



**UNIVERSITI PUTRA MALAYSIA**

***CARBON SEQUESTRATION OF MINING WASTE IN REDUCING  
CARBON DIOXIDE EMISSION THROUGH MINERAL CARBONATION***

**SHARIFAH NUR MUNIRAH BINTI SYED HASAN**

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By

**SHARIFAH NUR MUNIRAH BINTI SYED HASAN**

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia,  
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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Doctor of Philosophy

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**November 2020**

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**Faculty : Forestry and Environment**

The process of extracting minerals from mining operation emits high carbon dioxide emission in the atmosphere. However, large quantities of waste materials produced from the mining operation can be utilized for carbon sequestration by mineral carbonation process. Therefore, this study was conducted to; (1) evaluate the potential characteristics of mining wastes such as gold, limestone and iron ore mine wastes for carbon sequestration; (2) enhance mineral carbonation process at varying particle size, temperature and pH in sequestering more carbon dioxide in carbonate form and; (3) develop potential application of mining wastes for long term carbon storage in brick production. Rock, soil, sludge and sediment samples were collected and analyzed for their characteristics including pH, particle-size distribution, mineralogical composition, morphological structure and chemical composition by integrating X-ray diffraction, scanning electron microscopy and energy dispersive X-ray analyses. The mineral carbonation experiment was conducted using mining waste at different particle size, temperature and pH. Brick production incorporating mining waste was produced at different mix design ratio and the effects of carbonation time and curing periods on carbon dioxide uptake were measured. Findings suggest that gold mine was identified as the source of MgO and Fe<sub>2</sub>O<sub>3</sub> due to the presence of magnesium-iron silicate minerals; limestone mine as the source of CaO due to high availability of calcium-bearing mineral; and iron mine contains iron-calcium-magnesium silicate minerals as the source of Fe<sub>2</sub>O<sub>3</sub>, CaO and MgO that can be used as feedstock for mineral carbonation process. Iron mining waste was further evaluated for mineral carbonation due to variety of potential minerals and has the highest average divalent cation content. The effect of mineral carbonation using iron mining waste shows that smaller size particles (<38 µm) have achieved a higher calcium, iron and magnesium carbonation efficiency of 3.81%, 6.66% and 6.43%, respectively. As the temperature increased at 200°C, the maximum calcium, iron and magnesium carbonation efficiency of 4%, 5.82% and 5.62%, respectively were obtained. Increasing the pH at pH 12 resulted in greater calcium, iron and magnesium carbonation efficiency of 5.56%, 5.85% and 5.83%, respectively. Acceptable carbonation efficiency was achieved under the favorable

conditions of ambient pressure. The incorporation of different types of mine waste indicates good durability of bricks, where limestone mine waste bricks have reduced water absorption and improved compressive strength of up to 0.52% and 40.23 N/mm<sup>2</sup>, respectively. Iron mine waste bricks show higher carbon dioxide uptake averaging 0.63%. Various mix design ratio and curing period are the most significant factors that affect the water absorption of carbonated brick specimens, while carbonation time had increased the compressive strength of brick specimens. Low carbon dioxide uptake can be improved by increasing the percentage of mining waste used up to 60% and lengthening the carbonation time up to 3 hours. Therefore, utilization of mining wastes as feedstock for mineral carbonation process can be regarded as a solution for waste minimization issue and seems to be an environmentally beneficial approach in reducing carbon dioxide emissions. This would be useful in promoting sustainable use of natural resources and for future mitigation strategies of mining-related issues.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

## **PENYERAPAN KARBON DARIPADA SISA LOMBONG DALAM MENGURANGKAN PELEPASAN KARBON DIOKSIDA MELALUI MINERAL PENGKARBONATAN**

Oleh

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Proses penggalian mineral dari aktiviti perlombongan membebaskan gas karbon dioksida yang tinggi di atmosfera. Walaubagaimana pun, sejumlah besar bahan buangan yang dihasilkan dari operasi perlombongan dapat digunakan untuk penyerapan karbon melalui proses pengkarbonan mineral. Oleh itu, kajian dilakukan untuk; (1) menilai potensi ciri-ciri sisa perlombongan seperti emas, batu kapur dan bijih besi untuk penyerapan karbon; (2) meningkatkan proses pengkarbonan mineral pada ukuran saiz zarah, suhu dan pH yang berbeza untuk penyerapan lebih banyak karbon dioksida dalam bentuk karbonat dan; (3) mengembangkan potensi penggunaan sisa perlombongan untuk penyimpanan karbon dalam jangka panjang dalam pembuatan batu bata. Sampel batu, tanah, enapcemar dan sisa mendapan dikumpulkan dan ciri-cirinya seperti pH, taburan saiz zarah, komposisi mineral, struktur morfologi dan komposisi kimia dianalisis menggunakan sinar-X difraksi, pengimbasan mikroskop electron dan analisis sinar-X penyebaran tenaga. Eksperimen pengkarbonan mineral telah dilakukan menggunakan sisa perlombongan pada saiz zarah, suhu dan pH yang berbeza. Pembuatan batu bata menggunakan sisa perlombongan telah dihasilkan pada nisbah campuran yang berbeza serta sifat fizikal dan mekanikal ditentukan. Kesan masa pengkarbonan dan tempoh pengeringan terhadap pengambilan karbon dioksida telah diukur. Penemuan menunjukkan lombong emas merupakan sumber MgO dan  $\text{Fe}_2\text{O}_3$  kerana kehadiran mineral magnesium-besi silikat; lombong batu kapur sebagai sumber CaO kerana mengandungi mineral kalsium yang tinggi; dan lombong bijih besi mengandungi mineral besi-kalsium-magnesium silikat sebagai sumber  $\text{Fe}_2\text{O}_3$ , CaO dan MgO yang berpotensi sebagai bahan mentah untuk proses pengkarbonan mineral. Sisa perlombongan besi dinilai lebih lanjut untuk pengkarbonan mineral kerana pelbagai potensi mineral dan mempunyai kandungan kation divalen tertinggi. Kesan pengkarbonan mineral menggunakan sisa perlombongan menunjukkan bahawa saiz zarah yang lebih kecil ( $<38 \mu\text{m}$ ) telah mencapai 3.81% kecekapan pengkarbonatan kalsium, 6.66% kecekapan

pengkarbonatan besi dan 6.43% kecekapan pengkarbonatan magnesium yang lebih tinggi. Apabila suhu meningkat pada 200°C, kecekapan pengkarbonatan kalsium, besi dan magnesium diperoleh pada tahap maksimum masing-masing sebanyak 4%, 5.82% dan 5.62%. Peningkatan pH kepada pH 12 menghasilkan kecekapan pengkarbonatan kalsium, besi dan magnesium yang lebih tinggi masing-masing sebanyak 5.56%, 5.85% dan 5.83%. Kecekapan pengkarbonatan yang diterima dicapai dalam keadaan tekanan persekitaran yang baik. Penggunaan pelbagai jenis sisa lombong di dalam batu bata menunjukkan daya tahan yang baik dari segi sifat fizikal dan mekanikal, di mana bata sisa lombong batu kapur berupaya mengurangkan penyerapan air menjadi 0.52% serta meningkatkan kekuatan mampatan pada 40.23 N/mm<sup>2</sup>. Batu bata sisa lombong bijih besi menunjukkan pengambilan karbon dioksida yang mengandungi purata lebih tinggi sebanyak 0.63%. Nisbah campuran yang berbeza dan tempoh pengeringan adalah faktor penting yang mempengaruhi penyerapan air spesimen bata berkarbonat, sementara waktu pengkarbonan dapat meningkatkan kekuatan mampatan spesimen bata. Pengambilan karbon dioksida yang rendah dapat ditingkatkan dengan meningkatkan peratusan penggunaan sisa perlombongan sebanyak 60% dan masa pengkarbonatan yang lebih lama iaitu selama 3 jam. Oleh itu, penggunaan sisa perlombongan sebagai bahan mentah untuk proses pengkarbonan mineral merupakan penyelesaian untuk masalah pengurangan sisa dan merupakan pendekatan yang bermanfaat untuk alam sekitar dalam mengurangkan pelepasan karbon dioksida. Inisiatif ini juga dapat mempromosikan penggunaan sumber asli secara lestari serta digunakan untuk strategi mitigasi yang berkaitan dengan perlombongan.

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## LIST OF ABBREVIATIONS

Al <sub>2</sub> O <sub>3</sub>	Aluminium oxide
AMD	Acid mine drainage
BOF	Basic-oxygen-furnace
BP	Borrow pit
BS	British Standard
Ca <sup>2+</sup>	Calcium
CaO	Calcium oxide
CaCO <sub>3</sub>	Calcium carbonate
Ca(OH) <sub>2</sub>	Calcium hydroxide
CASH	Calcium aluminosilicate hydrate
CCS	Carbon capture and storage
C-S-H	Calcium-silicate-hydrate
CO <sub>2</sub>	Carbon dioxide
EAF	Electric-arc-furnace
EDX	Energy dispersive X-ray
EDTA	Edetic acid
EMS	Environmental management system
EPA	Environmental Protection Agency
Fe <sup>2+</sup>	Iron
FeCO <sub>3</sub>	Iron carbonate
FeO	Iron oxide
Fe <sub>2</sub> O <sub>3</sub>	Iron (III) oxide
FESEM	Field emission scanning electron microscopy
GHG	Greenhouse gases
H <sup>+</sup>	Hydrogen
HCl	Hydrochloric acid
HDPE	High density polyethylene
HG	High grade
HNO <sub>3</sub>	Nitric acid
H <sub>2</sub> O <sub>2</sub>	Hydrogen peroxide
IEA	International Energy Agency
IPCC	Intergovernmental Panel on Climate Change
LG	Lower grade
LOI	Loss of ignition
Mg <sup>2+</sup>	Magnesium
MgCO <sub>3</sub>	Magnesium carbonate
MgO	Magnesium oxide
Mg(OH) <sub>2</sub>	Magnesium hydroxide
MgSiO <sub>3</sub>	Magnesium silicates
MnO	Manganese (II) oxide
MS	Malaysian Standard
NaCl	Sodium chloride
NaHCO <sub>3</sub>	Sodium bicarbonate
NaOH	Sodium hydroxide
NH <sub>4</sub> OH	Ammonium hydroxide
NOAA	National Oceanic and Atmospheric Administration
OP1	Open pit 1



OP2	Open pit 2
OPC	Ordinary Portland cement
PET	Polyethylene terephthalate waste
PID	Proportional-integral-derivative
PMMA	Polymethylmethacrylate
POFA	Palm oil fuel ash
RCA	Recycle concrete aggregate
SE	Secondary electron
SEM	Scanning electron microscopy
SiO <sub>2</sub>	Silica
SLG	Super lower grade
SOM	Soil organic matter
TGA	Thermogravimetric analysis
USDA	United States Department of Agriculture
XRD	X-ray diffractogram
XRF	X-ray fluorescence

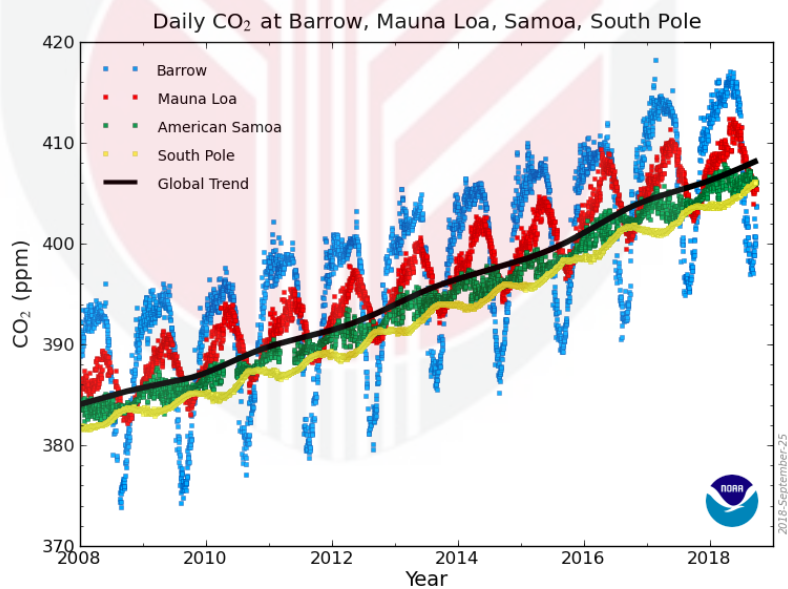
## CHAPTER 1

### INTRODUCTION

#### 1.1 Background of the Research

##### 1.1.1 Global Issue on Carbon Dioxide Emissions

Emissions of carbon dioxide (CO<sub>2</sub>) are likely to increase due to global energy demand and economic growth throughout the year. High concentration of anthropogenic CO<sub>2</sub> emission is becoming a global issue because of CO<sub>2</sub> is the major gas which causes high greenhouse gases (GHGs) accumulation in the atmosphere and lead to climate change (Sipilä et al., 2008). Furthermore, global atmospheric CO<sub>2</sub> concentration has risen every year in the last 10 years, where the highest increment is 3.4 ppm from 399.41 ppm in 2015 to 402.81 ppm in 2016 (NOAA, 2018) (Figure 1.1).



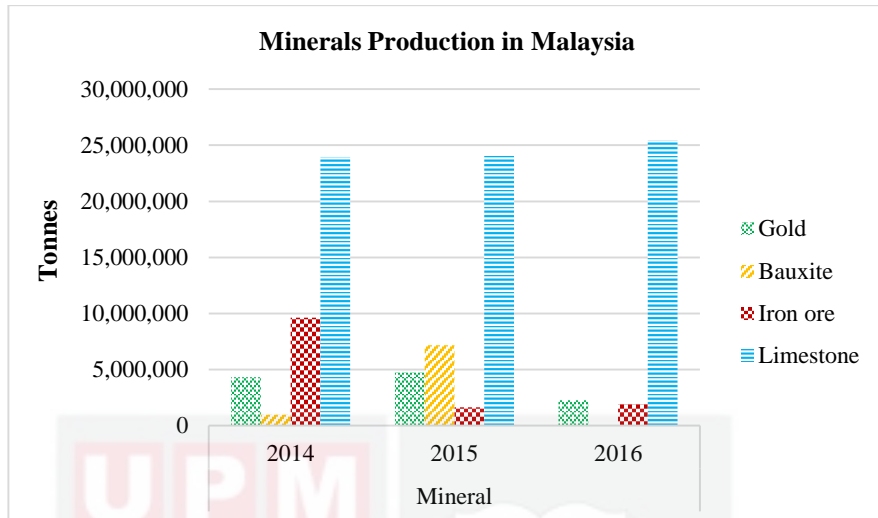
**Figure 1.1 : Trend line shows global average CO<sub>2</sub> levels. The line symbolize the U.S. State: South Pole, Antartica (yellow), American Samoa (green), Mauna Loa, Hawaii (red) and Barrow, Alaska (blue). The average of the smoothed curves for each year is shown in black line. [Adopted from National Oceanic and Atmospheric Administration (2018)]**



GHG emission and climate change has become one of the environmental issues that is caused by the increase in CO<sub>2</sub> emission from human activities such as fossil fuel burning, transportation and industrial activities. The release of anthropogenic GHG into the atmosphere consists of carbon dioxide, nitrous oxide, methane, water vapour and ozone. The GHG emission especially CO<sub>2</sub> is the main driver of the current global warming issue that contributes about 6,511 million metric tons of total CO<sub>2</sub> equivalent emission in 2016 (EPA, 2016; Muhd-Nor et al., 2016). Southeast Asian countries such as Malaysia, Thailand, Indonesia, Brunei, Singapore, Philippines, Vietnam, Myanmar, Laos, and Cambodia are also affected by global warming, where combustion of fossil fuel is the major activity which contributes in increasing CO<sub>2</sub> discharge into the atmosphere (Muhd-Nor et al., 2016; Wilson et al., 2009). Besides, GHG emissions also come from deforestation activity, where the trees are cut down and burned for development and agriculture purposes. Other than that, transportation, industrial activities and land-use changes are also leading to the increase of CO<sub>2</sub> emissions. Thus, the human activities have a significant effect on the climate on the earth.

To inhibit global climate change by 2050 as recommended by the International Energy Agency (IEA), a decrease of approximately 13% of the accumulated CO<sub>2</sub> emission per year is required. In Malaysia, release of CO<sub>2</sub> is at a current level of 257.69 million tons (Mt) in 2014 and is expected to increase to 12.1 tons of CO<sub>2</sub> emission per capita by 2020 (Zaid et al., 2015). Malaysia has targeted a 45% decline in GHG emissions by the year 2030, as reported in the policy commitment (Ministry of Energy and Natural Resources, 2019), whereas Indonesia and Singapore are expected to decrease GHG emissions by 29% and 36% respectively by 2030 (Muhd-Nor et al., 2016). Therefore, an effective approach is needed to achieve the goal of reducing CO<sub>2</sub> emission in the future.

The mining industry contributes for about 21% of global anthropogenic CO<sub>2</sub> emissions (IPCC, 2014), hence proper mitigation strategies are required in the mining sector to reduce CO<sub>2</sub> emissions for long term. Metallic minerals (e.g., iron ore, gold, and bauxite) and non-metallic mineral including limestone, are the main minerals production in Malaysia which provide high economic value to mining industry (Figure 1.2) (Department of Mineral and Geoscience Malaysia, 2018). As a result, the increase in minerals demand lead to subsequent increase in air pollutants such as CO<sub>2</sub> in the environment. For instance, metal ore extraction from an open pit mine will normally release large quantities of CO<sub>2</sub> into the atmosphere (Pandey et al., 2017). Mining industry faces major challenge for reducing the air pollutants emissions in the atmosphere and thus, requires an effective mitigation in controlling global warming.



**Figure 1.2 : The production of minerals in Malaysia of metallic minerals (e.g., gold, bauxite, iron ore) and non-metallic mineral (e.g., limestone) from 2014 to 2016. [Source from Department of Mineral and Geoscience Malaysia (2018)]**

### 1.1.2 Carbon Dioxide Emission from Mining Activities

Mining industry is among the main sources of environmental pollution such as through the release of anthropogenic CO<sub>2</sub> emissions into the atmosphere and alteration of land that leads to soil contamination. This is because mining activities releases various toxic contaminants into the soil and environment at different stages of its commissioning from mining to metal cleaning, transportation, and finally to its disposal as end product. In general processes, mining activities such as the extraction and materials processing that remove rocks and soils from open pit mine to obtain metal ore will normally release excessive amounts of CO<sub>2</sub> that affects anthropogenic GHG accumulation in the atmosphere (Pandey et al., 2017). During surface mining activities, the land would suffer several alterations that will lead to environmental contamination. Mining operations such as collection of valuable topsoil using scrapers and bulldozers and land cleaning for vegetation clearance, potentially destroying flora and fauna and causing plant and soil quality destruction, and also more dust release in the surrounding area (Tabatabaei and Mohammadi, 2013).

Removal of top soils is the basic operation involved in mining processes resulting in the removal of seed bank and root stocks, reduction of organic matter and nutrient contents, modification of soil texture and structure, and severe deterioration in quality of soil (Mensah, 2014). Minerals extraction is achieved through drilling and blastings activities after clearance of the topsoil and vegetation. The drilling process produces dust that comes from rock crushing and grinding activities (Pandey et al., 2017). The effect of drilling process contributes to the conveyance and spread of pollutants into the environment, and as well as the source of dust (Mandal et al., 2012). The explosion emitted CO<sub>2</sub>, nitrogen, and water that are considered a major

gaseous material in the ultimate environment. The topsoil is transported via the haul road for dumping involving excavator, scraper, loader, and dumper after drilling and blasting process, which lead to large emission of small particulates from the overburden substances (Tabatabaei and Mohammadi, 2013). The most plentiful sources of dust in open-cast mines are strongly released from vehicle traffic during mining operations. Therefore, various stages of mining operations such as land cleaning, drilling and blasting, crushing and grinding of rocks, filling, dropping, and road transport produce a lot of gaseous pollutants that may lead to accumulation of anthropogenic GHG and soil contamination in surrounding areas.

### **1.1.3 Carbon Sequestration Technique in Reducing Carbon Dioxide Emission**

Several techniques for reducing CO<sub>2</sub> emissions have been utilized particularly from the mining sector. Among them, CO<sub>2</sub> sequestration or carbon capture and storage (CCS) by mineral carbonation is the most effective method that can help in decreasing 20% of CO<sub>2</sub> emission (Benson and Cole, 2008) by permanently storing CO<sub>2</sub> in solid carbonates (Arce et al., 2017; Jorat et al., 2015a; Jorat et al., 2015b; Jorat et al., 2017; Lackner et al., 1995; Li and Hitch, 2017; Manning, 2008; Manning et al., 2013; Moosdorf et al., 2011; Renforth et al., 2009; Renforth, 2011; Renforth et al., 2011; Sipilä et al., 2008; Washbourne et al., 2015). This process occurs when the CO<sub>2</sub> from the gas flows is stored in rock and soil (Assima et al., 2013a; Harrison et al., 2013a; Wilson et al., 2009). For instance, natural processes that consist CO<sub>2</sub> chemical reaction to form carbonic acid and bicarbonate and the mixing of atmospheric gases into the oceans through physical exchange (Power et al., 2013) are highly preferred because it somehow can lower the environmental impact. Sequestering CO<sub>2</sub> in soils or other materials containing calcium content and the availability of CO<sub>2</sub> in the substrate are important factors in the carbonates formation, where this process is considered as passive CO<sub>2</sub> sequestration (Manning et al., 2013; Jorat et al., 2018). In this study, passive CO<sub>2</sub> sequestration is applied which involves the uptake of CO<sub>2</sub> uptake by mineral carbonation reaction from carbonates minerals formed in the rocks and soils to mitigate CO<sub>2</sub> in the atmosphere.

Waste materials or residues are produced by extraction and materials process in a form of gases, solids, and water from mining operations. This process increases the GHG emission as the CO<sub>2</sub> is released into the atmosphere (Assima et al., 2014b; Harrison et al., 2013b), and generates greater amount of waste that has small or no economic benefit. Nevertheless, mining wastes in a constructive way can help reduce GHG emission to the atmosphere because of its ability to store CO<sub>2</sub>. In order to reduce CO<sub>2</sub> in the atmosphere, passive CO<sub>2</sub> sequestration process by mineral carbonation is needed which can trap CO<sub>2</sub> in stable carbonates (Assima et al., 2013a; Assima et al., 2014c; Wilson et al., 2009). Reaction between CO<sub>2</sub> with ions such as calcium (Ca<sup>2+</sup>), magnesium (Mg<sup>2+</sup>), and hydrogen (H<sup>+</sup>) forming carbonate or bicarbonate that captured carbon by weathering of primary minerals (Assima et al., 2014b; Assima et al., 2014c; Lechat et al., 2016; Power et al., 2013; Renforth, 2011; Wilson et al., 2009). This natural carbonation process offers huge potential as CO<sub>2</sub> storage to reduce GHG emission for mining waste. Moreover, mining residues from various types of mining processes could potentially sequester CO<sub>2</sub> and provides

economic values with the least technology costs (Assima et al., 2012; Assima et al., 2013a; Lechat et al., 2016). Large amount of mining wastes which contains plenty of Ca-Fe-Mg have a potential be used as a feedstock for passive CO<sub>2</sub> sequestration. Thus, mining waste materials can be used as a feedstock for carbon sequestration for long-term reduction of CO<sub>2</sub> emissions.

From another point of view, CO<sub>2</sub> storage in construction industry has also been explored in a more recent year. Industrial activities such as cement production, steel making and other construction material production also emit high CO<sub>2</sub> into the atmosphere. Approximately 5% of global CO<sub>2</sub> emissions come from the production of cement (Khan et al., 2018). Nevertheless, recovered CO<sub>2</sub> can be recycled as a curing agent in the production of carbonated concrete products to minimize CO<sub>2</sub> emissions for long term (El-Hassan and Shao, 2014). The concept of carbonation curing process is almost similar to the mineral carbonation process, where CO<sub>2</sub> is stored in the form of thermodynamically stable CaCO<sub>3</sub> in the concrete product. Application of CO<sub>2</sub> storage in construction industry has been discovered in cement-bonded cellulose fiberboards (He et al., 2019), concrete block (El-Hassan and Shao, 2014) and mortar mixes incorporating cement kiln dust (CKD) (Sharma and Goyal, 2018) in sequestering CO<sub>2</sub> in concrete product. Furthermore, the use of waste materials will serve as additional binder in carbonation process that can increase CO<sub>2</sub> uptake and improve the properties of the carbonation byproduct. Since mining waste materials have potential for carbon sequestration, further utilization of it for the manufacture of bricks can act as a permanent CO<sub>2</sub> storage technique and at the same time enhance the durability properties of carbonated products that can be used for construction purposes.

## **1. 2 Problem Statements**

The population growth, high income and rapid growth of urban cities in developing countries have causes several environmental issues such as global warming. Human activities have contributed to the increase in atmospheric anthropogenic CO<sub>2</sub> emissions and can lead to climate change. Global warming and climate change have potential effects on the environment such as severe climate (e.g., storm, floods, and droughts), sea level rise and altered crop growth. Malaysian economy is evolving rapidly in that most of the CO<sub>2</sub> emissions continue to increase as the Malaysian economy develops. The increasing population and environmental deterioration are becoming major obstacles in achieving 40% CO<sub>2</sub> reduction target by the year 2020 (Shahid et al., 2014), where mining industry is among the significant contributor to climate change. Thus, an effective mitigation is needed in monitoring CO<sub>2</sub> emission from mining industry for long term.

Mining activities yield large quantities of waste materials which are typically stored in tailings and waste dump at the mining site. Furthermore, the large quantities of waste at mining site if are not well-managed, can be harmful to the environment and human health. However, mineral wastes from mining have a potential to sequester CO<sub>2</sub> permanently because these minerals are able to trap CO<sub>2</sub> from the atmosphere, when they are exposed to the atmosphere and rain water. Thus, the potential

utilization of mining waste for carbon sequestration is required in reducing CO<sub>2</sub> in the atmosphere.

Carbon sequestration or CCS is the technology used to reduce CO<sub>2</sub> emission, which is an environmentally sound approach of storing CO<sub>2</sub> permanently in carbonates form. This technology is widely applied to alkaline wastes from industry such as cement kiln dust, steel slags, fines from aggregate production and construction waste (Renforth et al., 2011). Carbon sequestration is also important for mining industry because they must reduce CO<sub>2</sub> emission and at the same time explore the potential of carbon sequestration through utilization of mining waste. However, less research on CCS through mineral carbonation is applied in Malaysia, especially in mining industry. Thus, the current research is emphasized on the discovery of the mining waste potential for carbon sequestration in mitigating GHG emission for long term.

### **1.3 Objectives of the Study**

The aim of this research is to evaluate the potential of carbon sequestration from mining waste to reduce CO<sub>2</sub> emissions in the atmosphere through mineral carbonation. The underlying objectives are expanded below.

1. To evaluate the characteristics of different types of mining wastes for carbon sequestration.
  - (i) To characterize the mineral phases and chemical composition of the mining wastes for carbon sequestration.
  - (ii) To identify the presence of natural silicate and/or carbonate minerals in mining wastes as the feedstock for the mineral carbonation process.
2. To enhance mineral carbonation process in sequestering more CO<sub>2</sub> in carbonate form.
  - (i) To determine the effect of particle size fraction, temperature and pH on mineral carbonation process from mining wastes.
  - (ii) To measure the carbonation efficiency from the mineral carbonation process using mining wastes.
3. To develop potential application of mining wastes in construction industry for long term carbon storage.
  - (i) To evaluate the physical and mechanical properties of mining waste bricks in carbon storage application.
  - (ii) To assess the potential carbon capture and storage of mining waste bricks.
  - (iii) To determine the effect of carbonation curing on brick properties.



## **1.4 Research Questions**

Waste materials from mining operations may have potential for carbon sequestration in reducing CO<sub>2</sub> emissions by enhancing the carbonation reaction and utilizing it for long term carbon storage. The underlying research questions are as follows:

1. What are the characteristics of mining waste suitable for carbon sequestered to reduce CO<sub>2</sub> emission?
2. What are the mineral and chemical composition of the mining wastes that can be influential for carbon sequestration?
3. What are the natural silicate and carbonate minerals present in mining wastes that can be utilized as feedstock for the mineral carbonation process?
4. What are the factors that influence mineral carbonation process from mining waste to sequester more CO<sub>2</sub>?
5. Does the particle size fraction, temperature and pH affect the mineral carbonation process from mining wastes?
6. How much is the carbonation efficiency from the mineral carbonation process using mining wastes?
7. How can carbonate minerals from mineral carbonation process of mining waste be utilized for permanent carbon storage?
8. What are the physical and mechanical properties of mining waste bricks? Does the bricks meet the standard specification for construction purposes?
9. How much is the CO<sub>2</sub> uptake of bricks containing mining waste?
10. Does the carbonation curing of mining waste bricks affect the properties of the brick product?

## **1.5 Significance of the Study**

Passive carbon sequestration from mining waste provides potential solution in storing CO<sub>2</sub> permanently for long term. Carbon mineralization is an approach for determining the potential of carbon capture of mining waste in sequester CO<sub>2</sub> in stable carbonates. Potential use of silicate minerals for carbon uptake through mineral carbonation can enhance carbon sequestration from mining waste. Furthermore, passive CO<sub>2</sub> sequestration of mineral carbonation provides the potential for carbon footprint evaluation in mining industry in Malaysia. The process of trapping and storing CO<sub>2</sub> can help stabilize concentrations of atmospheric CO<sub>2</sub> that would otherwise be released into the atmosphere. CCS is an attractive solution that is capable of meeting demand for CO<sub>2</sub> reduction (Li and Hitch, 2017). The use of this technology may provide long term benefits to the environment through the adoption of carbon sequestration technique.

Mine wastes, instead of being regarded as waste materials have a potential for atmospheric carbon sink in reducing CO<sub>2</sub> emission to the atmosphere. Current research has explored the mining waste potential from metal mining industry such as iron ore, gold; and non-metal industry including limestone because they produce

large amount of waste which may be used as a feedstock for carbon sequestration process. Besides, they are the main contributor to country's mineral sector and the availability of Mg-Ca-Fe-silicate minerals in mining waste makes it favorable for carbonates reactions. Mine waste samples from tailings, stockpiles, and waste dump are investigated in this study because these wastes may provide long term atmospheric CO<sub>2</sub> storage in order to reduce GHG emission to the atmosphere. Utilization of mining waste as carbonates and value added by-product from carbonation process, have the potential to be used as green building product for permanent CO<sub>2</sub> storage. In this research, waste materials from gold, limestone and iron ore mining are utilized as raw materials in brick production, where CO<sub>2</sub> is being captured and stored from escaping into the atmosphere.

## **1.6 Scope of the Study**

This study focuses on the potential of mining waste such as the waste materials from gold, limestone and iron ore mining for carbon sequestration in mitigating climate change. Samples of mine wastes consisting of rock, soil, sludge and sediment were obtained from active mine sites, which are discovered in Pahang and Perak states. Data from mining waste will help describe its mineralogy and chemical composition in association with its potential as feedstock for mineral carbonation. Direct aqueous mineral carbonation experiment of mining waste is performed using different particle size fraction, pH and temperature to sequester more CO<sub>2</sub> in carbonate form. The carbonation efficiency of various types of mining waste are evaluated. In order to capture CO<sub>2</sub> permanently, mining waste is further utilized to develop bricks product. The effect of brick properties such as density, water absorption and compressive strength using various proportion of mining waste, which is 40%, 50% and 60% are evaluated to produce good quality bricks for construction purposes that satisfy the standard engineering requirements. The CO<sub>2</sub> uptake of mining waste bricks is identified and the influence of carbonation time and curing periods on the brick properties are explored.

## **1.7 Organization of the Thesis**

Chapter one focuses on the general issues on CO<sub>2</sub> emissions and application of carbon sequestration technology in reducing CO<sub>2</sub>. This chapter also provides the statement of the problems, the objectives of the study, significance of the study, scope of the study and the outlines of the thesis. Chapter two provides a review of the literature on carbon sequestration and mineral carbonation. This includes the advantages and disadvantages of carbon sequestration, classification of mining wastes and its potential as a feedstock for carbon sequestration, and potential by-product from mineral carbonation using mining waste for industrial use. Chapter three explains the details of the study area including geological history of mining site, field sampling, sample collection and sample preparation for different types of waste. Sample analysis was further describes using X-ray diffractogram (XRD), scanning electron microscopy (SEM), energy dispersive X-ray (EDX) and thermogravimetric analysis (TGA). Mineral carbonation experiments using the mining waste were further described. The methodology for the production of bricks

using different mixture proportion of mining waste for long-term carbon storage was also explained in detail, including its physical and mechanical properties and the carbonation curing procedure. Chapter four discusses the results of the characterization of mining waste such as pH, particle-size distribution, mineral and chemical composition as feedstock for mineral carbonation, where the first objective was achieved. The mineral carbonation process using silicate minerals from mining wastes were also discussed. Findings from mineral carbonation experiment were discussed on the effects of particle size, temperature and pH on carbonation efficiency, achieving the second objectives. The physical and mechanical properties of bricks containing mining wastes in terms of density, water absorption and compressive strength were described and was compared with the standard specification of bricks for construction purposes. The potential of carbon capture and storage of mining waste bricks was also explained. The effect of CO<sub>2</sub> uptake and carbonation of bricks properties were discussed in detail, achieving the third objectives. Last but not least, Chapter five summarizes the conclusions and implications from the study and provides recommendations for the future studies.



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## BIODATA OF STUDENT

Sharifah Nur Munirah binti Syed Hasan (GS46860) was born on 15<sup>th</sup> June 1990 at Hospital Besar Alor Setar, Kedah. She lived in Alor Setar, Kedah for 14 years and moved to Kangar, Perlis in year 2005. She is currently residing in Kangar, Perlis. She has one elder brother, two elder sister and a younger brother. Her parents are government pensioner. She has completed her primary education from year 1997 to 2002 at Sekolah Kebangsaan Taman Uda, Alor Setar, Kedah and her secondary education from year 2003 to 2004 at Sekolah Menengah Kebangsaan Sultanah Asma, Alor Setar, Kedah, and moved to Sekolah Menengah Kebangsaan Derma, Kangar, Perlis from year 2005 to 2007. She continued her studies in Matriculation (Biological Science) at Kolej Matrikulasi Perlis (KMP) from 2008 to 2009 (1 year) after the Sijil Pelajaran Malaysia (SPM) examination. She obtained Bachelor of Applied Science (Hons) in Biodiversity Conservation and Management from Universiti Malaysia Terengganu (UMT) in year 2012. Then, she further her study in Master of Environment (coursework) in Environmental Science at Universiti Putra Malaysia (UPM) for 2 years and graduated in year 2015. Currently, she is a Doctor of Philosophy (PhD) student in Environmental Pollution Control Technology (Carbon Sequestration) at Universiti Putra Malaysia (UPM) since year 2016 and will be graduating soon in year 2021 at the age of 31.



## LIST OF PUBLICATIONS

### Journal

**Syed-Hasan, S.N.M.\***, Kusin, F.M., Jusop, S., Mohamat-Yusuff, F. (2018). Potential of soil, sludge and sediment for mineral carbonation process in Selinsing gold mine, Malaysia. *Minerals*, 8: 257–270 (**ISI; IF: 2.250; Q2**).

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