-HT and ACCeCu-HT were 3.61, 1.16 and 0.50 mmol/g for CO2, 0.43, 0.57 and 0.31 mmol/g for SO<sub>2</sub> and 0.82, 0.96 and 0.78 mmol/g for NO<sub>x</sub>.

Overall, all objectives have been satisfied and it can be said that hydrothermally treated AC with CeO<sub>2</sub> has the potential to be used as adsorbent for CO<sub>2</sub> capture as well as SO<sub>2</sub> and NO<sub>x</sub> and the ability is comparable to the carbon monolith. Therefore, it is suitable to be applied in industry particularly flue gas system for CO<sub>2</sub>, SO<sub>2</sub> and NO<sub>x</sub> adsorption.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Master Sains

### PENYINGKIRAN SERENTAK KARBON DIOKSIDA, SULFUR DIOKSIDA DAN NITROGEN OKSIDA MENGGUNAKAN KARBON YANG DIAKTIFKAN YANG DIUBAH SUAI DENGAN LOGAM OKSIDA

Oleh

#### HAZIMAH BINTI MADZAKI

November 2018

### Pengerusi : Profesor Madya Ir. Wan Azlina Wan Abdul Karim Ghani, PhD Fakulti : Kejuruteraan

Gas rumah hijau berfungsi sebagai selimut untuk memastikan bumi hangat. Walau bagaimanapun, pelepasan gas rumah hijau yang berlebihan di atmosfera menyebabkan suhu bumi meningkat di atas ambang yang mengakibatkan banyak masalah alam sekitar. Karbon dioksida (CO<sub>2</sub>) adalah penyumbang terbesar kepada masalah ini, tetapi gas lain juga turut menjadi penyumbang kepada masalah yang sama. Pelepasan gas rumah hijau yang utama di Malaysia adalah dari industri stesen janakuasa yang menghasilkan elektrik melalui pembakaran bahan api fosil. Loji kuasa arang batu mengeluarkan kepekatan CO<sub>2</sub> yang tinggi serta sulfur dioksida (SO2) dan oksida nitrogen (NOx), dua daripada gas yang paling banyak dan berbahaya selain daripada CO<sub>2</sub>. Loji kuasa arang batu ini menggunakan banyak teknologi untuk menyingkirkan gas-gas ini dari aliran gas serombong. Walau bagaimanapun, teknologi yang digunakan adalah berbeza untuk setiap gas; penangkapan dan penyimpanan karbon untuk CO2 dan desulfurisasi gas serombong untuk SO<sub>2</sub> dan pembakar NO<sub>x</sub> rendah untuk kawalan NO<sub>x</sub>. Proses penyingkiran yang pelbagai dan kompleks ini boleh menjadi sangat mahal disebabkan oleh modal dan kos operasi yang tinggi. Oleh itu, adalah penting untuk mengintegrasikan proses yang kompleks ini menjadi satu langkah tunggal untuk penyingkiran serentak CO<sub>2</sub>, SO<sub>2</sub> dan NO<sub>x</sub> untuk mengurangkan masa dan kos untuk pembersihan gas serombong.

Manuskrip ini memperkenalkan cara pengubahsuaian permukaan karbon aktif menggunakan rawatan hidrotermal dan impregnasi menggunakan logam oksida. Prestasi penjerapan  $CO_2$  oleh karbon diaktifkan yang diubahsuai diuji dengan menggunakan unit penjerapan  $CO_2$  sementara penyingkiran serentak  $CO_2$ ,  $SO_2$  dan  $NO_x$  dilakukan dengan menggunakan unit penjerapan gas serombong dengan aliran gas serombong yang sebenar dari pembakaran

arang batu dalam relau elektrik. Dua jenis oksida logam digunakan dalam kajian ini iaitu cerium oksida (CeO<sub>2</sub>) dan tembaga oksida (CuO). Kajian penjerapan terdiri daripada karbon aktif hidroterma dirawat dengan oksida logam (ACCe-HT, ACCu-HT dan ACCeCu-HT) dan juga mengaktifkan karbon yang diresapi dengan oksida logam (ACCe-WI, ACCu-WI dan ACCeCu-WI). Objektif penyelidikan adalah untuk menyiasat sifat kimia fizikal karbon diaktifkan yang diubahsuai dengan pelbagai logam oksida dan untuk menyiasat kapasiti penjerapan dalam penyingkiran serentak karbon dioksida, sulfur dioksida dan nitrogen oksida.

Dalam kajian ini, prestasi karbon diaktifkan yang diubahsuai oleh rawatan hidrotermal dan teknik impregnasi telah dibandingkan. Sampel yang disediakan dicirikan oleh teknik-teknik yang berbeza menggunakan analisis permukaan permukaan Brunauer-Emmet-Teller (BET), Scanning Electron Microscopy (SEM) dan X-ray Difraction (XRD). Dari analisis BET, luas permukaan dan jumlah liang AC mentah ialah 1,288 m<sup>2</sup>/g dan 0.38 cm<sup>3</sup>/g.. Selepas pengubahsuaian, nilai-nilai ini berkurang dengan ACCe-HT yang mempunyai luas permukaan tertinggi di antara sampel yang diubahsuai yang lain iaitu 1,169 m<sup>2</sup>/g. Keputusan ini sesuai dengan imej SEM bahawa permukaan AC dilitupi dengan logam oksida dan puncak logam oksida muncul dalam spektrum XRD. Penjerapan CO<sub>2</sub> diuji menggunakan unit penjerapan CO<sub>2</sub>, di mana 10% gas CO2 disalurkan melalui sampel sehingga titik terobosan dicapai. Melalui kajian penjerapan, ia didapati ACCe-HT mempunyai kapasiti penjerapan tertinggi iaitu 0.86 mmol/g dengan masa terobosan 19.33 min manakala ACCe-WI, ACCu-WI, ACCeCu-WI, ACCu-HT dan ACCeCu-HT mempunyai kapasiti penjerapan sebanyak 0.12, 0.52, 0.06, 0.59 dan 0.56 mmol/g. Dalam kajian isotherm menggunakan tiga model isoterm; Freundlich, Sips dan Toth, didapati bahawa model Toth Isotherm menunjukkan kesesuaian dengan data eksperimen dengan pekali korelasi 0.9910. Dalam penjerapan serentak dari kajian gas serombong menggunakan sampel rawatan hidrotermal, kapasiti penjerapan untuk ACCe-HT, ACCu-HT dan ACCeCu-HT adalah 3.61, 1.16 dan 0.50 mmol/g untuk CO<sub>2</sub>, 0.43, 0.57 dan 0.31 mmol/g untuk SO<sub>2</sub> dan 0.82, 0.96 dan 0.78 mmol/g untuk NO<sub>x</sub>.

Secara keseluruhannya, semua objektif telah dicapai dan dapat dikatakan bahawa AC yang diubahsuai dengan  $CeO_2$  mempunyai potensi untuk digunakan sebagai penjerap untuk menangkap  $CO_2$  serta  $SO_2$  dan  $NO_x$  dan keupayaan adalah setanding dengan karbon monolit. Oleh itu, ia sesuai untuk digunakan dalam industri terutama sistem asap serombong untuk penjerapan  $CO_2$ ,  $SO_2$  dan  $NO_x$ .

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfillment of the requirement for the degree of Master of Science. The members of the Supervisory Committee were as follows:

#### Wan Azlina Binti Wan Ab Karim Ghani, PhD

Associate Professor, Ir. Faculty of Engineering Universiti Putra Malaysia (Chairman)

### Thomas Choong Shean Yaw, PhD

Professor, Ir. Faculty of Engineering Universiti Putra Malaysia (Member)

#### Noraziah Muda

Principal Researcher Tenaga Nasional Berhad Research Malaysia (Member)

> **ROBIAH BINTI YUNUS, PhD** Professor and Dean School of Graduate Studies Universiti Putra Malaysia

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Signature: Name of Chairman of Supervisory Committee:	Assoc. Prof. Ir. Dr. Wan Azlina Wan Ab Karim Ghani
Signature: Name of Member of Supervisory Committee:	Prof. Dr. Ir. Thomas Choong Shean Yaw
Signature: Name of Member of Supervisory Committee:	Puan Noraziah Muda

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### LIST OF ABBREVIATIONS

1/n	system heterogeneity
AC	activated carbon
AC ACCeCu-HT	activated carbon hydrothermal treated with cerium oxide
ACCECU-III	and copper oxide
ACCeCu-WI	activated carbon impregnated with cerium oxide and
Accedu-m	copper oxide
ACCe-HT	activated carbon hydrothermal treated with cerium oxide
ACCe-WI	activated carbon impregnated with cerium oxide
ACCu-HT	activated carbon hydrothermal treated with copper oxide
ACCu-WI	activated carbon impregnated with copper oxide
AEE	2-2-aminoethylamino ethanol
AEPDNH <sub>2</sub>	N-2-aminoethyl 1,3-propanediamine
AIO	aluminium oxide
AMP	2-amino 2- methyl 1-propanol
ARG-C	argan fruit shell activated carbon
BET	Brunauer-Emmett-Teller
C	outlet concentration
C <sub>0</sub>	initial concentration
Ca	calcium
CaCO₃	calcium carbonate
CCS	carbon capture and storage/sequestration
CDM	Clean Development Mechanism
Ce	cerium
CeO <sub>2</sub>	cerium oxide
СМ	carbon monolith
СМС	carboxymethylcellulose
CNT	carbon nanotube
Со	cobalt
СО	carbon monoxide
CO <sub>2</sub>	carbon dioxide
COP21	Conferences of Parties 21
Cr	chromium
CS	raw coconut shell biochar
Cu	copper
CuO	copper oxide
C <sub>x</sub> H <sub>y</sub>	hydrocarbon
DEA	diethanol amine
DGA	diglycol- amine
DIPA	di-2-propanolamine
DOE	Department of Environmental
ESA	electrical swing adsorption

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EU F Fe FGD GA GHG $H_2$ $H_2O$ $H_3PO_4$ HCL He HNO_3 HP5 HT IPCC	European Union feed flow rate iron flue gas desulphurization glucosamine greenhouse gases hydrogen water phosphoric acid hydrochloric acid helium nitric acid persian ironwood was activated by h <sub>3</sub> po <sub>4</sub> hydrothermal treatment Intergovernmental Panel on Climate Change
IUPAC K	International Union of Pure and Applied Chemistry potassium
K <sub>2</sub> CO <sub>3</sub>	potassium carbonate
k⊧	Freundlich adsorption constant
КОН	potassium hydroxide
ks	adsorption constant
k⊤	Toth adsorption constant
m	heterogeneity parameter
MDEA	N-methyldiethanolamine
MEA	monoethanol amine
Mg	magnesium
MgO	magnesium oxide
MO	metal oxide
MWCNT	multi-walled carbon nanotube based
N N/A	nitrogen
N/A Na <sub>2</sub> CO <sub>3</sub>	not available sodium carbonate
NaOH	sodium hydroxide
N-CS	amine treated coconut shell biochar
NH <sub>3</sub>	ammonia
Ni	nickel
NiO	nickel oxide
NO	nitrogen oxide
NO <sub>2</sub>	nitrogen dioxide
NOx	oxides of nitrogen
NRE	Ministry of Natural Resources and Environment
O <sub>2</sub>	oxygen
P123	poly(ethylene glycol)-block-poly(propylene glycol)-block-
	poly(ethylene glycol)

PAH	nalvavalia aramatia hydrogarhan
PEI	polycyclic aromatic hydrocarbon
	polyethylenimine
PSA	pressure swing adsorption
PSAC	palm shell activated carbon
PSAC-Ce	palm shell activated carbon activated by cerium
PZ	piperazine
q	adsorption capacity
qf	amount of adsorbed CO <sub>2</sub>
<b>q</b> <sub>m,S</sub>	maximum adsorption capacity
<b>q</b> m,T	maximum adsorption capacity
qs	absolute amount adsorbed
q⊤	amount of adsorbed CO <sub>2</sub>
R <sup>2</sup>	correlation coefficient
S	sulphur
S	system heterogeneity
SEM	Scanning Electron Microscopy
SM	silica monolith
SO <sub>2</sub>	Sulphur dioxide
SOx	oxides od sulphur
t	reaction time
TEA	triethanol amine
TETA	triethylene tetra amine
TNB	Tenaga Nasional Berhad
TNBR	Tenaga Nasional Berhad Research
TSA	temperature swing adsorption
US	United States
UTP	Universiti Teknologi Petronas
VOCs	volatile organic compounds
W	amount of adsorbent
ŴI	wet impregnation
XRD	X-ray Diffraction

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### CHAPTER 1

#### INTRODUCTION

### 1.1 Background

Global warming is an alarming issue faced globally. Since the industrial revolution, humans have been altering this process by adding more carbon dioxide and other harmful gases to the atmosphere. Global atmospheric concentration of the greenhouse gases (GHG) show significant seasonal and year-to year variability, but all show on going upward trends. Figure 1.1 shows the global mean GHG concentrations from the year 1978 to 2015.

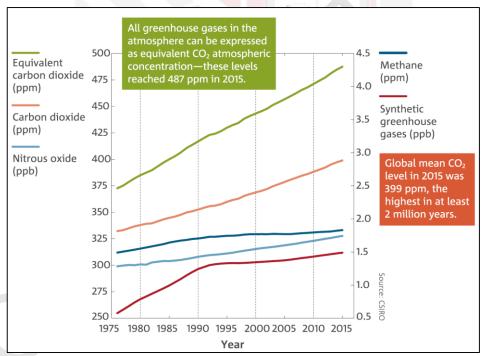
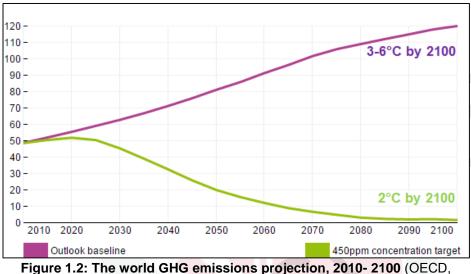


Figure 1.1: Global mean GHG concentrations from the year 1978 to 2015 (CSIRO, 2016).

The world leaders have decided to take actions to reduce emissions to avoid 2 °C of earth warming, which is the level outlined as a safe threshold for climate change. It is forecasted that, if no action is taken to reduce the emission, the earth temperature will increase to three to six degree Celsius. Figure 1.2 shows



the world greenhouse gases, GHG emissions projection of baseline scenario (no action taken) versus targeted GHG concentration level which is 450 ppm.

Figure 1.2: The world GHG emissions projection, 2010- 2100 (OECD 2012).

There are two causes of global warming which are categorized into natural and anthropogenic forcing. Natural forcing is the tilt of the earth axis that have time scales of thousands of years and variation in interaction between the ocean and atmosphere can result in climate variations of yearly, decadal and century time scales while anthropogenic forcing is human induced activities which lead to increasing of the concentration of GHG into the atmosphere (MET, 2009). Greenhouse effect is the process in which the infrared radiation was absorbs by water vapor, carbon dioxide (CO<sub>2</sub>), methane and other atmospheric gases to warms the Earth's surface (Chiang & Juang, 2017; Yaumi *et al.*, 2017; Mondal *et al.*, 2012). However, excessive greenhouse gases in the atmosphere cause rises in earth's temperature that led to various environmental problems such as continuous rise of water-level in sea, the increasing number of ocean storms and floods (Hamza *et al.*, 2016; Mondal *et al.*, 2012).

Since 1990s, the economic growth in Malaysia has been increased in parallel with urbanization, stimulating in high energy demand. As stated in Tenth Malaysia Plan (EPU, 2010), by 2020, it is predicted that three quarters of the Malaysia population will be living in the urban areas. Bekhet & Othman (2017) studied on the theory of urbanization affects the environmental transition and concluded that as Malaysia is developing at a fast rate of urbanization with huge numbers of industrial areas and well-planed housing settlements has leads to higher level of atmospheric pollution. As the numbers of wealthier citizens increases, the demand for energy intensive products also increases. Hence, Malaysian government is trying to reduce the energy used through

transportation industry by focusing on urban transportation network (Bekhet & Othman, 2017; Zhang *et al.*, 2017; Shahbaz *et al.*, 2016; Lau *et al*, 2014).

The Malaysian Department of Environmental (DOE) has reported in 2010 the air pollution in the country main contributors includes power stations industry, motor vehicles industry and open burnings. Production of electricity in Malaysia is mainly from burning of fossil fuel such as oil, coal or natural gas that has huge consequences for the environment. Malaysia used petroleum and other liquids and natural gas as the main energy sources followed by coal, hydroelectricity and biomass. Since Malaysia relies so much on the oil and natural gas to sustain its economic growth, it has cause many environmental problem such air pollution and global warming. The concern to the environment has caused the government to find an alternative energy sources from the used of coal to renewable energy (EIA, 2017).

Fossil fuel power plants have the highest CO<sub>2</sub> emission and coal-fired power plant is the main contributor among them. Coal has become much more competitive with natural gas-fired power because of the low fuel price and has gained a larger portion of power generation in Peninsula Malaysia in the past few years. There are several coal-fired power plants in Peninsular Malaysia which are Jimah Power Station in Negeri Sembilan, Manjung Power Station in Perak, Sultan Salahuddin Abdul Aziz Shah Power Station in Selangor and Tanjung Bin Power Station in Johor (Wikipedia, 2017). Among all this coalfired power plant, Manjung Power Station is owned by Tenaga Nasional Berhad (TNB), the largest electric utility company in Malaysia.

CO<sub>2</sub> is one of the major pollutants in the atmosphere, which contribute to greenhouse effect. The rise in concern towards the earth climate has attracted extensive effort to introduce efficient and cost-effective technologies for capturing or reducing CO<sub>2</sub> from large point sources like coal-fired power plant. One of the main technologies in reducing greenhouse gases is postcombustion capture of CO<sub>2</sub> because it has the potential to be retrofitted to existing coal-fired power plant without requiring substantial changes to the combustion process (Hajari, 2017; Rackley, 2017; Thiruvenkatachari et al., 2015). Technologies such as absorption, cryogenic and membrane separation have been proposed to capture CO<sub>2</sub> from the flue gases (Keller et al., 2018; Mohshim et al., 2018; Singh et al., 2018; Querejeta et al., 2017; Lee & Park, 2015; Ruiz et al., 2013; Choi et al., 2009; Zhou et al., 2012). However, these technologies have some disadvantages. For example in absorption, solvent degradation and corrosion may occur and affects the CO<sub>2</sub> removal efficiency (Liang et al., 2016; Shakerian et al., 2015; Fytianos et al., 2014). For cryogenic process, the H<sub>2</sub>O must be completely removed to prevent corrosion, fouling and plugging which leads to higher cost (Kang et al, 2017). In membrane separation, multiple stages of separation or recycling is needed since it cannot always give high separation degrees which makes the cost of membranes reactor is high (Nakhjiri et al., 2018; Mondal et al., 2012; Olajire, 2010).

Adsorption was considered as one of the most promising technologies in the commercial and industrial applications because of the low energy requirement, cost advantage, and ease of applicability over a relatively wide range of temperatures and pressures (Yue *et al.*, 2018; Serafin *et al.*, 2017; Kongnoo *et al.*, 2016; Younas *et al.*, 2016; Thiruvenkatachari *et al.*, 2009; Duffy *et al.*, 2006; Song, 2006). CO<sub>2</sub> capture by adsorption technology had drawn much research effort recently and had done lots of work to improve capture performance (Dassanayake & Jaroniec, 2018; Pan *et al.*, 2018; Irani *et al.*, 2017; Querejeta *et al.*, 2017; Kwiatkowski *et al.*, 2016). Activated carbon (AC) as an agent for adsorption has been used for many years in many field because this amorphous material has large surface area and pore volume. Gas-phase adsorption by activated carbon is a separation process in which adsorbate molecules are transferred to the pore surface of solid activated carbon (Sumathi *et al.*, 2010).

ACs as suitable candidates for CO<sub>2</sub> capture are highly anticipated, however, there is some restriction which is the low selectivity of AC towards CO<sub>2</sub>. It has been known that the surface chemistry of AC can affect the adsorption chemistry which can be enhanced greatly using chemical modification like introducing the basic groups to the surface of AC (Chiang & Juang, 2016). To this day, many metal oxides have been used to further improve the CO<sub>2</sub> capture capacity. Still, one major problem for metal oxide modification is that the CO<sub>2</sub> adsorption capacity is still too moderate (Wang *et al.*, 2014). Loading metal oxides onto the surface of the AC cause a reduction in the surface area as well as pore volume. This is due to the deposition of the metal particles into the interior structures and blocks some parts of the micropores and mesopores (Rashidi & Yusup, 2016).

Other technique that can modify AC physically is hydrothermal treatment. Hydrothermal treatment is commonly used in application like biodiesel production, activation of chars and carbonation of biomass (Breulmann *et al.*, 2017; Ding *et al.*, 2018; Jain *et al.*, 2016; Lagazzo *et al.*, 2016; Soltani *et al.*, 2017). There are only a few works devoted to hydrothermal treatment of carbon materials with the purpose of modification of their porous and surface structure (Hlaing et al., 2014; Wu, Li, Liu, & Jin, 2016; Xiao, Guo, Zhao, & Han, 2016). The chemical and physical properties, high speed, high yield, low cost and being eco-friendly (due to sealed system condition) are some of the characteristic of hydrothermal treatment (Deng *et al.*, 2016). Hence hydrothermal treatment can enhanced the pore structure of an absorbent for better adsorption.

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Coal-fired flue gas stream which also contains oxides of nitrogen NO<sub>x</sub> and sulphur oxides SO<sub>x</sub>, two of the most widespread and dangerous gases other CO<sub>2</sub> (Sumathi *et al.*, 2010b), hence, the removal of these gases should also be taken into consideration. Many technologies have been used to remove these gases from flue gas. However, the process applied can only remove one gas at a time; flue gas desulphurization of SO<sub>2</sub>, NO<sub>x</sub> control and carbon capture and storage for CO<sub>2</sub>. This multistage and complex removal processes can be very

expensive due to high capital and operating cost (Hajari, 2017; Yi, Huang, et al., 2014). Hence, it is important to integrate the multistage process into single step process for the simultaneous removal of  $CO_2$ ,  $SO_2$  and  $NO_x$  from flue gas.

This paper focus on modification of activated carbon surface using wet impregnation and hydrothermal treatment with metal oxides. The adsorption performance of the modified activated carbon is evaluated by using a CO<sub>2</sub> and flue gas adsorption unit. Recent study on doping of various oxides of metals; magnesium (Mg), calcium (Ca), copper (Cu), cobalt (Co), nickel (Ni), iron (Fe) and chromium (Cr), onto AC enhance the adsorption capacity of CO<sub>2</sub> and among all, it is reported that AC impregnated with CuO results in higher CO2 percentage removal (Rashidi and Yusup, 2016; Hosseini et al., 2015; Kim et al., 2010; Moradi, 2014; Fenrong et al., 2010; Yong and Mata, 2001). A study by Sumathi et al. (2010b) on selection of best impregnated AC for removal of  $SO_2$  and  $NO_x$  shows that AC impregnated with cerium oxide (CeO<sub>2</sub>) has the best removal compare to other metal oxides; nickel (Ni), iron (Fe), vanadium (V). The study of simultaneous capture of CO<sub>2</sub>, SO<sub>x</sub> and NO<sub>x</sub> reported is very limited. Currently, the removals only focus on  $CO_2$  or  $SO_x$  and/or  $NO_x$ . Hence, in this study, two types of metal oxides which are cerium oxide and copper oxide are used for modification of AC to allow simultaneous adsorption of CO<sub>2</sub>, SO<sub>x</sub> and NO<sub>x</sub> from flue gas stream.

### 1.2 Problem Statement

At present, research in the field of adsorbents had made rapid progress for adsorbing pollutant in flue gas. AC is preferred as  $CO_2$  adsorbent due to its environmentally friendly nature, low cost, high porous structure, high surface area and good mechanical properties. However, other than  $CO_2$  coal-fired flue gas stream also contains traces of oxides of nitrogen  $NO_x$  and sulphur oxides  $SO_x$ . Hence, the removal of these gases simultaneously is also considered in order to save cost and time compare to when they are captured separately.

A lot of attention is given in modification of the surface chemistry of existing carbon sorbents so that adsorption capacity is increased and selectivity is improved. Since  $CO_2$  is an acidic gas, the basicity properties of metal oxides can increase the adsorption capacity of  $CO_2$  since metal oxides is an electron donor that can attract  $CO_2$  molecules. However, impregnating metal oxides onto the surface of the AC cause a reduction in the surface area as well as pore volume. This is due to the deposition of the metal particles into the interior structures and blocks some parts of the pores. Hence hydrothermal treatment is also introduced in modification of AC to alter the porous and surface structure as well as loading the metal oxides particles onto AC surface.

Copper oxide (CuO) and cerium oxide (CeO<sub>2</sub>) are selected to be loaded to AC surface by the two modification methods; wet impregnation and hydrothermal treatment. Based on previous study, CuO has better CO<sub>2</sub> adsorption while

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 $CeO_2$  gave higher adsorption capacity for  $SO_2$  and  $NO_x$  compared to other metal oxides. Hence, these metal oxides are used in this study for simultaneous adsorption of  $CO_2$ ,  $SO_2$  and  $NO_x$ .

### 1.3 Research Objectives

The objectives of the research are:

- 1. To investigate the physical-chemical properties of raw and modified activated carbon (AC) with various metal oxides.
- 2. To analyse adsorption capacity of simultaneous removal of carbon dioxide (CO<sub>2</sub>), sulphur dioxide (SO<sub>2</sub>) and nitrogen oxides (NO<sub>x</sub>).

### 1.4 Scope and Limitation of Research

The scope of the research includes preparing AC modified with metal oxides via hydrothermal treatment and impregnation. The modified activated carbons were then characterized by using BET analysis to estimate the surface area, pore size, pore volume, pore diameter and pore distribution. The surface morphology of the samples is determined by Scanning Electron Microscopy (SEM). Determinations of the structure and fingerprint characterization of crystalline materials were done by using X-ray Diffraction (XRD). The performance of the modified AC in capturing CO<sub>2</sub> is tested using a CO<sub>2</sub> adsorption unit. The method that gives better adsorbent performance for CO<sub>2</sub> capture will be screened for simultaneous removal of CO<sub>2</sub> and SO<sub>2</sub>/NO<sub>x</sub> from flue gas testing. It is expected that the adsorption study of these modified AC will provide a new insight on their ability as simultaneous CO<sub>2</sub> and SO<sub>2</sub>/NO<sub>x</sub> adsorbent.

There are some notable limitations in this research. There is lack of previous studies on modification of AC with metal oxides via hydrothermal treatment. Therefore, the literatures that have similar aim that is to enhance the pore structure of the AC using the same type of precursors are being referred to. Other than that, the measure used to collect the data is restricted. The inlet and outlet concentration of the adsorption of gases need to be done using different batch of coal combustion due to the lack of gas analyzer. Hence the breakthrough curve and adsorption capacity cannot be calculated accurately. Thus, it is important to ensure the combustion of coal is done using the same parameter throughout the experiment.

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### **BIODATA OF STUDENT**

Hazimah binti Madzaki is a master's student in Chemical Engineering under the supervision of Assoc. Prof. Dr. Wan Azlina Wan Ab Karim Ghani. Her interest is in environment engineering, sustainable energy and gas cleaning. Prior to enrolling at Universiti Putra Malaysia (UPM), she worked as research assistance under the same supervision for one month. She graduated from Universiti Teknologi MARA (UiTM) with Diploma in Chemical Engneering and Bachelor of Engineering (Hons) Chemical and Process in 2011 and 2014 respectively. In February of 2015, he pursued his Master's degree in Chemical and Environmental Engineering under the supervision of Associate Professor Dr Wan Azlina in UPM.

### LIST OF PUBLICATIONS

- Madzaki, H., Ghani, W. A. W. A. K., Yaw, T. C. S., and Muda, N. (2017). Carbon dioxide adsorption using activated carbon impregnated with carbon dioxide. *Proceedings of the 4th Postgraduate Colloquium for Environmental Research* 2017 (POCER 2017), 44-45.
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