

**SYNTHESIS OF SORBENTS FOR CO₂ CAPTURE
FROM BIOGENESIS CALCIUM WASTE**

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**SYNTHESIS OF SORBENTS FOR CO₂ CAPTURE FROM BIOGENESIS
CALCIUM WASTE**

by

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LIST OF ABBREVIATIONS

AGS	Anadara granosa shells
Al	Aluminium
Al(NO ₃) ₃	Aluminium nitrate
Al(NO ₃) ₃ .9H ₂ O	Aluminum nitrate enneahydrate
Al ₂ O ₃	Aluminium oxide
BET	Brunauer–Emmett–Teller
Ca(NO ₃) ₂	Calcium nitrate
Ca(OH) ₂	Calcium hydroxide
Ca ₁₂ Al ₁₄ O ₃₃	Mayenite
CaCO ₃	Calcium carbonate
CaO – AGS	CaO derived from calcined anadara granosa shells
CaO – MMS	CaO derived from calcined meretrix meretrix shells
CaO – TVCS	CaO derived from calcined textile venus clam shells
CaO	Calcium oxide
CaZrO ₃	Calcium zirconate
CO ₂	Carbon dioxide
GDP	Gross domestic product
GHG	Greenhouse gas
H ₂	Hydrogen gas
H ₂ O	Distilled water
MEA	Monoethanolamine
Mg	Magnesium
MgO	Magnesium oxide

MMS	Meretrix meretrix shells
MRT	Mass Rapid Transit
MT	Metric tonnes
N ₂	Nitrogen gas
NDC	Nationally Determined Contribution
O ₂	Oxygen gas
SCM	Shrinking core model
SEDA	Sustainable Energy Development Authority
SEM	Scanning electron microscopy
Si	Silicon
TGA	Thermogravimetric analyzer
Ti	Titanium
TVCS	Textile venus clam shells
UNFCCC	United Nations Framework Convention on Climate Change
Wt %	Weight percentage
XRD	X-ray diffraction
XRF	X-ray fluorescence
Zr	Zirconia
ZrO ₂	Zirconium oxide

SINTESIS PENJERAP UNTUK MENJERAP CO₂ DARIPADA SISA KALSIUM BIOGENESIS

ABSTRAK

Pemanasan global telah menjadi salah satu isu global penting kerana kepekatan karbon dioksida (CO₂) semakin meningkat. Penjerapan CO₂ dengan menggunakan kalsium oksida (CaO) yang diperolehi daripada sisa semula jadi telah menarik perhatian ramai penyelidik. Pemanfaatan sorben hijau untuk penjerapan CO₂ ini dapat meminimumkan kos sorben yang menguntungkan semua kos pengeluaran. Dalam kajian ini, kerang sisa semulajadi yang merupakan kerang anadara granosa, kerang meretrix meretrik dan kerang venus kerang tekstil telah digunakan sebagai sumber untuk mendapatkan CaO. Dalam analisis pendarfluor sinar-X (XRF), lebih daripada 97 % berat CaO ditemui di dalam kerang sisa semulajadi selepas pengkalsinan pada 900 °C. Beberapa parameter telah diselidik termasuk tempoh pengkarbonan, suhu pengkarbonan, jenis bahan tambahan logam ditambah dan peratusan berat bahan tambahan logam yang ditambahkan di sorben. Kapasiti penjerapan tertinggi didapati apabila tempoh pengkarbonan adalah 60 minit. Julat suhu pengkarbonan yang dikaji dalam kajian ini adalah 400 – 800 °C. Kapasiti penjerapan tertinggi untuk kerang anadara granosa yang dikalsin (CaO – AGS) dan kerang meretrik meretrik yang dikalsin (CaO – MMS) didapati pada 800 °C manakala kulit kerang venus tekstil (CaO – TVCS) dan CaO komersial mempunyai kapasiti penjerapan yang tertinggi pada 700 °C. Walau bagaimanapun, CaO komersial mempunyai kapasiti penjerapan tertinggi berbanding dengan yang lain kerana ia mempunyai kawasan permukaan tertinggi (100.63 m²/g). Kajian regenerabiliti juga telah dibuatkan untuk semua pemjerap pada 700 °C untuk

11 kitaran. CaO – TVCS telah didapati sebagai sorben terbaik kerana kapasiti penjerapan yang lebih tinggi dan penurunan kecekapan kecil (24.27 %) dalam kajian regenerabiliti. Kajian kinetik penjerap juga dinilai. Tenaga pengaktifan untuk CaO – AGS, CaO – MMS, CaO komersial semasa pertama tahap process pengkarbonan adalah 47.13, 18.19, 26.98 dan 30.18 kJ/mol masing-masing. Tenaga pengaktifan untuk CaO – AGS, CaO – MMS, CaO – TVCS dan CaO komersial semasa tahap kedua process pengkarbonan adalah 20.19, 20.74, 21.09 dan 15.18 kJ/mol masing-masing. Kapasiti penjerapan CaO – TVCS ditambah dengan bahan tambahan logam (Al_2O_3 dan ZrO_2) telah menurun dengan peningkatan peratusan berat bahan tambah logam. Kapasiti penjerapan penjerap yang ditambah dengan bahan tambahan logam didapati bergantung pada peratusan berat CaO dalam penjerap. Selepas menjalankan kajian regenerabiliti, CaO:ZrO₂ dengan peratusan berat 90:10 telah didapati sebagai penjerap terbaik kerana penurunan kecekapan yang lebih rendah (19.05 %) berbanding dengan CaO:Al₂O₃ dengan peratusan berat 90:10 (22.47 %) dan CaO – TVCS (38.04 %). Oleh itu, dapat disimpulkan bahawa CaO:ZrO₂ dengan peratusan berat 90:10 adalah alternatif yang berpotensi untuk CaO kerana ia mempunyai ketahanan dan kestabilan yang lebih tinggi daripada CaO.

SYNTHESIS OF SORBENTS FOR CO₂ CAPTURE FROM BIOGENESIS

CALCIUM WASTE

ABSTRACT

Global warming has become one of the essential global issues as the world concentration of CO₂ is rising. Carbon dioxide (CO₂) capture by using calcium oxide (CaO) derived from natural waste shells has attracted the attentions of many researchers. Utilization of these green sorbents for CO₂ capture is able to minimize the cost of the sorbents which is favourable to the whole production cost. In this research, the natural waste shells which are anadara granosa shells, meretrix meretrix shells and textile venus clam shells were utilized as the sources to obtain CaO. In the X-ray fluorescene (XRF) analysis, more than 97 wt % of CaO was found in the natural waste shells after calcination process at 900 °C. Several parameters were investigated including carbonation duration, carbonation temperature, type of metal additives added and weight percentage of metal additive added to sorbent. The highest sorption capacity was found when the carbonation duration was 60 minutes. The range of the carbonation temperature studied in this research was 400 – 800 °C. The highest sorption capacities for calcined anadara granosa shells (CaO – AGS) and calcined meretrix meretrix shells (CaO – MMS) were at 800 °C while calcined textile venus clam shells (CaO – TVCS) and commercial CaO had their highest carbonation conversion at 700 °C. However, commercial CaO had the highest sorption capacity as compared to others since it had the highest surface area (100.63 m²/g). Regenerability study also had been carried for all the sorbents at 700 °C for 11 cycles. CaO – TVCS had been found as the best sorbent due to its higher carbonation conversion and minor

efficiency drop (24.27 %). Kinetic study of the sorbent samples was evaluated. The activation energy for CaO – AGS, CaO – MMS, CaO - TVCS and commercial CaO during the chemical reaction controlled stage of carbonation reaction were 47.13, 18.19, 26.98 and 30.18 kJ/mol respectively. The activation energy for CaO – AGS, CaO – MMS, CaO - TVCS and commercial CaO during the product layer diffusion controlled stage of carbonation reaction were 20.19, 20.74, 21.09 and 15.81 kJ/mol respectively. The sorption capacities of the CaO – TVCS added with metal additives (Al_2O_3 and ZrO_2) were decreased with the increasing weight percentage of metal additives. The sorption capacities of the sorbent samples added with metal additives were found relied on the weight percentage of CaO in the sorbent samples. After conducting the regeneration study, CaO: ZrO_2 with 90:10 weight percentage had been found as the best sorbent samples owing to its lower efficiency drop (19.05 %) in comparison to CaO: Al_2O_3 with 90:10 weight percentage (22.47 %) and CaO – TVCS (38.04 %). Therefore, it can be concluded that CaO: ZrO_2 with 90:10 weight percentage is a potential alternative to CaO as it has higher durability and stability than CaO.

CHAPTER ONE

INTRODUCTION

1.1 Carbon Dioxide (CO₂)

Carbon dioxide (CO₂) is one of the greenhouse gases emitted from human activities. In fact, there are two sources of CO₂ emission such as natural CO₂ source and anthropogenic CO₂ source. The examples of natural sources of carbon dioxide are respiration of plants and animals, emissions from volcanic eruptions, ocean releases as well as the decomposition of organic matters. In contrast, the anthropogenic CO₂ sources are emissions from transportation sectors, chemical and petroleum production, deforestation and also combustion of the fossil fuels including oil, coal and natural gas. For instance, approximately 44 % of anthropogenic CO₂ emissions is from coal fired power plants, oil fired power plants as well as the gas fired power plants (Wang, et al., 2011). There are around 7 coal fired power plants can be found in Malaysia (Williams, 2016). Additionally, Environmental Protection Agency claimed that the allowance of CO₂ that could be emitted from power plants was 1000 pounds/megawatt-hour of energy generated (Barringer, 2012).

In 2011, the highest CO₂ emission in Malaysia is from the energy industries due to fuel in order to produce electricity (55 %), followed by the transportation sector (21 %) and manufacturing and construction sectors (11 %) (Ministry of Natural Resources and Environment Malaysia, 2015). Figure 1.1 presents an overview of the CO₂ emissions by sources in Malaysia in 2013. From the pie chart shown in Figure 1.1, it can be seen that by far the highest percentage of CO₂ emission is still caused by electricity and heat production, followed by transportation sector and manufacturing

industries and construction sector. Therefore, attention should be drawn in these three sectors in order to mitigate the CO₂ emissions in Malaysia.

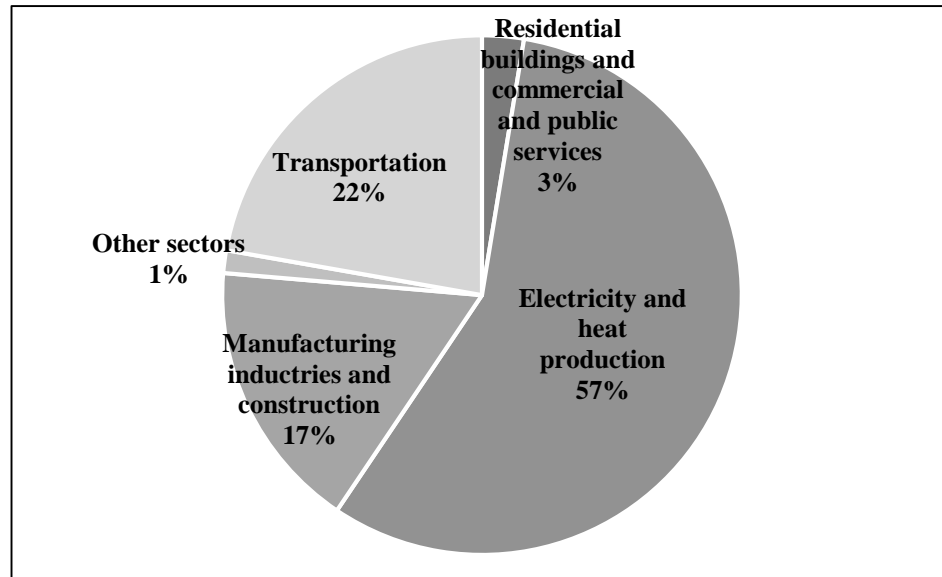


Figure 1.1: Percentage of CO₂ emissions by sectors in Malaysia in 2013 (Trading Economics, 2017)

The accumulation of CO₂ and other greenhouse gases in the atmosphere serve as ‘blanket’ that absorbs the sunlight with the radiations which bounce off the surface of the earth. Excessive concentration of CO₂ in the atmosphere will trap the heat and warm the earth and hence further increase the temperature of the earth. The rising of CO₂ concentration will cause global warming and devastating environmental effects such as the frequent occurrence of storms, floods as well as droughts (Safaai, et al., 2011). Meanwhile, the effects of global warming are climate change, melting of the glaciers, massive crop failure as well as the human health damage.

Another effect that caused by the increasing of CO₂ is ocean acidification. Up to the present time, anthropogenic CO₂ emissions that acidify the ocean is at an alarming level. The most compelling evidence is around 27 % of CO₂ from combustion

of fossil fuels together with deforestation since the industrial revolution has dissolved in the ocean (Nina, 2014). The pH of the ocean will drop with the increasing amount of CO₂ dissolves in the ocean because carbonic acid will be produced when CO₂ dissolved and reacted with water (H₂O) in the ocean. For instance, the pH of the ocean is reduced from 8.2 to 8.1 (Ocean Portal Team, 2018). Besides, there is a prediction that the ocean pH will reduce to 7.8 by the end of this century and causing the ocean to become more acidic (Ocean Portal Team, 2018). Hence, the marine ecosystem will be affected if the pH drops inasmuch as carbonate ion (CO₃²⁻) is vital substrate in the biotic calcification process (Fabricius, et al., 2011). The example of the ocean that had been affected by drop in pH is Southern Ocean (Antarctic and Southern Ocean Coalition, 2018).

1.2 CO₂ Emissions

Figure 1.2 depicts the global CO₂ concentration has risen from 338.8 ppm in 1980 to 402.87 ppm in 2016 (Ritchie and Roser, 2017). As can be seen in Figure 1.2, there is an increment of 64.07 ppm in these 36 years. Apart from that, the total global CO₂ emissions in 2015 and 2016 are tabulated in Table 1.1. The total global CO₂ emissions is increasing from 33303.9 million tonnes (2015) to 33432.0 million tonnes (2016) and the total CO₂ emissions discharged from Asia Pacific is the highest as compared to others. In Asia Pacific, the largest CO₂ contributors are China (9123 million tonnes), India (2271.1 million tonnes), followed by Japan (1191.2 million tonnes) (BP Statistical Review of World Energy, 2017).

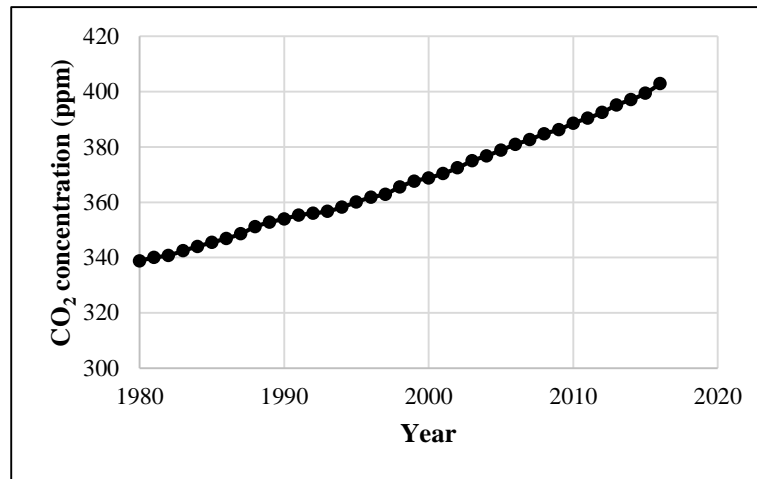


Figure 1.2: Global CO₂ concentration (Ritchie and Roser, 2017)

Table 1.1: Total global CO₂ emissions (BP Statistical Review of World Energy, 2017)

	CO ₂ emissions (million tonnes)	
	2015	2016
North America	6458.1	6348.0
South & Central America	1377.9	1348.2
Europe & Eurasia	6240.7	6258.5
Middle East	2127.5	2167.8
Africa	1192.5	1209.0
Asia Pacific	15907.2	16100.5
Total	33303.9	33432.0

Malaysia is one of the countries which contributes to the global CO₂ emission since it is mainly relies on fossil fuels and coals to generate electricity. In 2016, the CO₂ emission of Malaysia was 263.8 million tonnes, which was comparatively low to the three largest CO₂ contributors to the global CO₂ emission, notably China (9123 million tonnes), U.S. (5350.4 million tonnes) and India (2271.1 million tonnes) (BP Statistical Review of World Energy, 2017). Figure 1.3 shows the CO₂ emissions per capita of Malaysia was risen from 1.25 metric tons in 1970 to 8.09 metric tons in 2015 (Knoema, 2017). There is an increment of 6.84 metric tons of CO₂ per capita in these 45 years. Henceforth, awareness should be raised for this increment of CO₂ emission in our country.

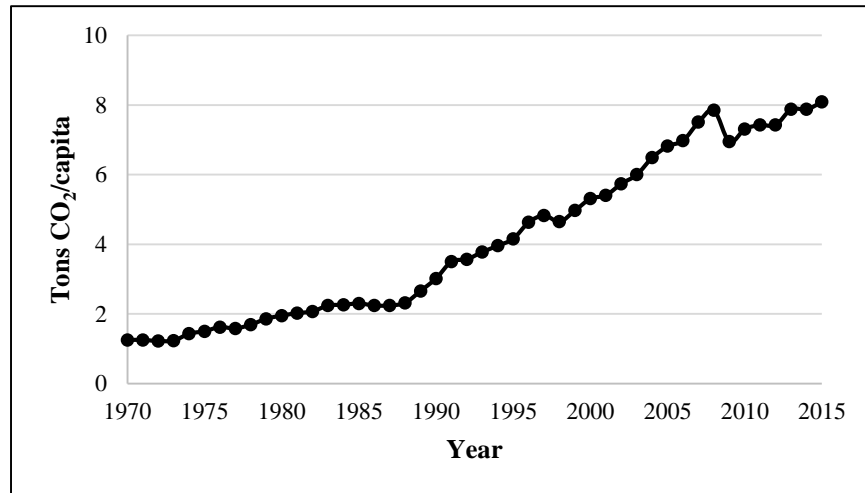


Figure 1.3: CO₂ emissions per capita of Malaysia from 1970 to 2015 (Knoema, 2017)

1.3 Efforts of Reducing CO₂ in Malaysia

Malaysia has participated in United Nations Framework Convention on Climate Change (UNFCCC) in January 2016 and ratified in November 2016 (Fulton, et al., 2017). According to the Minister of Natural Resources and Environment, Malaysia has a target to reduce the CO₂ emissions by 300,000 tonnes by year 2030 and enables Malaysia in achieving the target determined by the Nationally Determined Contribution (NDC) proposed to UNFCCC under 2015 Paris Agreement (Lye, 2017).

Malaysia is a country which typically depends on the combustion of fossil fuels and coals in the industries. Hence, adoption of renewable energy will aid in reducing the CO₂ emissions indirectly. Table 1.2 tabulates the acts/laws/policies in Malaysia to reduce CO₂ emissions. Apart from that, Eleventh Malaysia Plan (2016 – 2020) has been introduced with the purpose of promoting the practice of energy efficient vehicles and public transport, notably Mass Rapid Transit (MRT) system. As an illustration, approximately 34,400 tonnes of annual CO₂ emissions is predicted will be reduced by the adoption of MRT (Lye, 2017).