

**INVESTIGATION OF PHYSICAL AND  
CHEMICAL PROPERTIES OF SPENT  
BLEACHING CLAY BASED GEOPOLYMERS  
USING ALKALINE ACTIVATED  
GEOPOLYMERIZATION PROCESS**

**MAK WEI YING**

**UNIVERSITI SAINS MALAYSIA**

**2023**

**INVESTIGATION OF PHYSICAL AND  
CHEMICAL PROPERTIES OF SPENT  
BLEACHING CLAY BASED GEOPOLYMERS  
USING ALKALINE ACTIVATED  
GEOPOLYMERIZATION PROCESS**

by

**MAK WEI YING**

**Thesis submitted in fulfillment of the requirements  
for the Degree of  
Master of Science**

**September 2023**

## ACKNOWLEDGEMENT

First and foremost, I would like to express my heartfelt gratitude to my main supervisor, ChM Dr. Shangeetha Ganesan, for the unwavering guidance, invaluable patience, and motivation throughout my research. This endeavor would not have been possible without her understanding and vast knowledge with dedicated involvement in every step throughout the process. At the same time, words cannot express my deepest appreciation to my co-supervisor, TS. Dr. Mastura binti Azmi from the School of Civil Engineering for the insightful remarks and suggestions, which impelled me to gain experience and broaden my research from multiple perspectives.

My sincere appreciation to Universiti Sains Malaysia for the financial support from the short-term research grant 304/PKIMIA/6313335 and RUI grant 1001/PKIMIA/8011137 that allowed me to undergo the research smoothly.

Getting through my research required more than academic support, and I have many people to thank for listening to and, at times, having to tolerate me over the past few years. Special thanks to my fellow lab mates, especially Siow Hao Sen, for his late-night feedback sessions, moral support, and stimulating discussions. I take this opportunity to express gratitude to all of the members of School of Chemical Sciences and School of Civil Engineering for their assistance and cooperation throughout the research, direct or indirectly.

Last but not least, my gratitude would be incomplete without mentioning my family, especially my parents as well as my sister for their unfailing support, continuous encouragement, and understanding throughout the years. This accomplishment would not have been possible without them.

## TABLE OF CONTENTS

<b>TABLE OF CONTENTS</b> .....	<b>iii</b>
<b>LIST OF TABLES</b> .....	<b>vi</b>
<b>LIST OF FIGURES</b> .....	<b>vii</b>
<b>LIST OF ABBREVIATIONS, SYMBOLS, AND UNITS</b> .....	<b>viii</b>
<b>ABSTRAK</b> .....	<b>xi</b>
<b>ABSTRACT</b> .....	<b>xiii</b>
<b>CHAPTER 1 INTRODUCTION</b> .....	<b>1</b>
1.1 Research Background.....	1
1.2 Problem statement.....	3
1.3 Research objective .....	4
1.4 Scope of research .....	5
1.5 Significance of research .....	5
1.6 Overview of thesis.....	6
<b>CHAPTER 2 LITERATURE REVIEW</b> .....	<b>7</b>
2.1 Aluminosilicate materials.....	7
2.1.1 Kaolin clay .....	7
2.1.2 Montmorillonite clay.....	8
2.1.3 Other types of clay .....	10
2.2 Spent bleaching clay (SBC) .....	11
2.2.1 SBC as problematic waste.....	11
2.2.2 Recycling and reutilizing of SBC .....	12
2.3 Alkaline activator solution .....	13
2.3.1 Sodium vs Potassium .....	13
2.3.2 Combination of alkaline activator solution .....	15
2.3.3 Solid vs Liquid alkaline activator .....	17

2.4	Geopolymer.....	18
2.4.1	Ordinary Portland Cement (OPC).....	19
2.4.2	Advantages of geopolymer .....	20
2.4.3	Clay-based geopolymer using different types of clay.....	21
2.5	Factors affecting the different properties of geopolymer.....	28
2.5.1	Concentration of alkaline activator solution .....	33
2.5.2	Solid: Liquid ratio (S/L ratio) .....	35
2.5.3	Si/Al ratio.....	36
2.5.4	Curing conditions.....	38
2.6	Recent progress and applications in clay-based geopolymer.....	40
<b>CHAPTER 3 MATERIALS AND METHODOLOGY.....</b>		<b>45</b>
3.1	Materials and Chemicals .....	45
3.2	Synthesis of clay-based geopolymers .....	46
3.2.1	Optimization of geopolymerization reaction .....	46
3.2.2	Spent bleaching clay-based geopolymer (SBCG).....	47
3.2.3	Calcium silicate-spent bleaching clay-based geopolymer .....	48
3.3	Characterization of spent bleaching clay and geopolymers.....	49
3.3.1	Fourier Transform Infrared Spectroscopy (FTIR) .....	49
3.3.2	Scanning Electron Microscopy coupled with Energy Dispersive X-ray (SEM/EDX) .....	49
3.3.3	Thermogravimetric analysis (TGA).....	49
3.3.4	N <sub>2</sub> Brunauer-Emmett-Teller (BET) analysis.....	50
3.3.5	X-Ray Diffraction (XRD) analysis .....	50
3.3.6	Compressive strength test .....	50
<b>CHAPTER 4 RESULTS AND DISCUSSION .....</b>		<b>52</b>
4.1	Optimization of geopolymerization reaction .....	52
4.1.1	Spent bleaching clay-based geopolymer (SBCG).....	52

4.1.2	Calcium silicate-spent bleaching clay-based geopolymer .....	56
4.2	Characterization of SBC based geopolymer .....	58
4.2.1	Functional group analysis .....	58
4.2.2	Elemental and surface morphology analysis.....	62
4.2.3	Thermal properties .....	68
4.2.4	Surface area and pore size analysis.....	71
4.2.5	X-Ray Diffraction (XRD) analysis .....	78
4.2.6	Mechanical properties analysis .....	81
4.3	Mechanism of geopolymerization.....	84
<b>CHAPTER 5 CONCLUSION AND RECOMMENDATIONS .....</b>		<b>93</b>
5.1	Conclusion .....	93
5.2	Future recommendations.....	95
<b>REFERENCES.....</b>		<b>96</b>

**LIST OF PUBLICATION**

## LIST OF TABLES

	<b>Page</b>
Table 2.1	Summary of clay-based geopolymers produced using different types of clay and waste materials..... 25
Table 2.2	Different mix design studies of geopolymers based on different parameters ..... 30
Table 4.1	Optimization of SBCG using a different mass ratio of NaOH to Na <sub>2</sub> SiO <sub>3</sub> and mass ratio of alkaline activator solution to SBC ..... 55
Table 4.2	Optimization of calcium silicate-SBC based geopolymers using a different mass ratio of SBC to CS with reference to the previous formulation..... 57
Table 4.3	EDX analysis of SBC, SBCG, CS, CS 5, CS 10, and CS 15 at 10k magnification. .... 68
Table 4.4	BET surface area, total pore volume and average pore size of the SBC, SBCG, CS, CS 10, CS 5, and CS 15 ..... 73

## LIST OF FIGURES

	<b>Page</b>
Figure 2.1	Structural diagram of kaolinite clay mineral (Adapted and modified from Dudkin et al., 2005). ..... 8
Figure 2.2	Structural diagram of montmorillonite clay mineral (Adapted and modified from Uddin, 2018). ..... 9
Figure 2.3	Factors affecting properties of clay-based geopolymers ..... 29
Figure 3.1	General steps to produce SBCG (* For calcium silicate-SBC-based geopolymer, CS is mixed with SBC before an alkaline activator solution is added) ..... 48
Figure 3.2	Flow chart of general representation of this research ..... 51
Figure 4.1	FTIR spectrum of the SBC, SBCG, CS, CS 5, CS 10, and CS 15..... 59
Figure 4.2	SEM images of (a) SBC and (b) SBCG, respectively at 10k magnification. .... 65
Figure 4.3	SEM images of (a) CS, (b) CS 5, (c) CS 10 and (d) CS 15, respectively at 10k magnification. .... 66
Figure 4.4	TGA graph of (a) SBC and SBCG, (b) CS, CS 5, CS 10, and CS 15..... 70
Figure 4.5	BET nitrogen adsorption-desorption linear isotherm of (a) SBC, (b) SBCG, (c) CS, (d) CS 10, (e) CS 5, and (f) CS 15 ..... 77
Figure 4.6	XRD diffractogram of (a)SBC and SBCG, (b)CS, CS 5, CS 10, and CS 15 ..... 79
Figure 4.7	Mechanism of geopolymerization process to form geopolymer backbone ..... 85
Figure 4.8	Conceptual design of geopolymerization process..... 87
Figure 4.9	Different nomenclature of geopolymer..... 90



## LIST OF ABBREVIATIONS, SYMBOLS, AND UNITS

°	Degree
3D	Three Dimension
Å	Angstrom
Al	Aluminium
Al <sub>2</sub> O <sub>3</sub>	Alumina
ASTM	American Society of Testing and Materials
BET	Brunauer-Emmett-Teller
BJH	Barrett, Joyner, and Halenda method
°C	Degree Celsius
°C/min	Degree Celsius per Minute
Ca	Calcium
CaCO <sub>3</sub>	Calcium Carbonate
CaO	Calcium Oxide
cm <sup>3</sup> /g	Centimetre cube per Gram
CO <sub>2</sub>	Carbon Dioxide
CS	Calcium Silicate
Cu	Copper
EN	European Norms
Fe	Iron
Fe <sub>2</sub> O <sub>3</sub>	Iron Oxide
FTIR	Fourier Transform Infrared Spectroscopy
g cm <sup>-1</sup>	Gram per Centimetre cube
g	Gram

GEP	Gene Expression Programming
GWP	Global Warming Potential
h	Hour
H	Hydrogen
H <sub>2</sub> O <sub>2</sub>	Hydrogen Peroxide
IPCC	Intergovernmental Panel on Climate Change
IUPAC	International Union of Pure and Applied Chemistry
JCPDS	Joint Committee on Powder Diffraction Standards
K	Kelvin
K <sup>+</sup>	Potassium Ion
K <sub>2</sub> SiO <sub>3</sub>	Potassium Silicate
kg/m <sup>3</sup>	Kilogram per Meter cube
kN	Kilonewton
KOH	Potassium Hydroxide
kV	Kilovolt
M	Molarity
m <sup>2</sup> /g	Metre square per Gram
Mg	Magnesium
mg	Milligram
mg/g	Milligram per Gram
MgO	Magnesium Oxide
mL/g	Millimetre per Gram
MPa	Megapascal
N/mm <sup>2</sup>	Newton per Millimetre square
Na	Sodium

Na <sup>+</sup>	Sodium Ion
Na <sub>2</sub> SiO <sub>3</sub>	Sodium Silicate
NaOH	Sodium Hydroxide
nm	Nanometre
NMR	Nuclear Magnetic Resonance
no.	Number
OH	Hydroxide
Pa	Pascal
PC	Portland Cement
ppm	Parts Per Million
S/L	Solid to Liquid ratio
SBC	Spent Bleaching Clay
SBCG	Spent Bleaching Clay-based Geopolymer
SEM/EDX	Scanning Electron Microscopy with Energy Dispersive X-Ray
SiO <sub>2</sub>	Silica
SO <sub>3</sub>	Sulphur Trioxide
TGA	Thermogravimetric Analysis
UTS	Unit Test Scientific
wt%	Weight Percent
XRD	X-Ray Powder Diffraction
Zn	Zinc
θ	Theta

**PENJELASAN CIRI-CIRI FIZIKAL DAN KIMIA GEOPOLIMER  
BERASASKAN TANAH LIAT PELUNTUR TERPAKAI MENGGUNAKAN  
PROSES PENGAKTIFAN ALKALI GEOPOLIMERISASI**

**ABSTRAK**

Penyelidikan ini meneroka sintesis dan pencirian geopolimer berasaskan tanah liat berpori menggunakan tanah liat peluntur yang tidak dikalsinasi (SBC) sebagai kompaun yang baru. Geopolimer, yang terkenal dengan kekuatan mekanikal dan ketahanan kimianya, biasanya dihasilkan menggunakan abu terbang atau tanah liat kaolin yang dikalsin. Penyataan masalah timbul daripada potensi besar SBC, sejenis keluaran sampingan dari industri penapisan minyak kelapa sawit yang kaya dengan bahan aluminosilikat, sering dibuang di tapak pembuangan sampah. Matlamat utama kajian ini adalah untuk mengoptimumkan proses geopolimerisasi untuk geopolimer berasaskan SBC dan geopolimer berasaskan silikat kalsium-SBC. Bergantung kepada parameter reka bentuk, geopolimer boleh dihasilkan dengan pelbagai sifat dan kelebihan, seperti kekuatan yang sangat baik dan rintangan haba di bawah persekitaran berasid. Hasil kajian ini menunjukkan bahawa nisbah jisim natrium hidroksida kepada natrium sikilat sebanyak 3.50, nisbah jisim larutan pengatif alkali kepada tanah liat peluntur sebanyak 2.25, dan penggunaan 10% kalsium silikat adalah keadaan yang terbaik. Dalam kajian ini, pasta campuran geopolimer diletakkan dalam ketuhar pada suhu 80°C selama 24 jam dan pada suhu bilik selama 7 hari. Usaha pencirian merangkumi analisis yang mendalam terhadap bahan mentah, termasuk tanah liat peluntur dan kalsium silikat, dan geopolimer berpori yang dihasilkan, penilaian atas sifat struktur, mekanikal, termal, dan kimianya. Pencirian ini termasuk spektroskopi FTIR, SEM/EDX, TGA, BET, XRD, dan ujian kekuatan mampatan. Selain itu,

penyelidikan mengenai mekanisme pembentukan geopolimer menerangkan reaksi kimia rumit dan transformasi struktur yang mendasari proses tersebut. Kesimpulannya, penyelidikan ini tidak hanya memperkenalkan pendekatan baru untuk memanfaatkan semula tanah liat peluntur tetapi juga menyumbang kepada pembangunan bahan geopolimer yang mesra alam dengan pelbagai aplikasi, menekankan kepentingan pengoptimuman proses geopolimerisasi dan meningkatkan pemahaman terhadap mekanisme yang terlibat.

**INVESTIGATION OF PHYSICAL AND CHEMICAL PROPERTIES OF  
SPENT BLEACHING CLAY BASED GEOPOLYMERS USING ALKALINE  
ACTIVATED GEOPOLYMERIZATION PROCESS**

**ABSTRACT**

This research explores the synthesis and characterization of porous clay-based geopolymers using uncalcined spent bleaching clay (SBC) as a novel precursor. Geopolymers, celebrated for their mechanical strength and chemical durability, have traditionally been synthesized using fly ash or calcined kaolin clay. The problem statement arises from the extensive potential of SBC, a waste product from the palm oil refining industry rich in aluminosilicate material, which is often discarded in landfills. The study's overarching aim is to optimize the geopolymerization process for both SBC-based and calcium silicate-SBC based geopolymers. Depending on the design parameters, geopolymer can be produced with various properties and advantages, such as excellent strength and thermal resistance under acidic environment. The result showed that a mass ratio of NaOH to Na<sub>2</sub>SiO<sub>3</sub> of 3.50, mass ratio of alkaline activator solution to SBC of 2.25, and a utilization of 10% CS has the best condition. In this study, the geopolymer paste was cured in an oven at 80°C for 24h and at room temperature for 7 days. Characterization efforts encompass an extensive analysis of raw materials, including SBC and calcium silicate, and the synthesized porous geopolymers, evaluating their structural, mechanical, thermal, and chemical properties. The characterization included FTIR spectroscopy, SEM/EDX, TGA, BET, XRD, and compressive strength test. Additionally, the investigation into the mechanism of geopolymer formation sheds light on the intricate chemical reactions and structural

transformations underpinning the process. In conclusion, this research not only introduces a novel approach to repurpose SBC but also contributes to the development of environmentally friendly geopolymeric materials with versatile applications, underlining the significance of optimizing the geopolymerization process and enhancing the understanding of the mechanisms at play.

## CHAPTER 1

### INTRODUCTION

#### 1.1 Research Background

Geopolymer is a synthetic alkali aluminosilicate material with an amorphous to semi-crystalline polymeric structure. It can be formed through the reaction of a solid aluminosilicate with highly concentrated aqueous alkaline hydroxide, with or without the presence of a silicate solution. Geopolymer is also known as an environmentally friendly material as it utilizes waste material or industrial by-products, and its production process causes a lesser environmental impact (Majdoubi et al., 2021). Depending on the design parameters, geopolymer can be produced with various properties and applications. The geopolymer can offer many advantages, such as excellent strength and thermal resistance under an acidic environment.

Geopolymers, a term coined by Davidovits in the 1970s, is an alternative cementitious material with the properties of ceramics (Davidovits, 1993). According to Ferone et al. (2015), the "geo" part symbolizes the constitutive relationship of the binders to geological materials, such as natural stones or minerals. The "polymer" part alludes to the composition of geopolymers with a three-dimensional network of monomers (Ferone et al., 2015). Geopolymers, which can also be defined as alkaline cement or inorganic polymers, are highly polymerized materials with a three-dimensional aluminosilicate structure. They resulted from the chemical interaction between a source of aluminosilicate and a strongly alkaline solution, for example, alkali silicate or alkali hydroxide (Aguirre-Guerrero et al., 2017; Wang et al., 2020). The aluminosilicate materials must be rich in aluminium and silicon, which can be found in natural minerals such as clay and by-product minerals. At the same time, the alkaline activator solution



is usually sodium-based or potassium-based (Rangan, 2014). Theoretically, with a strongly alkali-activated solution, any aluminosilicate material can be used to produce geopolymers (Khalifa et al., 2020).

According to the Malaysian Palm Oil Board (2022), Malaysia is currently the world's second-largest oil palm producer, with 18.45 million tonnes of crude palm oil produced in 2022 (Malaysian Palm Oil Board, 2022). After extraction, the crude palm oil must undergo a refining process, which includes degumming, neutralization, bleaching, and deodorization (Abdelbasir et al., 2022). The refining process aimed to increase the stability, flavour, and aesthetic appeal of the edible oil by removing unwanted impurities and traces of metal ions from the crude palm oil (Moshi, 2017). In Malaysia, the most habitual practice to refine crude palm oil is by physical refining process using fresh bleaching clay due to its high specific surface (Su et al., 2018). This method is widely used due to its efficacy, high recovery, and less operating cost (Wafti et al., 2011). As a result, when the spent bleaching clay (SBC) is disposed of, it may still include up to 40 wt% of residual oil as well as pollutants such as coloring pigments, metallic traces, and fibers (Loh et al., 2017). The SBC might be viewed as a problematic waste with limited reusable benefits when it is not treated, which could result in critical issues for the country's finances and ecology.

Normally, 0.5 – 1.0 wt% of fresh bleaching clay was employed in the refining process of the crude palm oil to remove the impurities to produce edible oil (Wafti et al., 2011). Malaysia is one of the world's top palm oil producers with 423 palm oil mills (Othman et al., 2020). Based on the reported crude palm oil production in 2020, Malaysia alone generated an estimated 191,400 tonnes of SBC at 1% clay dosage added (Nordin et al.,

2021). The amount of SBC produced is predicted to increase annually as the demand for palm oil increases.

In this study, we aimed to investigate the feasibility of utilizing spent bleaching clay (SBC), a waste product from the oil palm refining industry rich in aluminosilicate materials, to produce an environmentally friendly geopolymer. Geopolymers are typically made by chemically reacting aluminosilicate materials with alkaline activator solution, but the potential usage of SBC in geopolymer production remains largely unexplored. This study innovatively explores the potential of repurposing SBC waste, which is traditionally discarded in landfills, for geopolymer production. This approach presents a novel and sustainable solution to waste management and contributes to the field of geopolymer technology. This study seeks to fill the knowledge gap by optimizing the incorporation of SBC into geopolymer formulations and discovering the properties of SBC-based geopolymer. Additionally, we aim to provide insights into the mechanism of these geopolymers. Ultimately, this study aims to offer a sustainable and efficient method in transforming waste materials into valuable, environmentally friendly product, which aligns with the global interest in sustainable practices and waste reduction.

## **1.2 Problem statement**

The increasing recognition of environmentally friendly geopolymers, known for their excellent mechanical properties and chemical durability, has prompted extensive research worldwide. However, despite the success in using various materials, such as fly ash and calcined kaolin clay, to produce geopolymers, the untapped potential of spent bleaching clay (SBC) remains a novel and unexplored territory. Spent bleaching

clay (SBC), a waste product from the oil palm refining industry, possesses unique properties that make it an intriguing candidate for geopolymer production. Comprised primarily of aluminosilicate material, SBC has a valuable source of aluminum and silicon, which are the essential components for geopolymerization process. Furthermore, its abundance and being a by-product of palm oil refining presented an opportunity for sustainable and cost-effective material utilization. However, while SBC exhibits potential, its utilization in geopolymer production necessitates the optimization of various parameters, including the composition of raw materials and the curing conditions. The challenge lies in revealing the desirable properties of SBC-geopolymer materials and defining the optimal parameters for the synthesis. This optimization is critical to utilize the unique properties of SBC fully. Therefore, this research aimed that through the optimization of key parameters, the SBC can be effectively utilized to produce geopolymers. Thereby contributing to the sustainable waste management practices and expanding the scope of environmentally friendly material in the industry.

### **1.3 Research objective**

The overall aim of this study is to synthesize and characterize a porous clay-based geopolymer from uncalcined spent bleaching clay (SBC). The specific objectives of this research are as follows:

1. To optimize the geopolymerization process of synthesizing SBC-based geopolymers and calcium silicate-SBC-based geopolymers.
2. To characterize the raw materials and synthesized porous geopolymers (SBC-based geopolymer and calcium silicate-SBC-based geopolymer).
3. To deduce the mechanism of the geopolymerization based on the analysis of the characterization techniques.

#### **1.4 Scope of research**

1. This research will focus on optimizing the geopolymerization process for the SBC-based geopolymers and calcium silicate-SBC based geopolymers. The scope includes varying the parameters on the amount of alkaline activator, the ratio between alkaline activator solution and SBC, and the amount of calcium silicate added. At the same time, the types of alkaline activator and curing conditions such as temperature and duration were controlled in this research.
2. This research will characterize the raw materials and geopolymers in terms of mechanical and chemical properties, including Fourier Transform Infrared spectroscopy (FTIR), Scanning Electron Microscopy coupled with Energy Dispersive X-ray (SEM/EDX), thermogravimetric analysis (TGA), N<sub>2</sub> Brunauer-Emmett-Teller analysis (BET), X-Ray Diffraction analysis (XRD), and compressive strength. The characterization aims to provide a comprehensive evaluation of the geopolymer synthesis.
3. This research will explore and elucidate the mechanism underlying the geopolymerization process, with specific focus on the utilization of the SBC. This encloses the investigation of the chemical reactions and structural transformations that contributed to the formation of geopolymer.

#### **1.5 Significance of research**

The research findings from this study will help in producing geopolymers that are more effective, long-lasting, and environmentally benign. Due to its profitable use from waste material than being generally discarded in landfills, geopolymer has attracted interest in recent years. This research assists in the synthesis of geopolymer utilizing the optimum formulation, which has the potential to affect the industries significantly.

## **1.6 Overview of thesis**

This thesis is structured along the background of this research, followed by the experimental design, and the research findings. Chapter 1 briefly describes the desired product and raw material used, which is the geopolymer and SBC from the palm oil industry, respectively. Chapter 2 comprehensively reviews the literature on aluminosilicate materials and alkaline activator solutions used for geopolymer production. The chapter is continued with the review of factors influencing the chemistry and properties of geopolymer such as the different ratios between aluminosilicate materials and alkaline activators, and the curing conditions. Lastly, several recent geopolymer applications were also addressed in this chapter.

Chapter 3 detailed the materials used to synthesize geopolymers and the characterization methods for both the raw materials and geopolymers. This chapter also covered the optimization of the formulation for geopolymer. The detailed methodology for completing the experimental part of the study is presented. Subsequently, Chapter 4 demonstrated the experimental findings from the formulation optimization to the characterization of the raw materials and geopolymers. This chapter included brief statements on the experimental results and a discussion backed by prior studies. Last but not least, Chapter 5 summarized the research findings from this study and suggested some future studies in order to bring spent bleaching clay-based geopolymer from the lab to practical use.

## CHAPTER 2

### LITERATURE REVIEW

#### 2.1 Aluminosilicate materials

The aluminosilicate material that is used in geopolymer synthesis should be rich in alumina ( $\text{Al}_2\text{O}_3$ ) and silica ( $\text{SiO}_2$ ) from the geological origin, industrial waste, or by-product, such as the metakaolin obtained by calcined kaolin clays, fly ash, slag, and rice husk ash. Generally, any material that contains alumina and silica in a reactive state that is readily dissolved in an alkaline environment can be used in the geopolymerization process. A significant amount of research has recently been done on clay-based geopolymer as clay is an abundant natural aluminosilicate source. Clay has been commonly used to produce ceramics, bricks, roof tiles, and buildings for ages ago. According to Zhang et al. (2022), clay is frequently utilized as a partial substitution in cement concrete to enhance its mechanical properties and durability. The utilization of natural clays such as kaolin, illite, and montmorillonite in geopolymer has gained the interest of many researchers due to its low cost, ample, and high availability in most countries (Farhan, et al., 2020; Neiber-Deiters et al., 2019).

##### 2.1.1 Kaolin clay

Compared to other types of clay, most studies have focused on the geopolymer synthesis from kaolinite and metakaolinite. Metakaolinite ( $\text{Al}_2\text{Si}_2\text{O}_7$ ) is a dehydroxylated form of the clay mineral kaolinite ( $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ ), which is formed by the calcination of kaolin clay. Kaolinite has a 1:1 dioctahedral layer structure where the silica sheet and the alumina sheet are bonded by oxygen atoms, while metakaolin has a more open structure compared to kaolin. Figure 2.1 demonstrates the structural

diagram of kaolinite clay mineral. Natural kaolin is rich in alumina and silica, with some presence of iron and potassium (Ayeni et al., 2021; Louati et al., 2016).

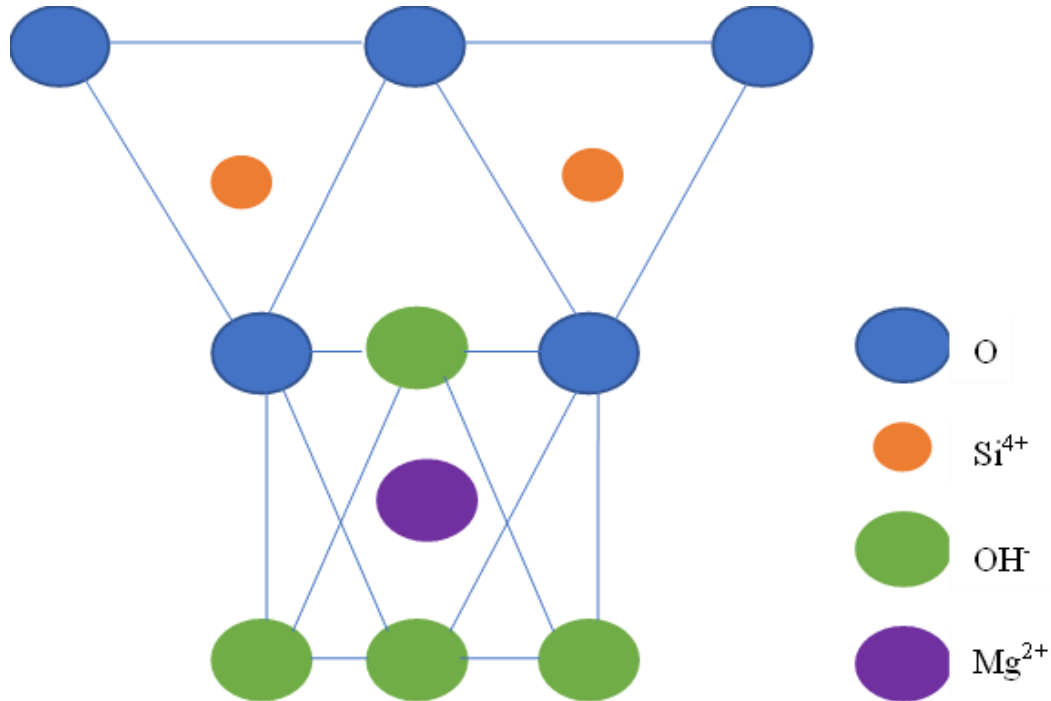


Figure 2.1 Structural diagram of kaolinite clay mineral (Adapted and modified from Dudkin et al., 2005).

### 2.1.2 Montmorillonite clay

Although most research is more focused on kaolinite, however, the smectite mineral group and the illite mineral group  $((K,H)Al_2(Si,Al)_4O_{10}(OH)_2 \cdot H_2O)$  were found to exhibit stronger characteristics (Uddin, 2018). This is due to their higher aluminosilicate ratio of 2:1 compared to kaolinite (1:1) (Heath et al., 2014; Uddin, 2018). In other words, the structural unit of smectite and illite clay is made up of two tetrahedral silica layers bonded by an octahedral alumina sheet. This finding can be further supported by research done on geopolymer synthesis using kaolinitic clay and

illite-kaolinitic clay, in which the clay is rich in illite clay mineral possesses superior mechanical characteristics to that of kaolinite-rich clay (Essaidi et al., 2014).

Montmorillonite clay mineral with the general formula of  $(Ca, Na, H)(Al, Mg, Fe, Zn)_2(Si, Al)_4O_{10}(OH)_2 \cdot H_2O$  is frequently used in the synthesis of clay-based geopolymers. Figure 2.2 demonstrates the structural diagram of the montmorillonite clay mineral group. The 2:1 dioctahedral layer structure of montmorillonite was bonded together by interlayer cations, electrostatic forces, Van der Waals forces, or hydrogen bonds (Uddin, 2018). As a result, when an alkali reactant attacked, the near-zero charges between the layers limited the replacement of other elements and hindered the exchange of ions. In other words, the chemical attack will begin at the surface and edge of the clay structure and progressively works its way within through the layers (Liew et al., 2016).

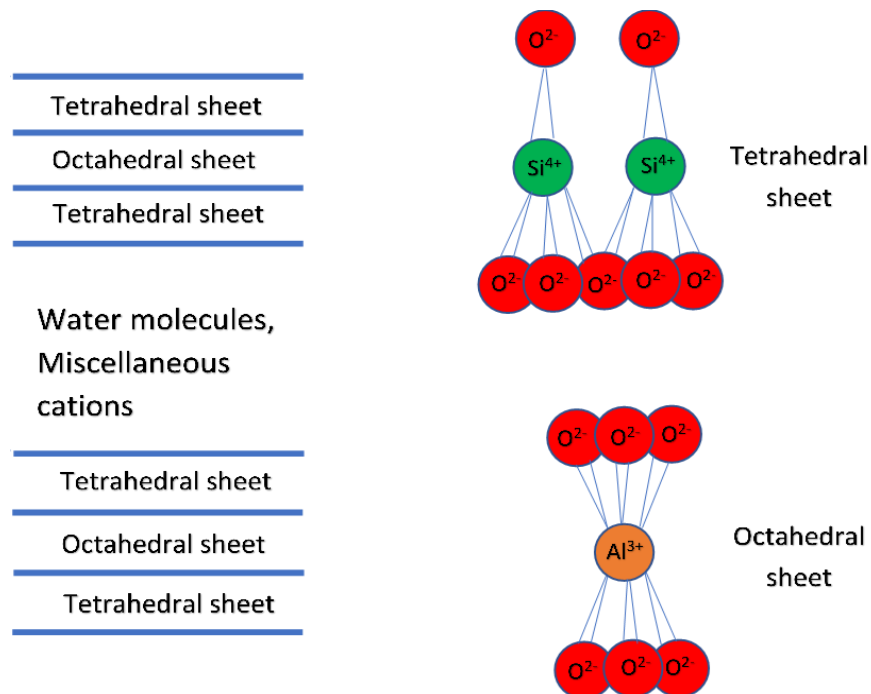


Figure 2.2 Structural diagram of montmorillonite clay mineral (Adapted and modified from Uddin, 2018).



### 2.1.3 Other types of clay

The possibility of producing clay-based geopolymer from calcined illite-smectite clay which is mainly composed of illitic layers and muscovite was shown by Hu et al. (2017). However, the raw clay consists of about 56% of silica and only 17% of alumina. Therefore, the addition of gibbsite clay mineral ( $\text{Al}(\text{OH})_3$ ) is necessary to lower the total Si/Al ratio (Hu et al., 2017). This is because when the Si/Al ratio is  $>2$ , the addition of silicate ions by the alkaline activator solution is limited (Buchwald et al., 2009). Although the meta-illite-smectite clay mineral was not completely dissolved as reported in the X-Ray Diffraction (XRD) analysis of geopolymer, the addition of 10wt% gibbsite has been proven to increase its compressive strength by about 20% (Hu et al., 2017).

Besides, laterite clay, which is a kind of iron-rich aluminosilicate material, has also attracted several researchers to synthesize geopolymers. The laterite clay is mainly composed of alumina and silica in the range of 37% to 57%, with a significant amount of iron oxide,  $\text{Fe}_2\text{O}_3$  in the range of 38% to 50%. The XRD analysis on the uncalcined laterite showed the presence of goethite ( $\alpha\text{-FeO}(\text{OH})$ ), hematite ( $\alpha\text{-Fe}_2\text{O}_3$ ), ilmenite ( $\text{FeTiO}_3$ ), quartz, and kaolinite as the main mineral phase (Kaze et al., 2018a; Kaze et al., 2021a; Kaze et al., 2021b; Sontia Metekong et al., 2021). After calcination, the kaolinite and goethite have dehydroxylated to metakaolinite and hematite, respectively (Kaze et al., 2021a; Kaze et al., 2021b; Matthew & Issac, 2020). The partial substitution of Al by Fe that causes a disorder in the clay structure makes it a potential raw material for poly(sialate-siloxo) type of geopolymer (Cong et al., 2021; Lassinantti Gualtieri et al., 2015; Kaze et al., 2021a). Kaze et al. (2018b) suggested that the highest flexural strength of 40MPa achieved by the Fe-rich lateritic clay-based geopolymer is due to the formation of ferrisilicate that enhances its mechanical properties (Kaze et al., 2018b).

Xu & van Deventer (2000) utilized various types of natural clay minerals as the precursor materials in geopolymer formation, which are almandine, andalusite, anorthite, augite, celsian, grossular, heulandite, hydroxyapophyllite, illite, kyanite, lepidolite, pumpellyite, sillimanite, sodalite, spodumene, and stilbite. Although using mainly kaolinite in the production of geopolymer resulted in a weak structure, adding kaolinite as a secondary source can improve its strength. Compared to all the minerals, stilbite showed the highest compressive strength of 18MPa (Xu & van Deventer, 2000).

## **2.2 Spent bleaching clay (SBC)**

The SBC comprises a majority of  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$ , followed by  $\text{MgO}$ ,  $\text{Fe}_2\text{O}_3$ , and  $\text{CaO}$  (Loh et al., 2017). After refining, the SBC mainly contains 20 – 40 wt.% of residual oil, metallic impurities, and other organic compounds such as carotenoids and chlorophyll (Beshara & Cheeseman, 2014; Dijkstra, 2020; Pollard et al., 1993, Sedghamiz et al., 2019). However, some processes could make the SBC contain substances besides the fresh bleaching clay and crude palm oil. In order to be employed in the synthesis of geopolymer with outstanding strength, the SBC should consist of around 45-55% of  $\text{SiO}_2$ , 20-28% of  $\text{Al}_2\text{O}_3$  and around 15-20% of  $\text{Fe}_2\text{O}_3$  and  $\text{CaO}$  (Murmu & Patel, 2020).

### **2.2.1 SBC as problematic waste**

The SBC that contains unsaturated oil and undesirable impurities makes the material unstable, which restricts the disposal options (Bachmann et al., 2020). The traditional disposal methods of SBC are through landfilling, incineration, and usage as compost without any prior processing. However, these methods usually cause secondary pollution to the environment (Zhang et al., 2022).

According to Loh et al. (2013), the most common approach in Malaysia is to haul and bury SBC at a solid waste disposal facility. First, unpleasant odours will be generated when the entrained oil rots (Abdelbasir et al., 2002). The degradation of the residual oil in it can cause fire and pollution hazards (Loh et al., 2013; Sedghamiz et al., 2019). Upon exposure to air, the entrained oil will undergo rapid oxidation and generate heat, which can lead to spontaneous combustion and an explosion. Furthermore, the entrained oil from the unprocessed SBC will leach into the soil and pollute the ground water.

As a result, the country suffers environmental and financial damages due to the disposal of SBC (Hatami et al., 2018). Hence, it can be considered a sort of waste that poses a problem in terms of handling and management, both in terms of nature and volume created, due to the lack of viable recovery options (Eliche-Quesada & Corpas-Iglesias, 2014).

### **2.2.2 Recycling and reutilizing of SBC**

With regards to issue mentioned above, it is crucial to find potential reuse of SBC to ensure the continuing feasibility of the ecosystem. SBC has been employed in a number of environmental and industrial applications due to its accessibility, affordability, and physical and chemical characteristics, including high surface area and ion exchange capacity, and chemical and mechanical stability (Phey et al., 2022). Some approaches to recycling SBC, including ultrasound-assisted in-situ transesterification of residual oil to methyl esters and the reuse of the clay material for bleaching after transesterification have been discussed (Boey et al., 2011a; Boey et al., 2011b).

To guarantee that the disposal of SBC is environmentally benign, a number of extraction methods are utilized to remove the residual oil from SBC, such as thermal treatment, solvent leaching, and supercritical carbon dioxide extraction (Su et al., 2018; Xu et al., 2019)). Mixing the extracted oil with plant waste can be used for biodiesel production and as a biofertilizer (Abdelbasir et al., 2022; Pollard et al., 1993). In addition, the SBC can be regenerated to be utilized in the adsorption field such as in the purification of heavy metals and wastewater treatment (Pollard et al., 1993; Mahmoud et al., 2012, Zhang et al., 2021).

### **2.3 Alkaline activator solution**

The alkaline activator solution plays a significant role in the synthesis of geopolymer. The reaction between an alkaline activator solution and an aluminosilicate material causes the breakdown of Si-O-Si and Al-O-Si covalent bonds, releasing the aluminate and silicate monomers (Aboulayt et al., 2017; Muraleedharan & Nadir, 2021; Petermann et al., 2010). The oligomeric aluminate and silicate species will undergo polymerization to form aluminosilicate gel, forming a geopolymer upon polycondensation (Juengsuwattananon et al., 2019; Sata & Chindaprasirt, 2020). The types of alkaline activator solutions used can affect the efficiency of the dissolution of aluminosilicate material and the polycondensation process, which further affects the microstructure and properties of the geopolymer formed (Zhao et al., 2021).

#### **2.3.1 Sodium vs Potassium**

Although most of the research targeted sodium ions, Na<sup>+</sup> and potassium ions, K<sup>+</sup> to form the alkaline activator solution, hypothetically, all types of alkali metal from Group 1 can be used to synthesis clay-based geopolymer (Hounsi et al., 2014). Other research

on geopolymers has also been done using different alkaline activators such as lithium and calcium (Askarian et al., 2019; Cong & Cheng, 2021). Different types of alkali metals of different sizes and chemical properties can affect the rate of formation and the microstructure of the geopolymer formed.  $\text{Na}^+$  ions that are relatively smaller show stronger pair formation with smaller silicate oligomers, while the larger  $\text{K}^+$  ions tend to form larger silicate oligomers (Gomez-Casero et al., 2021; Hendricks et al., 1991; Hounsi et al., 2014; Leong et al., 2016). In other words, the use of NaOH solution increases the dissolution of aluminosilicate material. This increases the number of monomeric silicate oligomers while the use of KOH solution increases the rate of polycondensation.

A comparative study on the physical and mechanical properties of geopolymers has been done by Gomez-Casero et al. (2021). The authors compared the properties of geopolymers that use different kinds of alkaline activating solutions, such as sodium hydroxide solution, potassium hydroxide solution, and the mixture of alkaline hydroxide with alkaline silicate solution. At the same concentration of alkaline activator solution, the geopolymers that are activated by KOH presented lower thermal conductivity and had a higher bulk density compared to geopolymers activated by NaOH. This means that the geopolymers produced have a higher insulating capacity and greater compressive strength. The finding was further supported by the FTIR spectra of geopolymers using KOH solution with a higher wavenumber of Si-O and Al-O bond stretching that may indicate a higher degree of polycondensation (Gomez-Casero et al., 2021). A similar study also reported a higher compressive strength in KOH-based geopolymers at low silicate content (Leong et al., 2016).

However, Leong et al. (2016) also showed that when the silicate content increase at which the ratio of sodium silicate ( $\text{Na}_2\text{SiO}_3$ ) to KOH or NaOH exceeds 1.5, the dissolution of NaOH-based geopolymer is higher and geopolymer produced have higher compressive strength (Leong et al., 2016). A similar study reported a higher compressive strength in geopolymer activated by NaOH solution at a silicate to sodium or potassium ratio of 1.9 (Zhang et al., 2017).

The smaller size  $\text{Na}^+$  ion is relatively more reactive than the  $\text{K}^+$  ion, the amount of energy required to form oligomers decreases, and the densification degree increases. This caused the smaller pores to agglomerate and resulted in larger pore sizes. A metakaolin-based geopolymer produced using NaOH alkaline activator exhibits a larger pore size but a lower porosity compared to the geopolymer that used KOH (Gharzouni et al., 2016). Another study by Hu et al. (2017) showed that under the same curing temperature of  $75^\circ\text{C}$ , the NaOH activator is more effective at dissolving silicate and aluminate than the KOH activator, proven by the higher Si and Al concentration in the solution (Hu et al., 2017). A similar statement on the effectiveness of sodium in the dissolution of aluminosilicate material was made by Phair & van Deventer (2002).

### **2.3.2 Combination of alkaline activator solution**

To synthesis a clay-based geopolymer, most of the research uses the combination of NaOH and  $\text{Na}_2\text{SiO}_3$  as the alkaline activator solution (Hodhod et al., 2020; Ogundiran & Kumar, 2015; Palacios et al., 2021; Salimi & Ghorbani, 2020; Singh, 2021; Tahmasebi Yamchelou et al., 2021; Zidi et al., 2021). However, there are also numerous studies found to use the combination of potassium hydroxide (KOH) and potassium silicate ( $\text{K}_2\text{SiO}_3$ ) as an activator solution to synthesis geopolymer (Dupuy et al., 2019;

Gharzouni et al., 2016; Selmani et al., 2017; Tippayasam et al., 2016). As a comparison, most studies practice the combined use of NaOH and Na<sub>2</sub>SiO<sub>3</sub> rather than a potassium-based alkaline activator solution because of its cost-effectiveness and availability (Heath et al., 2014; Muraleedharan & Nadir, 2021). The alkaline activator solution does not limit to the combination of sodium or potassium hydroxide and its silicates but also carbonates and different kinds of silicates (Autef et al., 2012; Pourabbas Bilondi et al., 2018; Helmy, 2016; Hendricks et al., 1991; Zhao et al., 2021).

The sole usage of alkaline silicates as an activator is not favorable. Research on the influence of different types of alkaline activator on the strength performance concluded that the clay geopolymer activated by only NaOH generally have higher compressive strength than Na<sub>2</sub>SiO<sub>3</sub>. At a mass ratio of alkaline activator to calcined clay of 0.7, clay geopolymer activated by NaOH achieved higher compressive strength at both 7 and 28 days compared to only the Na<sub>2</sub>SiO<sub>3</sub> activator. Besides that, the NaOH activator also showed a higher compressive strength from the sodium silicate pentahydrate activator with an alkaline solution to calcined clay ratio in the range of 0.8-1 (Bature et al., 2021).

Although the usage of silicates solely is not recommended due to their low stability and their production that involves high temperatures which is not environmentally friendly (Autef et al., 2012; Petermann et al., 2010; Slaty et al., 2013), yet some of the research that included silicates in the alkaline activator solution reported better structure and strength (Singh et al., 2005). According to Bature et al. (2021), geopolymer can be synthesized by using an activator solution with the addition of silicates at a correct formulation, while using NaOH solely favors the production of alkali-activated material (Bature et al., 2021).

Geopolymers that are activated by only NaOH solution detected crystalline sodalite and type-A zeolite as the majority instead of amorphous geopolymeric gel. However, geopolymers with the addition of Na<sub>2</sub>SiO<sub>3</sub> in the activator showed a less ordered structure without detectable zeolitic formation (Juengsuwattananon et al., 2019; Zhang et al., 2013). In other words, crystallization occurs instead of geopolymerization without the addition of Na<sub>2</sub>SiO<sub>3</sub> solution because the silicates can obstruct the formation of crystalline zeolite. Geopolymers activated by only NaOH solution also report lower compressive strength, higher thermal conductivity, and longer setting times than the geopolymers activated with the addition of Na<sub>2</sub>SiO<sub>3</sub> (Gomez-Casero et al., 2021; Jun et al., 2021; Laskar & Talukdar, 2017; Palacios et al., 2021).

### **2.3.3 Solid vs Liquid alkaline activator**

Comprehensively, a geopolymer is formed when an aluminosilicate material is activated by an alkaline activator in a highly alkaline environment. Nevertheless, the activator is not necessarily in liquid form but can also exist in powdered form. The method used to produce geopolymer using an alkaline activator solution can be known as the wet mixing method. This method of preparing geopolymer paste is widely used in most of the research in synthesizing geopolymer (Eliche-Quesada et al., 2021; Ghani et al., 2021; Piriyaikul & Iamchaturapatr, 2018; Qin et al., 2021; Yurt, 2020). However, the usage of an alkaline activator in liquid form has some disadvantages such as being time-consuming because the alkaline activator needs to cool down to room temperature before mixing with the aluminosilicate source (Zhao et al., 2021).

On the other hand, the powder-activated geopolymer, also known as a one-part geopolymer, is produced by dry mixing the alkaline activator with the aluminosilicate



source. In this dry mixing method, the alkali activator and the aluminosilicate source must be mixed and milled into a fine powder before being used. They are known as one-part geopolymers because they can be formed by adding only water directly to the powdered geopolymer precursor (Bayuaji et al., 2017; Luukkonen et al., 2018; Muraleedharan & Nadir, 2021). Hence, this method can reduce the risk of corrosion caused by direct contact with the alkaline activator solution (Assi et al., 2020; Wu et al., 2021). Moreover, the one-part geopolymer is also more convenient as in-situ applications can be achieved, unlike the conventional two-part geopolymers that are mostly used in the precast industry due to the availability of controlled conditions (Alrefaei et al., 2019).

However, one of the drawbacks of this method is that the dissolution of the aluminosilicate source requires a highly alkaline environment but adding water to the system causes the alkalinity to increase more slowly. Therefore, the alkali activator with the appropriate dissolution kinetic is significant in the formation of geopolymer gel (Askarian et al., 2019; Dong et al., 2020). Ke et al. (2014) produced a one-part geopolymer using the powder blends of dried red mud and NaOH pellets, and it reported a compressive strength of 9.8MPa after 28 days. However, the publication also reported that the geopolymer produced has a limited strength development due to the excess alkalis in the system.

## **2.4 Geopolymer**

In recent years, geopolymer technology has attracted much interest among researchers. Geopolymers are a new environmentally friendly material and a new element of sustainable architecture because of their minimal carbon footprint in the atmosphere

during manufacturing, ambient temperature production, and high compressive and flexural strengths (Janosevic et al., 2018). Geopolymers are also an option for concrete binders due to their exceptional physical, chemical, and mechanical properties. They can also be used in various applications due to their outstanding physical properties and depending on the composition of the raw materials used.

#### **2.4.1 Ordinary Portland Cement (OPC)**

Concrete is one of the most widely utilized materials in the building and construction sector due to its great durability (Almutairi et al., 2021). For the time being, the construction industry plays a significant role in all economic sectors. The high daily demand for concrete and the negative environmental impact caused by its production has made it crucial to develop a sustainable alternative.

Ordinary Portland Cement (OPC), which is used to make concrete by mixing it with water and aggregate, is one of the most energy-intensive industries and is responsible for some of the most contentious global concerns (Gayathri et al., 2016; Madloul et al., 2011). The production of OPC requires the combustion of a large amount of fossil fuels, emitting tonnes of greenhouse gases into the environment and contributing to global warming (Kong & Sanjayan, 2008). According to McCaffrey (2002), the production of 1 tonne of OPC resulted in the release of 1 tonne of CO<sub>2</sub>. Additionally, 1.5 tonnes of limestone are needed for production, which could result in a shortage of natural resources.

Therefore, a suitable alternative has to be implemented in the construction industry to reduce the emission of greenhouse gases and to preserve our natural resources.

Geopolymer technology might be viewed as one of the potential replacements for producing OPC due to its equivalent cementitious properties and lower atmospheric CO<sub>2</sub> emissions of up to 80% (Cong & Cheng, 2021; Rao & Liu, 2015). Depending on the raw materials used, clay-based geopolymers may be a replacement for OPC, where clay has greater thermal stability and shrinkage properties.

#### **2.4.2 Advantages of geopolymer**

Numerous research has demonstrated that geopolymer outperformed OPC in terms of mechanical strength and durability properties. Over the years, from their application as alternative building materials, geopolymers have evolved to incorporate their usage as fireproof materials. The geopolymers' ceramic-like properties and the zeolite material make-up, which is amorphous to semi-crystalline, make it highly resistant to fire and corrosion and can immobilize toxic and radioactive material (Bakri et al., 2011; Salwa et al., 2013).

These outstanding results enable a wide range of applications for geopolymer, including environmental protection and building materials processing, which are primarily of interest to civil engineering communities, and even as a nuclear waste encapsulation medium (Davidovits, 1994a; Guo et al., 2017; Yakubu et al., 2017; Ji and Pei, 2019). Davidovits (1994b) suggested that geopolymers with the mentioned properties can be designed for toxic waste containment and acid-resistant geological barriers (Davidovits, 1994b). Carabba et al. (2017) demonstrated that geopolymers with strong chlorine resistance could be utilized in the marine environment, such as undersea supports and coastal bridges. Apart from the excellent chemical durability and mechanical properties

of geopolymers, the ease of production from industrial wastes or substandard materials gives added value to the geopolymers (Davidovits, 1991; Davidovits, 1993).

### **2.4.3 Clay-based geopolymer using different types of clay**

Hamdi et al. (2019) investigated the feasibility of producing geopolymer from a mixture of Tunisian illito-kaolinitic clay with different ratios of phosphogypsum and waste bricks. The phosphogypsum mainly contains  $\text{SO}_3$  and  $\text{CaO}$ , while waste bricks from the brick industry contain about 75% of aluminosilicate. From this study, the utilization of 8% phosphogypsum with 14M sodium hydroxide (NaOH) activator solution shows the highest compressive strength of 36MPa and the highest density of  $2296\text{kg/m}^3$  after 28 days. Under the same condition, when the calcined clay is replaced by waste bricks, the result shows the highest compressive strength of 32MPa with nearly the same mechanical property (Hamdi et al., 2019).

Aziz et al. (2021) studied the potential of using calcined kaolin clay and natural perlite, which is a kind of silica-rich hydrated volcanic glass, to produce geopolymers with different properties and applications. The perlite consists of a high molar ratio of Si/Al of 9.48 with 73% of silica content. By substituting kaolin clay with perlite at different ratios, the synthesized geopolymers demonstrated different microstructure, mineralogical composition, and mechanical properties. The substitution of 10% kaolin clay showed a compact geopolymer with no cracks on the surface and the least porosity while the substitution of 50% produced porous light-weighted geopolymers that are suitable as an insulating material (Aziz et al., 2021).

Yang et al. (2020) reviewed the addition of bentonite clay, consisting mainly of montmorillonite clay mineral with approximately 84% aluminosilicate materials to produce porous geopolymer. The pore size and pore distribution increase as the amount of bentonite increases up to 60%. Although the increasing bentonite amount decreased the density of the geopolymer, the compressive strength increased and reached a maximum of 59.6MPa, making it a potential thermal insulation material compared to other porous geopolymers (Yang et al., 2017).

The research was done by Keppert et al. (2018) on the possibility of producing geopolymer using two different red-clay-based ceramic powders, which is a kind of by-product from brick factories. The ceramic wastes, namely type H and type L, are mainly composed of quartz, microcline, muscovite, and albite clay minerals, with a higher amorphous aluminosilicate and CaO content in type L. As a result, the geopolymer formed using type L waste contains more amorphous matter and the higher content of CaO causes a higher porosity in the geopolymer. This research also proved that clay-based geopolymers could be formed, although the amorphous aluminosilicate content of the raw material is lower than 30% (Keppert et al., 2018).

Sarkar and Dana (2021) studied the partial replacement of metakaolin with red ceramic waste in geopolymer. The ceramic waste produced by crushing the fired red clay articles contains about 83% aluminosilicate with a significant presence of illite and muscovite. The replacement of 33% of ceramic waste reported almost the same compressive strength and fine-grained texture observed from the FESEM micrographs compared to the purely metakaolin-based geopolymer. When the amount of ceramic

waste increases to 50%, the compressive strength shows a sharp decline, and the unreacted large grains are observed (Sarkar and Dana, 2021).

A similar study was done by Rovnanik et al. (2018) using waste red brick powder to replace metakaolin. The waste material was obtained by crushing the red clay bricks from the industry, containing about 67% of aluminosilicate materials. Although the substitution of 25% of the waste showed insignificant changes in the rheological, the geopolymer reported the highest compressive and flexural strength (Rovnanik et al., 2018). Furthermore, partial replacement of metakaolin was done using four different types of calcined Spanish clays and biomass bottom ash. Generally, the clays consist of quartz and illite as major components while hematite and montmorillonite as minor. Despite the types of clays, the substitution of 33% clay and 33% biomass bottom ash produced geopolymers with better mechanical strength, higher bulk density, and slightly higher thermal conductivity (Eliche-Quesada et al., 2021).

Clay-based geopolymer that is produced by only laterite clay usually results in weak mechanical properties, but this can be improved by the addition of other natural materials or industrial by-products. As an example, the addition of halloysite clay. Halloysite is a 1:1 clay mineral type similar to kaolinite, and the calcined clay mainly consists of meta-halloysite, ilmenite, and anatase. When the meta-halloysite substituted up to 50% of the clay, the increased amount of amorphous aluminosilicate content and its tubular structure can promote the dissolution reaction (Khale and Chaudhary, 2007). As a result, the homogeneity of geopolymer becomes higher, forming a denser and more compact structure reported with the highest compressive strength of 45MPa (Kaze et al., 2020). Lemougna et al. (2017) reported the geopolymer with a denser

microstructure and the highest compressive strength of 65MPa by substituting 50% ground granulated blast-furnace slag for laterite clay.

A summary of clay-based geopolymers produced using different types of clay and waste materials is shown in Table 2.1, with NA representing no data reported in respective publications.