UNIVERSITI PUTRA MALAYSIA

EFFECT OF WATER RATIO AND AGING TIME ON GLASS IONOMER CEMENT DERIVED FROM CALCIUM FLUOROALUMINOSILICATE-BASED GLASS

MOHAMMAD ZULHASIF AHMAD KHIRI

ITMA 2022 7

EFFECT OF WATER RATIO AND AGING TIME ON GLASS IONOMER CEMENT DERIVED FROM CALCIUM FLUOROALUMINOSILICATE-BASED GLASS

By

MOHAMMAD ZULHASIF AHMAD KHIRI

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Doctor of Philosophy

October 2021

DEDICATIONS

To my beloved parents,
My inspiration father, Ahmad Khiri Takim
My lovely mother, Zainun Musa
Thank for your unlimited support and encouragement

To my wife, Fathiah Syazana and her family,
For make my spiriting up throughout my studies journey until the end
Thank for your thousand love and care

To my sibling,
For makes my life happiness
Thank for your lovely care

To all my wonderful and kind friends,
For makes my life always in a colorful dream
Thank for your infinite help

To all my lecturers,
For their infinite teach and advice throughout my studies journey
Thank for your thousand supports
To me for the last,
May Allah bless me where ever I am
And
May I have success in this world and the hereafter
Amin

Without all of them, this great success would not be possible
Thank you everyone

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the Degree of Doctor of Philosophy

EFFECT OF WATER RATIO AND AGING TIME ON GLASS IONOMER CEMENT DERIVED FROM CALCIUM FLUOROALUMINOSILICATE-BASED GLASS

By

MOHAMMAD ZULHASIF AHMAD KHIRI

October 2021

Chair : Khamirul Amin Matori, PhD
Institute : Nanoscience and Nanotechnology

Glass ionomer cement (GIC) is produced from a reaction between calcium fluoroaluminosilicate (CFAS) glass powder and polyacrylic acid (PAA). These kinds of cement are widely used primarily in dentistry for a long time ago for various applications such as adhesive and tooth restorative. In this research, the GIC is designed to perform good in mechanical and antibacterial properties. CFAS glass system is seen as a suitable combination due to its structure similar with natural tooth and ability to release fluoride ions for inhibiting the bacterial growth. The compound of SiO_2 and CaO in the glass system have been replaced by waste materials to achieve the usability of waste materials in this research. Soda–lime–silica (SLS) glass and clam shell (CS) are used to act as SiO_2 and CaO sources. The SLS–CS–CaF_2–Al_2O_3–P_2O_5 glass system was used to synthesized CFAS glass by the conventional melt-quench technique and act as a based-glass in the fabricating GIC. In this study, GIC B5 [25SLS–15CS–20CaF_2–20Al_2O_3–20P_2O_5] with initial ratio of water at 1.2 revealed as the optimum sample based on the physical, structural, mechanical and antibacterial properties. From the mechanical results, the highest Vickers microhardness of GIC B5 is 191.33 HV while the highest compressive strength is 119.14 MPa at 28 days of aging time. Besides that, fluoride release by the GIC B5 was found directly proportional to the aging time between 1 to 28 days and recorded range from 74.62-194.82 ppm. The GIC also shows the antibacterial activity recorded the average diameter of the inhibition zone range between 19.33 and 28.00 mm. The antibacterial activity of GIC shows it is related to the fluoride ion release. It can be concluded that the fluoride release by GIC to the agar medium causes the inhibition of bacteria (S. mutans). Therefore, the GIC derived from CFAS glass in this study has high potential in dental material applications especially GIC B5 sample due to its good physical, structural and mechanical properties as well as its antibacterial activity.

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

KESAN NISBAH AIR DAN MASA PENUAAN SIMEN IONOMER KACA YANG BERASASKAN DARI KACA KALSIUMFLUROALUMINOSILIKAT

Oleh

MOHAMMAD ZULHASIF AHMAD KHIRI

Oktober 2021

Pengerusi : Khamirul Amin Matori, PhD
Institut : Nanosains dan Nanoteknologi

Simen ionomer kaca (GIC) atau dikenali sebagai simen polialkenoat kaca dihasilkan daripada tindak balas antara serbuk kaca kalsium fluoroaluminosilikat (CFAS) dan asid poliakrilik (PAA). Simen jenis ini banyak digunakan terutamanya dalam bidang pergigian sejak dulu untuk pelbagai aplikasi seperti pelekat dan pemulihan gigi. Dalam penyelidikan ini, GIC dirancang untuk memberikan sifat mekanikal dan sifat antibakteria yang baik. Sistem kaca CFAS dilihat sebagai kombinasi yang sesuai kerana strukturnya yang serupa dengan gigi semula jadi dan mempunyai keupayaan untuk melepaskan ion fluorida bagi menghalang pertumbuhan bakteria. SiO_2 dan CaO dalam sistem kaca telah digantikan dengan bahan buangan untuk mencapai kebolehgunaan bahan buangan dalam penyelidikan ini. Kaca soda–kapur–silika (SLS) dan shell clam (CS) masing-masing digunakan untuk bertindak sebagai sumber SiO_2 dan CaO. Sistem kaca SLS–CS–CaF_2–Al_2O_3–P_2O_5 digunakan untuk mensintesiskan kaca CFAS dengan teknik peleburan peleburan konvensional dan bertindak sebagai gelas berdasarkan pembuatan GIC. Dalam kajian ini, GIC B5 [25SLS–15CS–20CaF_2–20Al_2O_3–20P_2O_5] dengan nisbah awal air pada tahap 1.2 dinyatakan sebagai sampel optimum berdasarkan sifat fizikal, struktur, mekanikal dan antibakteria. Dari hasil mekanikal, kekerapan mikro Vickers tertinggi GIC B5 ialah 191.33 HV sementara kekuatan mampatan tertinggi ialah 119.14 MPa pada 28 hari masa penuaan. Selain itu, pelepasan fluorida oleh GIC B5 didapati berkadar terus dengan masa penuaan antara 1 hingga 28 hari dan direkodkan dari julat 74.62-194.82 ppm. GIC juga menunjukkan aktiviti antibakteria mencatatkan purata diameter julat zon perencatan antara 19.33 dan 28.00 mm. Aktiviti antibakteria GIC menunjukkan ia berkaitan dengan pembebasan ion fluorida. Dapat disimpulkan bahawa pembebasan fluorida oleh GIC ke medium agar menyebabkan perencatan bakteria (S. mutans). Oleh itu, GIC dari kaca CFAS dalam kajian ini mempunyai potensi yang tinggi dalam aplikasi bahan pergigian terutama sampel GIC B5 kerana sifat fizikal, struktur dan mekanikalnya yang baik berserta juga dengan ciri aktiviti antibakterianya.

ACKNOWLEDGEMENTS

In the name of Allah S.W.T., the Most Gracious and Most Merciful

First and foremost, I would like to speaks Alhamdullilah for all work I have done to complete my thesis and grateful to Allah for the blessing in my life. First of all, I would like to thank to Allah for giving me the functional body and good thinking to learn the huge knowledge until completing my doctoral studies.

I would like to appreciate my supervisor, Assoc. Prof Dr. Khamirul Amin Matori for his supervision and unlimited support from the beginning until the end of my doctoral studies. His comment and suggestion in this research are very helpful to me since I was new in this field of study. With his guide, I have overcome various problems well and complete my research perfectly.

Besides that, I would like to appreciate and acknowledge support from members of the supervisory committee, Dr. Che Azurahanim Che Abdullah, Dr. Mohd Hafiz Mohd Zaid and Dr. Norhazlin Zainuddin for their endless help, encouragement, and support. Thanks for the inspiration and motivation being given to me.

I would like to extend my thanks and appreciation to all lecturer in Department of Physics and all members of Ceramic Ultrasonic Laboratory (CURL) laboratory for their help and moral support during complete my research. Special thanks to my senior and junior, for their kindness and word of motivation when preparing my journal publication.

Sincere thank goes to staff members of Department of Material Synthesis and Characterization Laboratory (MSCL), Institute of Advanced Technology and Department of Physics, Faculty of Sciences for their kindness including cooperation and technical support during my research. Besides that, I also acknowledge the financial support for my study from Universiti Putra Malaysia through the Graduate Research Fellowship (GRF) through my studies.

Finally, I would like to thank my parent, Ahmad Khiri Takim and Zainun Musa for their moral support and endless prays. Not forget to siblings that always give me advice on the true path to success in study and life. The endless support of my lovely wife and her family.

I hope the blessing goes to the entire person who is direct or indirect help to complete my thesis and may Allah repay their kindness in the world and hereafter.

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

Khamirul Amin bin Matori, PhD
Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Chairman)

Che Azurahanim binti Che Abdullah, PhD
Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Member)

Mohd Hafiz bin Mohd Zaid, PhD
Senior Lecturer
Faculty of Science
Universiti Putra Malaysia
(Member)

Norhazlin binti Zainuddin, PhD
Senior Lecturer
Faculty of Science
Universiti Putra Malaysia
(Member)

____
ZALILAH MOHD SHARIFF, PhD
Professor and Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 9 March 2022

Declaration by graduate student

I hereby confirm that:

- this thesis is my original work;
- quotations, illustrations and citations have been duly referenced;
- this thesis has not been submitted previously or concurrently for any other degree at any other institutions;
- intellectual property from the thesis and copyright of thesis are fully-owned by Universiti Putra Malaysia, as according to the Universiti Putra Malaysia (Research) Rules 2012;
- written permission must be obtained from supervisor and the office of Deputy Vice-Chancellor (Research and Innovation) before thesis is published (in the form of written, printed or in electronic form) including books, journals, modules, proceedings, popular writings, seminar papers, manuscripts, posters, reports, lecture notes, learning modules or any other materials as stated in the Universiti Putra Malaysia (Research) Rules 2012;
- there is no plagiarism or data falsification/fabrication in the thesis, and scholarly integrity is upheld as according to the Universiti Putra Malaysia (Graduate Studies) Rules 2003 (Revision 2012-2013) and the Universiti Putra Malaysia (Research) Rules 2012. The thesis has undergone plagiarism detection software.

Signature: ________________________ Date: __________________

Name and Matric No.: Mohammad Zulhasif Ahmad Khiri, GS50490

Declaration by Members of Supervisory Committee

This is to confirm that:

- the research conducted and the writing of this thesis was under our supervision;
- supervision responsibilities as stated in the Universiti Putra Malaysia (Graduate Studies) Rules 2003 (Revision 2012-2013) are adhered to.

Signature: ______________________
Name of Chairman of
Supervisory Committee: Khamirul Amin Matori

Signature: ______________________
Name of Member of
Supervisory Committee: Che Azurahanim Che Abdullah

Signature: ______________________
Name of Member of
Supervisory Committee: Mohd Hafiz Mohd Zaid

Signature: ______________________
Name of Member of
Supervisory Committee: Norhazlin Zainuddin

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS AND SYMBOLS

CS	Clam shell
SLS	Soda–lime–silica
HA	Hydroxyapatite
FA	Fluorapatite
$CaCO_3$	Calcium carbonate
CaO	Calcium oxide
$Ca(OH)_2$	Calcium hydroxide
SiO_2	Silica
CaF_2	Calcium fluoride
Al_2O_3	Aluminium oxide
P_2O_5	Phosphorus pentoxide
Na_2O	Sodium oxide
$CaAl_2Si_2O_8$	Anorthite
$3Al_2O_3 \cdot 2SiO_2$	Mullite
$Ca_5(PO_4)_3(OH)$	Hydroxyapatite
$Ca_5(PO_4)_3F$	Fluorapatite
CFAS	Calcium fluoroaluminosilicate
PAA	Polyacrylic acid
GIC	Glass ionomer cement
CaP	Calcium phosphate
wt.	Weight
wt.%	Weight percentage
a.u.	Arbitrary unit
Ppm	Parts per million

rpm	Revolutions per minute
Mm	Micrometer
T_g	Glass transition temperature
T_c	Glass crystallization temperature
T_m	Glass melting temperature
V_m	Molar volume
MPa	Megapascal
Kgf	Kilogram force
F	Force
ρ	Density
H_v	Vickers hardness
XRF	X-ray fluorescence
XRD	X-ray diffraction
FTIR	Fourier transforms infrared
TGA	Thermal gravimetric analysis
DSC	Differential scanning calorimetry
FESEM	Field emission scanning electron microscopy
EDX	Energy dispersive X-ray
ISO	International Organization for Standardization
S. mutans	*Streptococcus Mutans*

CHAPTER 1

INTRODUCTION

1.1 General Introduction

This chapter focus on the introduction of the research background of the biomaterial, Calcium Fluroaluminosilicate (CFAS) glass derived from vitreous waste, Fluorapatite (FA) and Glass Ionomer Cement (GIC). The vitreous wastes such as Soda−Lime−Silica (SLS) glass and Clam Shell (CS) are used in these studies to synthesized CFAS glass and then fabricated the GIC for a potential material used in dental material application. The specific characteristic of GIC is needed to ensure the materials can function in excellent condition for dentistry. Thus, detailed information about the fabrication and uses of the GIC is explained to ensure the objectives of the studies are successful at the end of the research.

1.2 Research Background

Biomaterials are referred to as biological or synthetic materials that have replaced any defective part of the human body, such as cell tissue, organs and body function, by means of an implantation method. Many researchers have defined biomaterials in specific ways in terms of biological content. According to the American National Institute of Health, the term biomaterials is defined as any substance or combination of substances, other than drugs, of synthetic or natural origin, which may be used for any period of time which increases or replaces, in part or in whole, any tissue, organ or function of the body in order to maintain or improve the quality of life of the individual (Bergman and Stumpf, 2013). Biomaterials must be biofunctional and biocompatible in order to perform the specific function of the host, either as implants or devices (Patel and Gohil, 2012). Biofunctional term is defined as the compensation for loss of function in diseased or damaged tissues, the complete replacement of the function of such tissues or the use for diagnostic purposes (Hudecki et al., 2019). According to Williams (1987), biocompatibility in terms of implantable means any material that has the ability to respond appropriately to the host in a specific application without harm occurring in the surrounding area. The use of biomaterials is currently widely used in various applications such as tissue, orthopedic and dental engineering applications. Biomaterials are usually produced in different types of materials, such as natural, glass, ceramic, metallic and polymer, depending on their specific function in medical applications (Shackelford, 1996).

Biomaterials can be categorized into three main functions, namely inert, reactive and resorbable (Hulbert et al., 1982). Bioinert materials have high chemical stability that is not triggered by a reaction or interacts with a biological reaction. Bioinert materials have high chemical stability that does not initiate a response or interact when introduced into a biological system (Hulbert et al., 1982; Yamamuro, 2004). Simply put, the introduction of bioinert material into the biological system, such as the body, will not cause the host to react. On the other hand, bioactive materials can be defined as a material that shows reactivity to the biological effect, including interfacial bonding, and allows the tissue to ingest into the surrounding cells. While bioresorbable materials can be explained as a chemical reaction by dissolving materials to the body's host, and then absorbed by the body (Hulbert et al., 1982).

Bioglass is a type of biomaterials that can apply in the biosystem of living things such as in the human body nor animal. It also acts as a specific biological response at an interface of a material that results in the formation of a bond between the tissues and materials (Hench 1993; Hench, 1998). Bioglass 45S5 was introduced by Hench in the late 1960's at University of Florida, United State. Bioglass 45S5 has been in clinical use since 1985 (Hench, 2006). This bioglass consist of 45% SiO_2, 24.5% CaO, 24.5% Na_2O and 6% P_2O_5 (Hench and Paschall, 1974; Hench, 2006). The formulation of Bioglass 45S5 was successful used as an artificial material in the human body due to its excellent biocompatibility and bioactivity (Li et al., 2017). Until now, various type of bioglass was created with different formula composition and method according to their role in biological function. It is seen as one of the optimal compositions in biomedical applications especially in orthopedic. Bioglass 45S5 is commonly used as a reference to other bioglass due to its ability to integrate with the biological host in suitable conditions (Hench, 2013; Fiume et al., 2018). However, Bioglass 45S5 has low in mechanical applications due to its intrinsic brittleness characteristic and poor load-bearing application (Baino et al., 2019**)**. Thus, it usually will be reinforced with other materials such as metal to improve the mechanical properties without affecting the bioglass properties itself. The CFAS glass is seen as a suitable glass system that can be featured in the biological application due to its improvised mechanical properties by elemental addition such as fluorine and aluminum.

The CFAS glass system is first introduced by Hill in 1991 during studies on ionomer dental glass as a potential bioactive glass in the biomaterial application (Hill et al., 1991). The CFAS glass consists of five core chemical compounds which are SiO_2, CaO, CaF_2, Al_2O_3 and P_2O_5. The SiO_2 and CaO are two main chemical composition that can be replaced by waste materials in this research. Both SiO_2 and CaO are easily available everywhere due to their abundance dump and less used. Therefore, the idea to replace the SiO_2 and CaO with waste materials such as SLS glass and CS in the synthesis of the CFAS glass is one of the novelties in this research.

According to Sinton and Course (2001), SLS glass consists of more than 66 mol.% of silicon dioxide, SiO_2 which is ideal for the extraction element of silicon, Si from the SLS glass for a new production of glassware. The development of the glass industry in Malaysia seen has positive economic growth for the past 5 years (Sunder, 2019). Recently, two largest glass producer companies from Japan and China were invested heavily in the Malaysian market economy for glass processing activities. Malaysia was focused on the factory industry to increase the financial income by participating in manufacturing glass production and leading to the consumption of glass containers for packaged food and beverages. However, the dump of glassware is also increased due to unwanted glass containers especially from SLS glass which is commonly used for glass sheet, window glass panes and glass containers (Sehgal and Ito, 1998; Sheng et al., 2002). Therefore, the use of SLS glass in synthesizing CFAS glass directly can overcome the dumping problem of glass every year.

The shell marine species consist of rich calcium sources in form of calcium carbonate ($CaCO_3$) which is composed of more than 98 wt.% of calcium (Buasri et al., 2013). Thus, shell marine species are seen as an alternative source of high calcium content from waste materials. In Malaysia, the fisheries industry has been played an important role as a major supplier of animal protein sources to the Malaysian population due to the wide surface area of Malaysia's sea (Oon, 1986; Mohamed et al., 2012; Hussin, 2016). The Malaysia's fisheries industry has dynamic and competitive developments, and even has the potential to be developed, especially in contributing to the country's export earnings. High demand on seafood sources causes the fisheries activities in Malaysia become one of the important sectors (FAO, 2006). As a result, the dumping of shells from shell marine species such as clam shell, crab and horseshoe crab increase by the years (Chee et al., 2011). Therefore, the usability of the CS is provenly high in calcium sources and a suitable candidate for the replacement of commercial calcium in synthesizing CFAS glass for reducing the dumping problem of the shells. Moreover, the cost of CS is not only cheap but also easy to get in Malaysia.

The CFAS glass system can be crystallized to form a glass-ceramic and known as FA. The FA crystal is slightly similar to the chemical composition of hydroxyapatite, HA present in enamel composition (Wei et al., 2003; Jantová et al., 2008). The FA crystal with the chemical formula of ($Ca_{10}(PO_4)_6F$) can be substituted the fluoride ion, F^- with hydroxide ion, OH^- to form HA crystal ($Ca_{10}(PO_4)_6(OH)$) (Wei et al., 2003; Jantová et al., 2008; Rahman et al., 2019). Both FA and HA are categorized in calcium phosphate salt group which is widely used in biomedical applications due to excellence in biocompatibility properties (Tung, 1998).

This $SiO_2–CaO–CaF_2–Al_2O_3–P_2O_5$ glass system has shown a positive development in dentistry application, especially for dental cement purposes (Hill et al., 1991). The fabrication of glass ionomer cement, GIC derived from $SiO_2–CaO–CaF_2–Al_2O_3–P_2O_5$ glass system is seen as an acceptable effort for considering the GIC is applied in the dentistry. One of the reasons is the additive of fluoride ion in the CFAS glass system act as an anti-cariogenic agent which is can prevent the formation of plaque on the surface of the enamel (Wiegand et al., 2007; Moreau and Xu, 2010). Anti-cariogenic effects of fluoride involved variety of mechanisms including enhancement of remineralization, reduction of demineralization, interference of pellicle and inhibition of microbial growth in the oral cavity (Featherstone and Ten Cate, 1988; Fejerskov and Ckarkson, 1996; Hamilton, 1996; Rölla and Ekstrand, 1996). In addition, it is also noted that fluoride ions present in blood plasma and saliva are essential for normal development in human body especially for hard tissue development (Jha et al., 1997; Stanton and Hill, 2000).
The glass polyalkenoate cement or commonly known as GIC was first introduced in the late 1960's by Wilson and Kent at the English Laboratory of the Government Chemist, London (Wilson and Kent, 1971; Nicholson et al, 1998; Khoroushi et al., 2013; De Caluwé et al., 2017). From a previous report, the GIC was invented and developed for dental application in the response to the increasing cases of tooth decay (Forss et al., 2013). Dental caries causes serious tooth problems especially pits and fissures on occlusal surfaces among children and adolescents. Thus, the GIC is widely used in various clinical branches of dental application such as restorative, sealant, luting, base and liner to prevent the caries effect problem (Yli-Urpo et al., 2005; Khoroushi and Keshani, 2013).

The GIC is water-based cement which is formed by the reaction of acid and base between aqueous polymer (acid) and glass powder (Wilson and Kent, 1971). According to Fennell and Hill (2001), the most of glasses used for GIC are based on the calcium-aluminosilicate glass system reported by Wilson and Kent. Commonly, the additive of fluoride ion, F^- and PO_4^{3-} were added to the glass system to improve the properties of the GIC for dental application (De Caluwé et al., 2017). The present of fluoride ion in the glass system increases the mechanical properties of GIC especially in compressive strength but decreases the setting time of cement. While, phosphate group, PO_4^{3-} increases the working time and setting time but decrease the compressive strength of GIC (De Caluwé et al., 2017).

The setting reaction of GIC occurs as soon as the glass powder and liquid are mixed. During the acid-base reaction, ions such Ca^{2+}, Al^{3+} and F^- are leaching out from the glass powder and then, cross-linking between polyacid and the surface of the calcium-aluminosilicate glass is immediately occurred (Nicholson et al, 1998). At this stage, the water molecule being an important role for hardening process. The reaction between metal ion on the glass surface continued to linking with polyacid slowly against the aging time up to years. As the reaction progress, the hydrogen bond originated from polymer acid in the liquid is replaced by a metal ionic bond. Then, formation of gelation is formed and the GIC started to hardening against aging time. From the previous researchers, the mechanical properties of the GIC depend on the type of the initial glass powder and aqueous polyacid (Yli-Urpo et al., 2005; Khoroushi and Keshani, 2013).

1.3 Problem Statement

In recent years, glass, glass-ceramics, and ceramics have widely attracted prodigious interest in a medical material application due to their properties toward the biological reaction (Rezaie et al., 2015). This kind of material is a biomaterial, which shows the compatibility properties in a biological system. In the medical, biomaterials such as bioglass are commonly used in various applications such as medicine, dentistry and orthopedic (Skallevold et al., 2019). Among the bioglass that received the most attention in dentistry was the CFAS glass with the SiO_2–CaO–CaF_2–Al_2O_3–P_2O_5 glass system due to similarity to enamel composition crystallized into FA crystal and consist of fluoride ion to prevent the formation of caries from the microbial growth.

At present, the manufacturing cost for medical applications, especially in dentistry is increased highly over the years due to the increasing cost of production, including the cost of raw materials, labor, manufacture, and development. Due to the high-cost production of bioglass, especially for CFAS glass, the compound of SiO_2 and CaO can be replaced by alternative potential sources to utilize the unwanted materials as one way of recycling.

Usually, the primary compound to produce glass is a pure raw material of SiO_2 compound. Pure SiO_2 is a very expensive cost, and it has a high melting point. Thus, by considering the SLS glass, it consists of a SiO_2 compound with a low cost of raw materials and low melting point compared to the pure SiO_2. According to previous research, SLS glass acts as an alternative source of SiO_2 to produce a new optical application production. CS is also a waste material that is rich in calcium sources. CS is easily obtained from various seashell types and not used causes it is low in cost. However, CS can be contaminated with heavy metals from factory waste into the sea. As a precautionary step, avoid using or collecting CS from the factory areas near the seaside for dental material applications. In addition, CS can be tested using XRF analysis to confirm the presence of heavy metals contamination.

Therefore, the usability of SLS glass and CS act as the SiO_2 and CaO sources in producing CFAS glass is a suitable way to reduce the production cost and indirectly reduce the landfill waste materials. Besides, there are limited reports and systematic research of physical, structural, thermal, and morphology properties of CFAS glass system derived from vitreous waste materials such as SLS glass and CS. In addition, there are no optimum properties of CFAS glass has been reported especially mechanical properties which are important to apply in dental application.

The fabrication of GIC derived from the CFAS glass by using partially waste material is one of the opportunities to discover the behavior of the GIC itself. GIC is usually used widely in various dental applications for the past 50 years, such as luting, restorative, sealant, lining, and base. Before the existence of GIC, amalgam is made of alloy metal and mercury liquid. It is most widely used to fill a cavity caused by tooth decay. Until now, the uses of amalgam in dentistry are still gaining popularity due to its low cost, ease of application, durability against force, and high strength in mechanical properties. Nevertheless, the uses of amalgam are declining in developed countries. One reason behind this is amalgam has a controversial issue due to the concern about mercury ion release and can cause toxicity in the human body. The GIC seen as the most advantageous and suitable material compared to other materials that used in the dental application.

Basically, GIC shows impressive characteristics such as biocompatible, bioactive, esthetical, anti-cariogenic, rough surface texture, and low solubility. However, GIC has poor mechanical properties. Thus, several studies on GIC related to modifying glass phases to improve the GIC properties, especially in mechanical properties. Besides that, the setting reaction is an important part where gives an effect to the properties of GIC, especially to mechanical properties. The setting reaction started when the glass powder is mix with PAA and then aging at a specific time such as 7, 14, 21, and 28 days in the deionized water. It generally weak and not stable in water after setting reaction; however, as the progression of reaction is complete, it becomes stronger and more resistant against moisture. The previous research reported that the longer time aging of GIC could improve the resistance to moisture, the cement's compressive strength. Another factor that can be considered to enhance the properties of GIC is the ratio of Glass:Polymer:Water during the setting reaction. The ratio is very important, especially during the cement's initial reaction due to the fast reaction between acid and base reaction. Water is the role important in setting time and aging time of GIC. Thus, previous researchers' various ratio of initial water was studied to identify water's role in the fabrication of GIC.

For that reason, this research has focused on the comprehensive study of GIC fabricated from precursor CFAS glass system to use as a biomaterial in dental material for wide application. In this study, the compound's suitable composition to produce the CFAS glass with SLS−CS−CaF_2−Al_2O_3−P_2O_5 glass system is modified based on the previous research. Two series of compositions prepare the CFAS glass to determine the effect of SLS glass, CS, and CaF_2 in the glass system. Moreover, the initial water ratios while fabricating the GIC from the CFAS glass vary at suitable values. Furthermore, the fabricated GIC has been experienced an aging time from 7 to 28 days to improve the physical, structural, and mechanical properties of GIC. The antibacterial activity of GIC related to fluoride release during the aging time will identify as an excellent potential of the GIC in this research. Thus, this study's findings will anticipate finding a potential application as a dental material application for various types of tooth restoration in dentistry.

1.4 Significant of Study

Nowadays, the dump of waste materials generated drastically and increased by every year is one of the problems for every country due to the limited space area of the landfill. The excessive and uncontrolled dumping of waste materials causes environmental pollution consequently the spread of infectious diseases. The usability of waste material such as SLS glass and CS are widely reported by other researchers as precursor of SiO_2 and CaO to respectively to reduce solid wastes. A great deal of research has been focused on the preparing and characterization of the SLS−CS−CaF_2−Al_2O_3−P_2O_5 based glass system using commercial high-grade chemical composition for dental application such as restorative, filling, luting, liner and base for enamel defect. In this study, the CFAS glass based on SLS−CS−CaF_2−Al_2O_3−P_2O_5 glass system is prepared using waste materials by replacing SiO_2 and CaO to SLS glass and CS respectively.

The GIC has been identified as the most widely uses of restorative material besides amalgam in dentistry. The major component in the GIC is consists of glass powder and polymer acid that can be changed based on its suitable function. The glass system commonly used as the glass component in the GIC is aluminosilicate glass. The additional elements are added to the glass system to improve the performance of the GIC such as calcium, fluorine, zinc, phosphate, lithium and others. Besides that, the presence of the crystal phase in the glass system of GIC might be can enhanced the properties of the GIC itself such as HA and FA crystal which is similar to the composition of enamel. According to the previous researches, the presence of the crystal phase in the glass system will be improved the mechanical properties of the GIC and it is very useful for the dental application.

To the best of the author’s knowledge, there are very few available literature reports on the production of CFAS glass using waste materials such as SLS glass and CS. In the present research, the CFAS based glass system is used as the initial glass for the fabrication of GIC. The presence of the FA crystal phase in the CFAS glass system is expected to provide significant useful in the physical, structural, thermal and mechanical properties of the GIC. Furthermore, the GIC is fabricated with varying different initial ratios of water and aging times in order to enhance the properties and the quality of the GIC as the final products. Consequently, it is expected that the optimum physical, structural and mechanical properties as well as antibacterial activity properties of GIC derived from CFAS glass have a high potential for various dental material applications. It is also could contribute as new knowledge in modifying GIC for the dentistry area.

1.5 Research Objectives

The main objective of this study is to develop and enhance the GIC which fabricated from the CFAS glass and PAA with specific ratio of water. Two series of CFAS glass based on SLS–CS–CaF_2–Al_2O_3–P_2O_5 system are designed with different glass composition. The working objectives of this research are:

1) To synthesis the CFAS glass from combination of commercial grade chemicals and vitreous waste materials via conventional melt-quenching technique.
2) To study the effect of difference batch formulation of SLS glass, CS and CaF_2 toward CFAS glass system on structural, thermal and microstructural properties.
3) To investigate the influences of initial ratios of water and aging times on physical, structural and mechanical properties of GIC derived from CFAS glass system.
4) To evaluate antibacterial properties of GIC against *S. mutans*.

1.6 Scope of Study

In order to achieve the objective of this study, the scopes of the study as following:

1) A series of precursor CFAS glass (Series 1) based on the stoichiometric equation of $[(45\text{-}x)SLS \cdot xCS \cdot 15CaF_2 \cdot 20Al_2O_3 \cdot 20P_2O_5]$ where x = 5, 10, 15 and 20 (wt.%), has been prepared using SLS glass, CS, CaF_2, Al_2O_3 and P_2O_5 powder by conventional melt-quenching method.
2) A series of precursor CFAS glass (Series 2) based on the stoichiometric equation of $[(45\text{-}x)SLS \cdot 15CS \cdot xCaF_2 \cdot 20Al_2O_3 \cdot 20P_2O_5]$ where x = 5, 10, 15 and 20 (wt.%), has been prepared using SLS glass, CS, CaF_2, Al_2O_3 and P_2O_5 powder by conventional melt-quenching method.
3) The chemical composition of SLS glass and CS are been measured using the XRF spectroscopy in order to confirm the percentage of the chemical oxide before use in the synthesis of CFAS glass.
4) The thermal properties of CFAS glass have been measured using DSC and TGA spectroscopy in order to identify the glass transition temperature (T_g) and glass crystallization temperature (T_c)
5) The chemical composition of the CFAS glass has been measured using XRF and EDX spectroscopy in order to confirm the chemical compound in weight percentage.
6) The GIC has been fabricated from the CFAS glass system in Series 1 and Series 2 at different initial ratios of water and aging times between 7 to 28 days.
7) The physical, structural, thermal, morphology and mechanical properties of CFAS glass and GIC have been analyzed using the Archimedes method, molar volume equation, XRD, FTIR, Raman spectroscopy, DSC/TGA, FESEM, EDX, Vickers microhardness and Universal testing machine for compressive strength.

8) The optimum GIC sample based on the structural and mechanical properties has been measured for fluoride ion release by ion chromatography and tested for antibacterial activity by disk diffusion method.

In the current study, two different series of CFAS glass are prepared from a combination of the high grade of commercial chemical and vitreous waste materials. Series 1 was varying the weight composition of SLS and CS while Series 2 varying the weight composition of SLS and CaF_2. Both SLS glass and CS act as compound sources of SiO_2 and CaO respectively. The CFAS glass is synthesized by a conventional melt-quenching technique at a temperature of 1450 °C. The properties of SLS glass, CS and CFAS glass have been characterized such as structural, thermal and morphological in order to study the behaviour of CFAS glass derived from waste materials. Next, the fabrication of GIC is derived from CFAS glass in Series 1 and Series 2 experiences two conditions which are varying the initial ratios of water and the aging times. The initial ratio of water for fabricating GIC was controlled at 1.0, 1.2 and 1.4. While, the period of aging time for GIC in the deionized water was varies at 7, 14, 21 and 28 days. The temperature of aging time is also controlled at 37 °C. The structural, thermal, morphological, and mechanical properties of GIC for both Series 1 and Series 2 are characterized to determine the optimum properties of GIC. The optimum condition of GIC in mechanical properties is selected for further characterization based on the results of Vickers microhardness and compressive strength.

The selected GIC was characterized with further study on physical properties, fluoride ion release and antibacterial activity. The structural, thermal, morphological, physical properties in this study were analyzed by X-ray Fluorescence (XRF), X-ray Diffraction (XRD), Fourier Transform Infrared (FTIR) and Raman Spectroscopy, Differential Scanning Calorimetry (DSC), Thermogravimetric Analysis (TGA), Field Emission Scanning Electron Microscopy (FESEM), Energy Dispersive X-Ray Analysis (EDX), density and molar volume. While, the fluoride ion release and antibacterial activity of the GIC was determined by Ion Chromatography and antimicrobial disk method respectively.

1.7 Outline of Thesis

The thesis designed and arranged as follows. Chapter 1 gives an introduction background of CFAS glass, FA and GIC derived from vitreous waste (SLS glass and CS). This chapter also including the problem statements, the objectives of research, the scopes of study and also the significant of the study. The review of the previous and current works reported on the uses of SLS glass and CS as a precursor, production of CFAS glass and fabrication GIC has been discussed in Chapter 2. This chapter also reviewed the physical, structural, thermal, mechanical and antibacterial properties of CFAS and GIC for current and previous works. The details on research designed, methodology and characterization used in this study were described in Chapter 3. Next, Chapter 4 represent the results and discussions concerning on the properties of CFAS glass in Series 1 and Series 2 with different chemical composition of SLS glass, CS and CaF_2. This chapter also represent the findings and focusing on the properties of GIC derived from CFAS glass in Series 1 and Series 2 at different initial ratios of water and aging times. Besides that, the physical, structural, mechanical and antibacterial properties of CFAS glass and GIC are deeply discussed in

Chapter 4. Finally, the conclusion had been made based on the results obtained from Chapter 4, the suggestion and recommendation for future works presented in Chapter 5. The references, publications and conferences attended by the author were placed at the last part of the thesis.

REFERENCES

Abd Manaf, L., Samah, M. A. A., & Zukki, N. I. M. (2009). Municipal solid waste management in Malaysia: Practices and challenges. *Waste Management*, 29(11), 2902-2906.

Al-Bader, R. M., Ziadan, K. M., & Al-Ajely, M. (2015). New glass compositions based on calcium-fluoro aluminosilicate for dental composite. *Journal of Advances in Chemistry*, 10(5), 2743-2752.

Al-Eesa, N. A., Johal, A., Hill, R. G., & Wong, F. S. L. (2018). Fluoride containing bioactive glass composite for orthodontic adhesives—Apatite formation properties. *Dental Materials*, 34(8), 1127-1133.

Al-Eshaikh, M. A., & Kadachi, A. (2011). Elemental analysis of steel products using X-ray fluorescence (XRF) technique. *Journal of King Saud University-Engineering Sciences*, 23(2), 75-79.

Al-Sanabani, J. S., Madfa, A. A., & Al-Sanabani, F. A. (2013). Application of calcium phosphate materials in dentistry. *International Journal of Biomaterials*, 2013, 1-13.

Ali, S., Farooq, I., Al-Thobity, A. M., Al-Khalifa, K. S., Alhooshani, K., & Sauro, S. (2019). An in-vitro evaluation of fluoride content and enamel remineralization potential of two toothpastes containing different bioactive glasses. *Bio-Medical Materials and Engineering*, 30(5-6), 487-496.

Almasri, K. A., Matori, K. A., & Zaid, M. H. M. (2017). Effect of sintering temperature on physical, structural and optical properties of wollastonite-based glass-ceramic derived from waste soda lime silica glasses. *Results in Physics*, 7, 2242-2247.

Amin, M., Lim, L. W., & Takeuchi, T. (2008). Determination of common inorganic anions and cations by non-suppressed ion chromatography with column switching. *Journal of Chromatography A*, 1182(2), 169-175.

American Public Health Association, American Water Works Association, Water Pollution Control Federation & Water Environment Federation. (2012). 22nd Edition. *Standard methods for the examination of water and wastewater*. Washington DC, American Public Health Association.

Ananthanarayanan, A., Kothiyal, G. P., Montagne, L., Tricot, G., & Revel, B. (2011). The effect of P_2O_5 on the structure, sintering and sealing properties of barium calcium aluminum boro-silicate (BCABS) glasses. *Materials Chemistry and Physics*, 130(3), 880-889.

Anstice, H. M., & Nicholson, J. W. (1995). Investigation of the post-hardening reaction in glass-ionomer cements based on poly (vinyl phosphonic acid). *Journal of Materials Science: Materials in Medicine*, 6(7), 420-425.

Awang-Hazmi, A. J., Zuki, A. B. Z., Noordin, M. M., Jalila, A., & Norimah, Y. (2007). Mineral composition of the cockle (Anadara granosa) shells of west coast of peninsular Malaysia and its potential as biomaterial for use in bone repair. *Journal of Animal and Veterinary Advances*, 591-594.

Azami, M., Jalilifiroozinezhad, S., Mozafari, M., & Rabiee, M. (2011). Synthesis and solubility of calcium fluoride/hydroxy-fluorapatite nanocrystals for dental applications. *Ceramics International*, 37(6), 2007-2014.

Aziz, S. Q., Aziz, H. A., Bashir, M. J. K., & Mojiri, A. (2015). Assessment of various tropical municipal landfill leachate characteristics and treatment opportunities. *Global NEST Journal*, 17(3), 439-450.

Baig, M. S., & Fleming, G. J. (2015). Conventional glass-ionomer materials: a review of the developments in glass powder, polyacid liquid and the strategies of reinforcement. *Journal of Dentistry*, 43(8), 897-912.

Baino, F., & Fiume, E. (2019). Elastic mechanical properties of 45S5-based bioactive glass–ceramic scaffolds. *Materials*, 12(19), 3244-3256.

Bakar, M. K. S., Othman, N. H., Sainudin, M. S., Khalid, F. S., & Muthusamy, K. (2020). Utilisation of mussel shell ash and palm oil leaves ash as admixture in concrete. *International Journal of Integrated Engineering*, 12(8), 310-317.

Barandehfard, F., Rad, M. K., Hosseinnia, A., Khoshroo, K., Tahriri, M., Jazayeri, H. E., Moharamzadeh, K. & Tayebi, L. (2016). The addition of synthesized hydroxyapatite and fluorapatite nanoparticles to a glass-ionomer cement for dental restoration and its effects on mechanical properties. *Ceramics International*, 42(15), 17866-17875.

Barbusinski, K., Kalemba, K., Kasperczyk, D., Urbaniec, K., & Kozik, V. (2017). Biological methods for odor treatment–A review. *Journal of Cleaner Production*, 152, 223-241.

Barry, T. I., Clinton, D. J., & Wilson, A. D. (1979). The structure of a glass-io nomer cement and its relationship to the setting process. *Journal of Dental Research*, 58(3), 1072-1079.

Barnes, P., & Bensted, J. (2002). *Structure and performance of cements*. CRC Press.

Basu, B., Sabareeswaran, A., & Shenoy, S. J. (2015). Biocompatibility property of 100% strontium-substituted SiO_2–Al_2O_3–P_2O_5–CaO–CaF_2 glass ceramics over 26 weeks implantation in rabbit model: Histology and micro-Computed Tomography analysis. *Journal of Biomedical Materials Research Part B: Applied Biomaterials*, 103(6), 1168-1179.

Behr, M., Rosentritt, M., Loher, H., Kolbeck, C., Trempler, C., Stemplinger, B., Kopzon, V. & Handel, G. (2008). Changes of cement properties caused by mixing errors: the therapeutic range of different cement types. *Dental Materials*, 24(9), 1187-1193.

Best, S. M., Porter, A. E., Thian, E. S., & Huang, J. (2008). Bioceramics: past, present and for the future. *Journal of the European Ceramic Society*, 28(7), 1319-1327.

Bernardo, E., Esposito, L., Rambaldi, E., Tucci, A., & Hreglich, S. (2008). Recycle of waste glass into glass–ceramic stoneware. *Journal of the American Ceramic Society*, 91(7), 2156-2162.

Bharatham, B. H., Bakar, A., Zuki, M., Perimal, E. K., Yusof, L. M., & Hamid, M. (2014). Development and characterization of novel porous 3D alginate-cockle shell powder nano biocomposite bone scaffold. *BioMed Research International*, 2014, 1-11.

Bianco, A., Cacciotti, I., Lombardi, M., Montanaro, L., Bemporad, E., & Sebastiani, M. (2010). F-substituted hydroxyapatite nanopowders: thermal stability, sintering behaviour and mechanical properties. *Ceramics International*, 36(1), 313-322.

Biswas, A., Wang, T., & Biris, A. S. (2010). Single metal nanoparticle spectroscopy: optical characterization of individual nanosystems for biomedical applications. *Nanoscale*, 2(9), 1560-1572.

Buasri, A., Chaiyut, N., Loryuenyong, V., Worawanitchaphong, P. & Trongyong, S. (2013). Calcium oxide derived from waste shells of mussel, cockle, and scallop as the heterogeneous catalyst for biodiesel production. *The Scientific World Journal*, 2013, 1-7.

Bulánek, R., Čičmanec, P., & Setnička, M. (2013). Possibility of VOx/SiO_2 complexes speciation: comparative multi-wavelength Raman and DR UV-vis study. *Physics Procedia*, 44, 195-205.

Bogdanov, B. I., Pashev, P. S., Hristov, J. H., & Markovska, I. G. (2009). Bioactive fluorapatite-containing glass ceramics. *Ceramics International*, 35(4), 1651-1655.

Boey, P. L., Maniam, G. P., Abd Hamid, S., & Ali, D. M. H. (2011). Utilization of waste cockle shell (Anadara granosa) in biodiesel production from palm olein: optimization using response surface methodology. *Fuel*, 90(7), 2353-2358.

Bovo N. (2007). *Structure—properties relationships in bioactive glasses for PAA-based polyalkenoate cements*. Doctoral dissertation, Imperial College London, United Kingdom.

Calver, A., Hill, R. G., & Stamboulis, A. (2004). Influence of fluorine content on the crystallization behavior of apatite-wollastonite glass-ceramics. *Journal of Materials Science*, 39(7), 2601-2603.

Campillo, M., Lacharmoise, P. D., Reparaz, J. S., Goni, A. R., & Valiente, M. (2010). On the assessment of hydroxyapatite fluoridation by means of Raman scattering. *The Journal of Chemical Physics*, 132(24), 244501.

Canillas, M., Pena, P., Antonio, H., & Rodríguez, M. A. (2017). Calcium phosphates for biomedical applications. *Boletín de la Sociedad Española de Cerámica y Vidrio*, 56(3), 91-112.

Casasola, R., Rincón, J. M., & Romero, M. (2012). Glass-ceramic glazes for ceramic tiles: a review. Journal of Materials Science, 47(2), 553-582.

Cattani-Lorente, M. A., Godin, C., & Meyer, J. M. (1994). Mechanical behavior of glass ionomer cements affected by long-term storage in water. *Dental Materials*, 10(1), 37-44.

Cao, W., & Hench, L. L. (1996). Bioactive materials. *Ceramics International*, 22(6), 493-507.

Carter, C. B. & Norton, M. G. (2007). *Ceramic Materials*. New York: Springer.

Chee, S. Y., Devakie, M. N., & Azizah, M. S. (2011). Phylogenetic study and barcoding of the blood cockle, Tegillarca granosa, found on the west coast of peninsular Malaysia using the COI gene. *Genetics and Molecular Research*, 10(2), 1237-1244.

Chemistry of Glass. (2016 July 16). Architectural Glass Newsletters-Pilkington NSG. Retrieved from https://www.pilkington.com/en/global /about/education/chemistry-of-glass.

Chen, Y., & Miao, X. (2005). Thermal and chemical stability of fluorohydroxyapatite ceramics with different fluorine contents. *Biomaterials*, 26(11), 1205-1210.

Chen, X., Hench, L. L., Greenspan, D., Zhong, J., & Zhang, X. (1998). Investigation on phase separation, nucleation and crystallization in bioactive glass-ceramics containing fluorophlogopite and fluorapatite. *Ceramics International*, 24(5), 401-410.

Chen, X., Chen, X., Brauer, D. S., Wilson, R. M., Hill, R. G., & Karpukhina, N. (2014). Novel alkali free bioactive fluorapatite glass ceramics. *Journal of Non-Crystalline Solids*, 402, 172-177.

Chen, Z. W., Gibson, W. M., & Huang, H. (2008). High-definition x-ray fluorescence: principles and techniques. *X-ray Optics and Instrumentation*, 2008, 1-10.

Chen, J., Wen, Z., Zhong, S., Wang, Z., Wu, J., & Zhang, Q. (2015). Synthesis of hydroxyapatite nanorods from abalone shells via hydrothermal solid-state conversion. *Materials & Design*, 87, 445-449.

Chimalawong, P., Kirdsiri, K., Kaewkhao, J., & Limsuwan, P. (2012). Investigation on the physical and optical properties of Dy^{3+} doped soda-lime-silicate glasses. *Procedia Engineering*, 32, 690-698.

Clifford, A., & Hill, R. (1996). Apatite-mullite glass-ceramics. *Journal of Non-Crystalline Solids*, 196, 346-351.

Clifford, A., Hill, R., Rafferty, A., Mooney, P., Wood, D., Samuneva, B., & Matsuya, S. (2001). The influence of calcium to phosphate ratio on the nucleation and crystallization of apatite glass-ceramics. *Journal of Materials Science: Materials in Medicine*, 12(5), 461-469.

Cook, W. D. (1982). Dental polyelectrolyte cements I: chemistry of the early stages of the setting reaction. *Biomaterials*, 3(4), 232-236.

Coats, A. W., & Redfern, J. P. (1963). Thermogravimetric analysis a review. *Analyst*, 88(1053), 906-924.

Crisp, S., Lewis, B. G., & Wilson, A. D. (1976a). Characterization of glass-ionomer cements: 2. Effect of the powder: liquid ratio on the physical properties. *Journal of Dentistry*, 4(6), 287-290.

Crisp, S., Lewis, B. G., & Wilson, A. D. (1976b). Glass ionomer cements: chemistry of erosion. *Journal of Dental Research*, 55(6), 1032-1041.

Crisp, S., Lewis, B. G., & Wilson, A. D. (1977). Characterization of glass-ionomer cements: 3. Effect of polyacid concentration on the physical properties. *Journal of Dentistry*, 5(1), 51-56.

Croll, T. P., & Nicholson, J. W. (2002). Glass ionomer cements in pediatric dentistry: review of the literature. *Pediatric Dentistry*, 24(5), 423-429.

Cubillas, P., Köhler, S., Prieto, M., Chaïrat, C., & Oelkers, E. H. (2005). Experimental determination of the dissolution rates of calcite, aragonite, and bivalves. *Chemical Geology*, 216(1-2), 59-77.

Czarnecka, B., Limanowska-Shaw, H., & Nicholson, J. W. (2002). Buffering and ion-release by a glass-ionomer cement under near-neutral and acidic conditions. *Biomaterials*, 23(13), 2783-2788.

Das, S., Lee, S. H., Kumar, P., Kim, K. H., Lee, S. S., & Bhattacharya, S. S. (2019). Solid waste management: Scope and the challenge of sustainability. *Journal of Cleaner Production*, 228, 658-678.

Davidson, C. L. (2006). Advances in glass-ionomer cements. *Journal of Applied Oral Science*, 14, 3-9.

De Aza, P. N., De Aza, A. H., Pena, P., & De Aza, S. (2007). Bioactive glasses and glass-ceramics. *Boletin-Sociedad Espanola De Ceramica Y Vidrio*, 46(2), 45-55.

De Caluwé, T., Vercruysse, C. W. J., Ladik, I., Convents, R., Declercq, H., Martens, L. C., & Verbeeck, R. M. H. (2017). Addition of bioactive glass to glass ionomer cements: Effect on the physico-chemical properties and biocompatibility. *Dental Materials*, 33(4), 186-203.

De Maeyer, E. A. P., Verbeeck, R. M. H., & Vercruysse, C. W. J. (1998). Reactivity of fluoride-containing calcium aluminosilicate glasses used in dental glass-ionomer cements. *Journal of Dental Research*, 77(12), 2005-2011.

DeSchepper, E. J., Thrasher, M. R., & Thurmond, B. A. (1989). Antibacterial effects of light-cured liners. *American Journal of Dentistry*, 2(3), 74-76.

Dimitrova-Lukacs, M., & Gillemot, L. (1993). Bioactive-bioinert composite ceramics. *Third Euro-Ceramic*, 3, 179-84.

Duan, R. G., & Liang, K. M. (1998). A study on the crystallization of $CaO–Al_2O_3–SiO_2$ system glasses. *Journal of Materials Processing Technology*, 75(1-3), 235-239.

Duminis, T. (2019). *Development and characterisation of novel fluorapatite glass-ceramics for use in glass ionomer cements*. Doctoral dissertation, Queen Mary University of London, United Kingdom).

Duverger, C., Nedelec, J. M., Benatsou, M., Bouazaoui, M., Capoen, B., Ferrari, M., & Turrell, S. (1999). Waveguide raman spectroscopy: a non-destructive tool for the characterization of amorphous thin films. *Journal of Molecular Structure*, 480, 169-178.

Diaz-Arnold, A. M., Vargas, M. A., & Haselton, D. R. (1999). Current status of luting agents for fixed prosthodontics. *The Journal of Prosthetic Dentistry*, 81(2), 135-141.

Dickey, B., Price, R., & Boyd, D. (2016). Evidence of a complex species controlling the setting reaction of glass ionomer cements. *Dental Materials*, 32(4), 596-605.

Dong, J., Ozaki, Y., & Nakashima, K. (1997). Infrared, Raman, and near-infrared spectroscopic evidence for the coexistence of various hydrogen-bond forms in poly (acrylic acid). *Macromolecules*, 30(4), 1111-1117.

Dorozhkin, S. V. (2013). A detailed history of calcium orthophosphates from 1770s till 1950. *Materials Science and Engineering: C*, 33(6), 3085-3110.

Ebrahimi-Kahrizsangi, R., Nasiri-Tabrizi, B., & Chami, A. (2011). Characterization of single-crystal fluorapatite nanoparticles synthesized via mechanochemical method. *Particuology*, 9(5), 537-544.

El-Ghannam, A. R. (2004). Advanced bioceramic composite for bone tissue engineering: Design principles and structure–bioactivity relationship. *Journal of Biomedical Materials Research Part A*, 69(3), 490-501.

El-Mallawany, R. (1993). Longitudinal elastic constants of tellurite glasses. *Journal of Applied Physics*, 73(10), 4878-4880.

Eliaz, N., & Metoki, N. (2017). Calcium phosphate bioceramics: a review of their history, structure, properties, coating technologies and biomedical applications. *Materials*, 10(4), 334-438.

Eslami, H., Solati-Hashjin, M., & Tahriri, M. (2009). The comparison of powder characteristics and physicochemical, mechanical and biological properties between nanostructure ceramics of hydroxyapatite and fluoridated hydroxyapatite. *Materials Science and Engineering: C*, 29(4), 1387-1398.

FAO (Food and Agricultural Organization) (2006). *Basic bivalve biology: taxonomy, anatomy and life history*. Food and Agricultural Organization of the United Nation. Retrieved at http://www.fao.org/docrep/007/y5720e/ y5720e07.htm.

Fahami, A., & Nasiri-Tabrizi, B. (2014). Mechanochemical behavior of $CaCO_3$–P_2O_5–CaF_2 system to produce carbonated fluorapatite nanopowder. *Ceramics International*, 40(9), 14939-14946.

Fathi, H., Johnson, A., van Noort, R., & Ward, J. M. (2005). The influence of calcium fluoride (CaF_2) on biaxial flexural strength of apatite–mullite glass–ceramic materials. *Dental Materials*, 21(9), 846-851.

Fantinel, F., Rieger, J., Molnar, F., & Hübler, P. (2004). Complexation of polyacrylates by Ca^{2+} ions. Time-resolved studies using attenuated total reflectance Fourier transform infrared dialysis spectroscopy. *Langmuir*, 20(7), 2539-2542.

Featherstone, J. D. (1999). Prevention and reversal of dental caries: role of low-level fluoride. *Community Dentistry and Oral Epidemiology*, 27(1), 31-40.

Featherstone, J. D. B., & Ten Cate, J. M. (1988). Physicochemical aspects of fluoride-enamel interactions. *Fluoride in Dentistry*, 1, 125-149.

Fennell, B., & Hill, R. G. (2001). The influence of poly (acrylic acid) molar mass and concentration on the properties of polyalkenoate cements Part I Compressive strength. *Journal of Materials Science*, 36(21), 5193-5202.

Fejerskov, O., & Clarkson, B. H. (1996). Dynamics of caries lesion formation. *Fluoride in Dentistry*, 187-213.

Feroz, S., & Khan, A. S. (2020). Fluoride-substituted hydroxyapatite. In A. S. Khan, & A. A. Chaudhry (Eds.), *Handbook of Ionic Substituted Hydroxyapatites* (pp. 175-196). Woodhead Publishing.

Fiume, E., Barberi, J., Verné, E., & Baino, F. (2018). Bioactive glasses: from parent 45S5 composition to scaffold-assisted tissue-healing therapies. *Journal of Functional Biomaterials*, 9(1), 24-50.

Fini, M., Savarino, L., Aldini, N. N., Martini, L., Giavaresi, G., Rizzi, G., Martini, D., Ruggeri, A., & Giardino, R. (2003). Biomechanical and histomorphometric investigations on two morphologically differing titanium surfaces with and without fluorohydroxyapatite coating: an experimental study in sheep tibiae. *Biomaterials*, 24(19), 3183-3192.

Fleming, G. J., Farooq, A. A., & Barralet, J. E. (2003). Influence of powder/liquid mixing ratio on the performance of a restorative glass-ionomer dental cement. *Biomaterials*, 24(23), 4173-4179.

Francis Thoo, V. W., Zainuddin, N., Matori, K. A., & Abdullah, S. A. (2013). Studies on the potential of waste soda lime silica glass in glass ionomer cement production. *Advances in Materials Science and Engineering*, 2013, 1-6.

Francisconi, L. F., Scaffa, P. M. C., Barros, V. R. D. S. P. D., Coutinho, M., & Francisconi, P. A. S. (2009). Glass ionomer cements and their role in the restoration of non-carious cervical lesions. *Journal of Applied Oral Science*, 17(5), 364-369.

Freire, W. P., Fook, M. V. L., Barbosa, E. F., Araújo, C. S., Barbosa, R. C., & de Sousa, W. J. (2015). Glass Ionomer Cement–Development and Characterization Microstructural. *Materials Science Forum*, 805,12-18.

Forss, H., Walsh, T., Hiiri, A., Nordblad, A., Mäkelä, M., & Hv, W. (2013). Sealants for preventing dental decay in the permanent teeth. *Cochrane Database of Systematic Reviews*, 2013(3), CD001830.

Forsten, L. (1998). Fluoride release and uptake by glass-ionomers and related materials and its clinical effect. *Biomaterials*, 19(6), 503-508.

Fu, K., Wang, S., Wang, G., & Wang, Y. (2018). The effects of calcium oxide on fluorapatite crystal morphology and mechanical property of functional glass-ceramics. *Ceramics International*, 44(16), 20531-20538.

Gunasekaran, A. Pandi, 2006. Raman and infrared spectra of carbonates of calcite structure. *Journal of Raman Spectroscopy*, 37(9), 892-899.

Gardiner, D. J. (1989). Introduction to Raman scattering. In Gardiner, D. J., & Pierre, R. (Eds.), *Practical Raman Spectroscopy* (pp. 1-12). Springer.

Gualtieri, M. L., Mugoni, C., Guandalini, S., Cattini, A., Mazzini, D., Alboni, C., & Siligardi, C. (2018). Glass recycling in the production of low-temperature stoneware tiles. *Journal of Cleaner Production*, 197, 1531-1539.

Germaine, G. R., & Tellefson, L. M. (1986). Role of the cell membrane in pH-dependent fluoride inhibition of glucose uptake by Streptococcus mutans. *Antimicrobial Agents and Chemotherapy*, 29(1), 58-61.

Grayson, K. (2019, September 16). Using Glass Modifiers to Change Glass Characteristics. [Web log post]. Retrieved from https://mo-sci.com/using-glass-modifiers/.

Griffin, S. G., & Hill, R. G. (1999). Influence of glass composition on the properties of glass polyalkenoate cements. Part I: influence of aluminium to silicon ratio. *Biomaterials*, 20(17), 1579-1586.

Griebenow, K., Bragatto, C. B., Kamitsos, E. I., & Wondraczek, L. (2018). Mixed-modifier effect in alkaline earth metaphosphate glasses. *Journal of Non-Crystalline Solids*, 481, 447-456.

Hamilton I.R., Bowden G.H.W. (1996). Fluoride effects on oral bacteria. In O. Fejerskov, J. Ekstrand, B. A. Burt (Eds.), *Fluoride in dentistry*. (2nd ed., pp.230-251). Copenhagen: Munksgaard.

Han, Z. X., & Wang, C. (2007). Determination of inorganic anions in drinking water by on chromatography. *Journal of Environment and Health*, (9), 1-5.

Hanna, W. A., Gharib, F. E., & Marhoon, I. I. (2011). Characterization of ceramic filled polymer matrix composite used for biomedical applications. *Journal of Minerals and Materials Characterization and Engineering*, 10(12), 1167-1178.

Hanifi, A. R., Genson, A., Pomeroy, M. J., & Hampshire, S. (2012). Independent but additive effects of fluorine and nitrogen substitution on properties of a calcium aluminosilicate glass. *Journal of the American Ceramic Society*, 95(2), 600-606.

Hart, A. (2020). Mini-review of waste shell-derived materials' applications. *Waste Management & Research*, 38(5), 514-527.

Harabi, A., Zaiou, S., Guechi, A., Foughali, L., Harabi, E., Karboua, N. E., Zouai, S., Mezahi, F.Z. & Guerfa, F. (2017). Mechanical properties of anorthite based ceramics prepared from kaolin DD2 and calcite. *Cerâmica*, 63(367), 311-317.

Hasanuzzaman, M., Rafferty, A., Sajja, M., & Olabi, A. G. (2016). Production and treatment of porous glass materials for advanced usage. *Reference Module in Materials Science and Materials Engineering*, 2016, 1-11.

Hejda, P., Holubová, J., Černošek, Z., & Černošková, E. (2017). The structure and properties of vanadium zinc phosphate glasses. *Journal of Non-Crystalline Solids*, 462, 65-71.

Hussain, F., & Ali, A. (2010). Elastic and plastic properties of soda lime glass by micro-indentation. *In Key Engineering Materials*, 442, 294-300.

Hupa, L. (2011). Melt-derived bioactive glasses. In H. O. Ylänen (Ed.), *Bioactive Glasses* (pp. 3-28). Woodhead Publishing.

Hautman, D. P. (1997). Determination of inorganic anions in drinking water by ion chromatography. *US Environmental Protection Agency*, 300(1) 1-36.

Hao, L., Gao, W., Yan, S., Niu, M., Liu, G., & Hao, H. (2019). Preparation and characterization of porous ceramics with low-grade diatomite and oyster shell. *Materials Chemistry and Physics*, 235, 121741- 121750.

Headley, T. J., & Loehman, R. E. (1984). Crystallization of a glass-ceramic by epitaxial growth. *Journal of the American Ceramic Society*, 67(9), 620-625.

Hembrick-Holloman, V., Samuel, T., Mohammed, Z., Jeelani, S., & Rangari, V. K. (2020). Ecofriendly production of bioactive tissue engineering scaffolds derived from egg-and sea-shells. *Journal of Materials Research and Technology*, 13729-13739.

Hench, L. L., & Paschall, H. A. (1974). Histochemical responses at a biomaterial's interface. *Journal of Biomedical Materials Research*, 8(3), 49-64.

Hench, L. L. (1991). Bioceramics: from concept to clinic. *Journal of the American Ceramic Society*, 74(7), 1487-1510.

Hench, L. L. (1993). *An introduction to bioceramics*. Singapore: World scientific.

Hench, L. L. (1998). Bioceramics, a clinical success. *American Ceramic Society Bulletin*, 77(7), 67-74.

Hench, L. L. (2013). Chronology of bioactive glass development and clinical applications. *New Journal of Glass and Ceramics*, 2(3), 1-7.

Hench, L. L. (2006). The story of Bioglass®. *Journal of Materials Science: Materials in Medicine*, 17(11), 967-978.

Hench, L. L., & Wilson, J. (1984). Surface-active biomaterials. *Science*, 226(4675), 630-636.

Hill, R. G. (2015). Bioactive Glass-Ceramics. In P. Duchey (Ed.), *Comprehensive Biomaterials Volume 1: Metallic, Ceramic and Polymeric Biomaterials* (pp. 181-187). Oxford: Elsevier.

Hill, R. G., Patel, M. & Wood, D. J. (1991). Preliminary studies on castable apatite-mullite glass-ceramics. In W. Bonfield, G. W., Hastings, & K. E. Tanner (Eds.), *Bioceramics: Proceedings of the 4th International Symposium on Ceramics in Medicine* (pp. 79-86). London: Butterworth-Heinemann.

Hill, R. G., & Wilson, A. D. (1988). A rheological study of the role of additives on the setting of glass-ionomer cements. *Journal of Dental Research*, 67(12), 1446-1450.

Hill, R., & Wood, D. (1995). Apatite-mullite glass-ceramics. *Journal of Materials Science: Materials in Medicine*, 6(6), 311-318.

Hill, R. G., Goat, C., & Wood, D. (1992). Thermal Analysis of a SiO_2- Al_2O_3-CaO-CaF_2 Glass. *Journal of the American Ceramic Society*, 75(4), 778-785.

Hill, R., Wood, D., & Thomas, M. (1999). Trimethylsilylation analysis of the silicate structure of fluoro-alumino-silicate glasses and the structural role of fluorine. *Journal of Materials Science*, 34(8), 1767-1774.

Höland, W., Rheinberger, V., & Frank, M. (1999). Mechanisms of nucleation and controlled crystallization of needle-like apatite in glass-ceramics of the SiO_2–Al_2O_3–K_2O–CaO–P_2O_5 system. *Journal of Non-Crystalline Solids*, 253(1-3), 170-177.

Höland, W., Rheinberger, V., Apel, E., van't Hoen, C., Höland, M., Dommann, A., Obrecht, M., Mauth, C., & Graf-Hausner, U. (2006). Clinical applications of glass-ceramics in dentistry. *Journal of Materials Science: Materials in Medicine*, 17(11), 1037-1042.

Höland, W., Rheinberger, V., Apel, E., Ritzberger, C., Eckert, H., & Mönster, C. (2007). Mechanisms of nucleation and crystallisation in high strength glass-ceramics. *Physics and Chemistry of Glasses-European Journal of Glass Science and Technology Part B*, 48(3), 97-102.

Hoque, M. E., Shehryar, M., & Islam, K. N. (2013). Processing and characterization of cockle shell calcium carbonate ($CaCO_3$) bioceramic for potential application in bone tissue engineering. *Journal of Material Sciences & Engineering*, 2(4), 132.

Holloway, D. G. (1973). *The physical properties of glass*. Wykeham: Pub. Ltd.

Hoornweg, D., & Bhada-Tata, P. (2012). *What a waste: a global review of solid waste management*. Washington, DC: World Bank.

Hudzicki, J. (2009). Kirby-Bauer disk diffusion susceptibility test protocol. *American Society for Microbiological*, 1-23.

Hudecki, A., Kiryczyński, G., & Łos, M. J. (2019). In M. J. Los, A. Hudecki, & E. Wiechec, (Eds.). *Biomaterials, Definition, Overview. In Stem Cells and Biomaterials for Regenerative Medicine* (pp. 85-98). Academic Press.

Hulbert, S. F., Hench, L. L., Forbers, D., & Bowman, L. S. (1982). History of bioceramics. *Ceramics International*, 8(4), 131-140.

Hussin M. (2016, June 21). Shellfish source of coastal fisheries. *Harian Metro*, Retrieved from https://www.hmetro.com.my/node/146872.

Iliadi, A., Baumgartner, S., Athanasiou, A. E., Eliades, T., & Eliades, G. (2014). Effect of intraoral aging on the setting status of resin composite and glass ionomer orthodontic adhesives. *American Journal of Orthodontics and Dentofacial Orthopedics*, 145(4), 425-433.

Islam, K. N., Bakar, M. Z. B. A., Noordin, M. M., Hussein, M. Z. B., Abd Rahman, N. S. B., & Ali, M. E. (2011). Characterisation of calcium carbonate and its polymorphs from cockle shells (Anadara granosa). *Powder Technology*, 213(1-3), 188-191.

ISO (International Organization for Standardization) (2007). *Dentistry-water-based cements—part 1: powder/liquid acid–base cements*. Geneva, Switzerland: International Organization for Standardization.

Jantová, S., Theiszová, M., Letašiová, S., Birošová, L., & Palou, T. M. (2008). In vitro effects of fluor-hydroxyapatite, fluorapatite and hydroxyapatite on colony formation, DNA damage and mutagenicity. *Mutation Research/Genetic Toxicology and Environmental Mutagenesis*, 652(2), 139-144.

Jha, L. J., Best, S. M., Knowles, J. C., Rehman, I., D Santos, J., & Bonfield, W. (1997). Preparation and characterization of fluoride-substituted apatites. *Journal of Materials science: Materials in Medicine*, 8(4), 185-191.

Jusoh, W. N. W., Matori, K. A., Zaid, M. H. M., Zainuddin, N., Khiri, M. Z. A., Rahman, N. A. A., Jalil, R.A. & Kul, E. (2019). Effect of sintering temperature on physical and structural properties of Alumino-Silicate-Fluoride glass ceramics fabricated from clam shell and soda lime silicate glass. *Results in Physics*, 12, 1909-1914.

Jung, S., Heo, N. S., Kim, E. J., Oh, S. Y., Lee, H. U., Kim, I. T., Hur, J., Lee, G. W., Lee, Y. C., & Huh, Y. S. (2016). Feasibility test of waste oyster shell powder for water treatment. *Process Safety and Environmental Protection*, 102, 129-139.

Kaewjaeng, S., Kothan, S., Chaiphaksa, W., Chanthima, N., Rajaramakrishna, R., Kim, H. J., & Kaewkhao, J. (2019). High transparency La_2O_3-CaO-B_2O_3-SiO_2 glass for diagnosis x-rays shielding material application. *Radiation Physics and Chemistry*, 160, 41-47.

Kamalanathan, P., Ramesh, S., Bang, L. T., Niakan, A., Tan, C. Y., Purbolaksono, J., Chandran & Teng, W. D. (2014). Synthesis and sintering of hydroxyapatite derived from eggshells as a calcium precursor. *Ceramics International*, 40(10), 16349-16359.

Kannan, S., Lemos, I. A. F., Rocha, J. H. G., & Ferreira, J. M. F. (2005). Synthesis and characterization of magnesium substituted biphasic mixtures of controlled hydroxyapatite/β-tricalcium phosphate ratios. *Journal of Solid-State Chemistry*, 178(10), 3190-3196.

Kalanatarifard, A., & Yang, G. S. (2012). Identification of the municipal solid waste characteristics and potential of plastic recovery at Bakri Landfill, Muar, Malaysia. *Journal of Sustainable Development*, 5(7), 11-17.

Karazi, S. M., Ahad, I. U., & Benyounis, K. Y. (2017). Laser Micromachining for Transparent Materials, In J. G. R. Sereni (Ed.), *Reference Module in Materials Science and Materials Engineering* (pp.1-13) Dublin, Ireland: Elsevier.

Kazuz, A., Radovanović, Ž., Veljović, D., Kojić, V., Miletić, V., Petrović, R., & Janaćković, D. (2020). α-Tricalcium phosphate/fluorapatite based composite cements: Synthesis, mechanical properties, and biocompatibility. *Ceramics International*, 46(16), 25149-25154.

Kent, B. E., Lewis, B. G., & Wilson, A. D. (1979). Glass ionomer cement formulations: I. The preparation of novel fluoroaluminosilicate glasses high in fluorine. *Journal of Dental Research*, 58(6), 1607-1619.

Khan, A. S., Aamer, S., Chaudhry, A. A., Wong, F. S., & Rehman, I. U. (2013). Synthesis and characterizations of a fluoride-releasing dental restorative material. *Materials Science and Engineering: C*, 33(6), 3458-3464.

Kheradmandfard, M., & Fathi, M. H. (2010). Preparation and characterization of Mg-doped fluorapatite nanopowders by sol–gel method. *Journal of Alloys and Compounds*, 504(1), 141-145.

Khoroushi, M., Mansoori-Karvandi, T., & Hadi, S. (2012). The effect of pre-warming and delayed irradiation on marginal integrity of a resin-modified glass-ionomer. *General Dentistry*, 60(6), 383-388.

Khoroushi, M., & Keshani, F. (2013). A review of glass-ionomers: From conventional glass-ionomer to bioactive glass-ionomer. *Dental Research Journal*, 10(4), 411-420.

Karimi, M., Jodaei, A., Sadeghinik, A., Ramsheh, M. R., Hafshejani, T. M., Shamsi, M., Orand, F. & Lotfi, F. (2017). Deep eutectic choline chloride-calcium chloride as all-in-one system for sustainable and one-step synthesis of bioactive fluorapatite nanoparticles. *Journal of Fluorine Chemistry*, 204, 76-83.

Kim, D., Abo-Mosallam, H., Lee, H. Y., Lee, J. H., Kim, H. W., & Lee, H. H. (2015). Biological and mechanical properties of an experimental glass-ionomer cement modified by partial replacement of CaO with MgO or ZnO. *Journal of Applied Oral Science*, 23(4), 369-375.

Kiran, P., Ramakrishna, V., Trebbin, M., Udayashankar, N. K., & Shashikala, H. D. (2017). Effective role of CaO/P_2O_5 ratio on SiO_2-CaO-P_2O_5 glass system. *Journal of advanced research*, 8(3), 279-288.

Koo, H. (2008). Strategies to enhance the biological effects of fluoride on dental biofilms. *Advances in Dental Research*, 20(1), 17-21.

Krishnan, V., & Lakshmi, T. (2013). Bioglass: A novel biocompatible innovation. *Journal of Advanced Pharmaceutical Technology & Research*, 4(2), 78-83.

Kurtulus, R., & Kavas, T. (2020). Investigation on the physical properties, shielding parameters, glass formation ability, and cost analysis for waste soda-lime-silica (SLS) glass containing SrO. *Radiation Physics and Chemistry*, 176, 109090-109120.

Kuwik, M., Pisarska, J., & Pisarski, W. A. (2020). Influence of oxide glass modifiers on the structural and spectroscopic properties of phosphate glasses for visible and near-infrared photonic applications. *Materials*, 13(21), 1-20.

Lad, P. P., Kamath, M., Tarale, K., & Kusugal, P. B. (2014). Practical clinical considerations of luting cements: A review. *Journal of International Oral Health*, 6(1), 116-120.

Larkin, P. (2017). *Infrared and Raman spectroscopy: Principles and Spectral Interpretation*. Stamford: Elsevier.

Laskar, I. B., Rajkumari, K., Gupta, R., Chatterjee, S., Paul, B., & Rokhum, L. (2018). Waste snail shell derived heterogeneous catalyst for biodiesel production by the transesterification of soybean oil. *RSC Advances*, 8(36), 20131-20142.

Lindon, J. C., Tranter, G. E., & Koppenaal, D. (2016). *Encyclopedia of spectroscopy and spectrometry*. Netherlands: Academic Press.

Lee, Y. K., Peng, Y. L. & Tomozawa, M. (1997). IR reflection spectroscopy of soda-lime glass surface during ion-exchange. *Journal of Non-Crystalline Solid*, 222, 125-130.

Lee, B. S., Lin, H. P., Chan, J. C. C., Wang, W. C., Hung, P. H., Tsai, Y. H., & Lee, Y. L. (2018). A novel sol-gel-derived calcium silicate cement with short setting time for application in endodontic repair of perforations. *International Journal of Nanomedicine*, 13, 261.

Lemos, J. A., Palmer, S. R., Zeng, L., Wen, Z. T., Kajfasz, J. K., Freires, I. A., Abranches, J. & Brady, L. J. (2019). The biology of Streptococcus mutans. *Gram-Positive Pathogens*, 435-448.

Leśniak, M., Szal, R., Starzyk, B., Gajek, M., Kochanowicz, M., Żmojda, J., Miluski, P., Dorosz, J., Sitarz, M. & Dorosz, D. (2019). Influence of barium oxide on glass-forming ability and glass stability of the tellurite–phosphate oxide glasses. *Journal of Thermal Analysis and Calorimetry*, 138(6), 4295-4302.

Letaïef, N., Lucas-Girot, A., Oudadesse, H., Dorbez-Sridi, R., & Boullay, P. (2014). Investigation of the surfactant type effect on characteristics and bioactivity of new mesoporous bioactive glass in the ternary system SiO_2–CaO–P_2O_5: structural, textural and reactivity studies. *Microporous and Mesoporous Materials*, 195, 102-111.

Li, Z., Khun, N. W., Tang, X. Z., Liu, E., & Khor, K. A. (2017). Mechanical, tribological and biological properties of novel 45S5 Bioglass® composites reinforced with in situ reduced graphene oxide. *Journal of the Mechanical Behavior of Biomedical Materials*, 65, 77-89.

Liao, Y., Brandt, B. W., Li, J., Crielaard, W., Van Loveren, C., & Deng, D. M. (2017). Fluoride resistance in Streptococcus mutans: a mini review. *Journal of Oral Microbiology*, 9(1), 1344509-1344519.

Liu, H., Yazici, H., Ergun, C., Webster, T. J., & Bermek, H. (2008). An in vitro evaluation of the Ca/P ratio for the cytocompatibility of nano-to-micron particulate calcium phosphates for bone regeneration. *Acta biomaterialia*, 4(5), 1472-1479.

Liu, Y. X., Yang, T. O., Yuan, D. X., & Wu, X. Y. (2010). Study of municipal wastewater treatment with oyster shell as biological aerated filter medium. *Desalination*, 254(1-3), 149-153.

Lin, J., Zhu, J., Gu, X., Wen, W., Li, Q., Fischer-Brandies, H., Wang, H. & Mehl, C. (2011). Effects of incorporation of nano-fluorapatite or nano-fluorohydroxyapatite on a resin-modified glass ionomer cement. *Acta Biomaterialia*, 7(3), 1346-1353.

Lin, Y. C., Amesho, K. T., Chen, C. E., Cheng, P. C., & Chou, F. C. (2020). A cleaner process for green biodiesel synthesis from waste cooking oil using recycled waste oyster shells as a sustainable base heterogeneous catalyst under the microwave heating system. *Sustainable Chemistry and Pharmacy*, 17, 100310-100325.

Liska, M., Wilson, A., & Bensted, J. (2019). Special Cements. In P. Hewlett & M. Liska (Eds.), *Lea's chemistry of cement and concrete* (pp. 585-640). Butterworth-Heinemann.

Lohbauer, U. (2010). Dental glass ionomer cements as permanent filling materials?–properties, limitations and future trends. *Materials*, 3(1), 76-96.

Loganathan, S., Valapa, R. B., Mishra, R. K., Pugazhenthi, G., & Thomas, S. (2017). Thermogravimetric analysis for characterization of nanomaterials. In S. Thomas, R. Thomas, A. K. Zachariah, & R. Kumar (Eds.), *Thermal and Rheological Measurement Techniques for Nanomaterials Characterization* (pp. 67-108). Amsterdam: Elsevier.

Loy, C. W., Matori, K. A., Lim, W. F., Schmid, S., Zainuddin, N., Wahab, Z. A., Alassan, Z. N. & Zaid, M. H. M. (2016). Effects of calcination on the crystallography and nonbiogenic aragonite formation of ark clam shell under ambient condition. *Advances in Materials Science and Engineering*, 2016, 1-8.

Loy, C. W., Whitten, A. E., de Campo, L., Appadoo, D., Zainuddin, N., Matori, K. A., Rehm, C., Sokolova, A., Wang, C., Xia, Q. & Whittle, T. A. (2018). Investigation of the siliceous hydrogel phase formation in glass-ionomer cement paste. *Physica B: Condensed Matter*, 551, 287-290.

Lucas, A. G., Mezahi, F. Z., Mami, M., Oudadesse, H., Harabi, A., & Le Floch, M. (2011). Sol–gel synthesis of a new composition of bioactive glass in the quaternary system SiO_2–CaO–Na_2O–P_2O_5: comparison with melting method. *Journal of Non-Crystalline Solids*, 357(18), 3322-3327.

Lusvardi, G., Malavasi, G., Menabue, L., Aina, V., & Morterra, C. (2009). Fluoride-containing bioactive glasses: surface reactivity in simulated body fluids solutions. *Acta Biomaterialia*, 5(9), 3548-3562.

Marghussian, V. K., & Mesgar, A. S. M. (2000). Effects of composition on crystallization behaviour and mechanical properties of bioactive glass-ceramics in the $MgO–CaO–SiO_2–P_2O_5$ system. *Ceramics International*, 26(4), 415-420.

Marie, P. J., & Hott, M. (1986). Short-term effects of fluoride and strontium on bone formation and resorption in the mouse. *Metabolism*, 35(6), 547-551.

Marinoni, N., D'Alessio, D., Diella, V., Pavese, A., & Francescon, F. (2013). Effects of soda–lime–silica waste glass on mullite formation kinetics and micro-structures development in vitreous ceramics. *Journal of Environmental Management*, 124, 100-107.

Marquis, R. E. (1995). Antimicrobial actions of fluoride for oral bacteria. Canadian *Journal of Microbiology*, 41(11), 955-964.

Marques Correia, L., Cecilia, J. A., Rodríguez-Castellón, E., Cavalcante, C. L., & Vieira, R. S. (2017). Relevance of the physicochemical properties of calcined quail eggshell (CaO) as a catalyst for biodiesel production. *Journal of Chemistry*, 2017, 1-12.

Matsuya, S., Maeda, T., & Ohta, M. (1996). IR and NMR analyses of hardening and maturation of glass-ionomer cement. *Journal of Dental Research*, 75(12), 1920-1927.

Matteucci, F., Dondi, M., & Guarini, G. (2002). Effect of soda-lime glass on sintering and technological properties of porcelain stoneware tiles. *Ceramics International*, 28(8), 873-880.

Mayanagi, G., Igarashi, K., Washio, J., Domon-Tawaraya, H., & Takahashi, N. (2014). Effect of fluoride-releasing restorative materials on bacteria-induced pH fall at the bacteria–material interface: An in vitro model study. *Journal of Dentistry*, 42(1), 15-20.

Mclean, J. W. (1994). Proposed nomenclature for glass-ionomer dental cements and related materials. *Quintessence International*, 25, 587-589.

McMillan, P. W. (1964). *Glass-ceramics*. London: Academic Press.

Mickenautsch, S., Mount, G., & Yengopal, V. (2011). Therapeutic effect of glass-ionomers: an overview of evidence. *Australian dental journal*, 56(1), 10-15.

Mitra, S. B. (1991). Adhesion to dentin and physical properties of a light-cured glass-ionomer liner/base. *Journal of Dental Research*, 70(1), 72-74.

Mohamed, M., Yusup, S., & Maitra, S. (2012). Decomposition study of calcium carbonate in cockle shell. *Journal of Engineering Science and Technology*, 7(1), 1-10.

Molla, A. R., Tarafder, A., & Karmakar, B. (2011). Synthesis and properties of glasses in the $K_2O-SiO_2-Bi_2O_3-TiO_2$ system and bismuth titanate ($Bi_4Ti_3O_{12}$) nano glass–ceramics thereof. *Journal of Materials Science*, 46(9), 2967-2976.

Moshaverinia, A., Ansari, S., Moshaverinia, M., Roohpour, N., Darr, J. A., & Rehman, I. (2008). Effects of incorporation of hydroxyapatite and fluoroapatite nanobioceramics into conventional glass ionomer cements (GIC). *Acta Biomaterialia*, 4(2), 432-440.

Moshaverinia, A., Roohpour, N., & Rehman, I. U. (2009). Synthesis and characterization of a novel fast-set proline-derivative-containing glass ionomer cement with enhanced mechanical properties. *Acta Biomaterialia*, 5(1), 498-507.

Moreau, J. L., & Xu, H. H. (2010). Fluoride releasing restorative materials: Effects of pH on mechanical properties and ion release. *Dental Materials*, 26(11), 227-235.

Moreno, E. C., Kresak, M., & Zahradnik, R. T. (1974). Fluoridated hydroxyapatite solubility and caries formation. *Nature*, 247(5435), 64-65.

Mustaffa, R., Mohd Yusof, M. R., & Abdullah, Y. (2015). A novelty of synthetic hydroxyapatite from cockle shell and characterization. *In Advanced Materials Research*, 1087, 429-433.

Mukhopadhyay, S. (2018). Bioactive glass-ceramics. *Fundamental Biomaterials: Ceramics*, 129-152.

Mukherjee, D. P., & Das, S. K. (2013). SiO_2–Al_2O_3–CaO glass-ceramics: effects of CaF_2 on crystallization, microstructure and properties. *Ceramics International*, 39(1), 571-578.

National Solid Waste Management Department (2012). *Solid waste lab Anual Report*. Retrieved from: https://jpspn.kpkt.gov.my/ resources/ index/ user1/ Sumber_Rujukan/ kajian/lab_sisa_pepejal.

Nasiri-Tabrizi, B., Fahami, A., Ebrahimi-Kahrizsangi, R., Khazraei, A., Yazdani, M. R., & Kajbafzadeh, M. J. (2013). A study on mechanochemical behavior of CaO–P_2O_5–CaF_2–ZrO_2 system to produce fluorapatite–zirconia composite nanopowders. *Powder Technology*, 243, 59-70.

Neel, E. A. A., Pickup, D. M., Valappil, S. P., Newport, R. J., & Knowles, J. C. (2009). Bioactive functional materials: a perspective on phosphate-based glasses. *Journal of Materials Chemistry*, 19(6), 690-701.

Nomoto, R., Komoriyama, M., McCabe, J. F., & Hirano, S. (2004). Effect of mixing method on the porosity of encapsulated glass ionomer cement. *Dental Materials*, 20(10), 972-978.

Nourmohammadi, J., Salarian, R., Solati-Hashjin, M., & Moztarzadeh, F. (2007). Dissolution behavior and fluoride release from new glass composition used in glass ionomer cements. *Ceramics International*, 33(4), 557-561.

Nicholson, J. W., Brookman, P. J., Lacy, O. M., Sayers, G. S., & Wilson, A. D. (1988). A study of the nature and formation of zinc polyacrylate cement using Fourier transform infrared spectroscopy. *Journal of Biomedical Materials Research*, 22(7), 623-631.

Nicholson, J. W. (1998). Chemistry of glass-ionomer cements: a review. *Biomaterials*, 19(6), 485-494.

Nicholson, J. W., Brookman, P. J., Lacy, O. M., & Wilson, A. D. (1988). Fourier transform infrared spectroscopic study of the role of tartaric acid in glass-ionomer dental cements. *Journal of Dental Research*, 67(12), 1451-1454.

Nicholson, J. W., & Wilson, A. D. (2000). The effect of storage in aqueous solutions on glass-ionomer and zinc polycarboxylate dental cements. *Journal of Materials Science: Materials in Medicine*, 11(6), 357-360.

Nicholson, J. W. (2016). Adhesion of glass-ionomer cements to teeth: a review. *International Journal of Adhesion and Adhesives*, 69, 33-38.

Njindam, O. R., Njoya, D., Mache, J. R., Mouafon, M., Messan, A., & Njopwouo, D. (2018). Effect of glass powder on the technological properties and microstructure of clay mixture for porcelain stoneware tiles manufacture. *Construction and Building Materials*, 170, 512-519.

Oon N. F. (1986). *Growth and mortality of the malaysian cockle (anadara granosa) under commercial culture analysis through length frequency data*. Fisheries Research Institute Report, Malaysia.

Ozabaci, M., Aksan, M. A., Kirat, G., Kizilaslan, O., & Yakinci, M. E. (2016). Preparation and characterization of $CaO-Al_2O_3-SiO_2$ (CAS) glass-ceramics. *Journal of Non-Crystalline Solids*, 454, 8-12.

Pajor, K., Pajchel, L., & Kolmas, J. (2019). Hydroxyapatite and fluorapatite in conservative dentistry and oral implantology—a review. *Materials*, 12(17), 2683.

Patel, N. R., & Gohil, P. P. (2012). A review on biomaterials: scope, applications & human anatomy significance. *International Journal of Emerging Technology and Advanced Engineering*, 2(4), 91-101.

Pashev, P., Bogdanov, B., & Hristov, Y. (2012). Crystallization kinetics and phase transformation of $SiO_2-Al_2O_3-P_2O_5-CaO-CaF_2$ glass. *Journal of the University of Chemical Technology & Metallurgy*, 47(4), 398-402.

Pearson, G. J., & Atkinson, A. S. (1991). Long-term flexural strength, of glass ionomer cements. *Biomaterials*, 12(7), 658-660.

Peitl, O., Zanotto, E. D., & Hench, L. L. (2001). Highly bioactive P_2O_5–Na_2O–CaO–SiO_2 glass-ceramics. *Journal of Non-Crystalline Solids*, 292(1-3), 115-126.

Penel, G., Leroy, G., Rey, C., Sombret, B., Huvenne, J. P., & Bres, E. (1997). Infrared and Raman micro spectrometry study of fluor-fluor-hydroxy and hydroxy-apatite powders. *Journal of Materials Science: Materials in Medicine*, 8(5), 271-276.

Pires, R., Nunes, T. G., Abrahams, I., Hawkes, G. E., Morais, C. M., & Fernandez, C. (2004). Stray-field imaging and multinuclear magnetic resonance spectroscopy studies on the setting of a commercial glass-ionomer cement. *Journal of Materials Science: Materials in Medicine*, 15(3), 201-208.

Pradiptama, Y., Purwanta, M., & Notopuro, H. (2019). Antibacterial effects of fluoride in streptococcus mutans growth in vitro. *Biomolecular and Health Science Journal*, 2(1), 1-3.

Prosser, H. J., Powis, D. R., & Wilson, A. D. (1986). Glass-ionomer cements of improved flexural strength. *Journal of Dental Research*, 65(2), 146-148.

Rafferty, A., Clifford, A., Hill, R., Wood, D., Samuneva, B., & Dimitrova-Lukacs, M. (2000). Influence of fluorine content in apatite–mullite glass-ceramics. *Journal of the American Ceramic Society*, 83(11), 2833-2838.

Rahman, N. A. A., Matori, K. A., Zaid, M. H. M., Zainuddin, N., Ab Aziz, S., Khiri, M. Z. A., Jalil, R.A & Jusoh, W. N. W. (2019). Fabrication of Alumino-Silicate-Fluoride based bioglass derived from waste clam shell and soda lime silica glasses. *Results in Physics*, 12, 743-747.

Ramay, H. R., & Zhang, M. (2004). Biphasic calcium phosphate nanocomposite porous scaffolds for load-bearing bone tissue engineering. *Biomaterials*, 25(21), 5171-5180.

Ranjkesh, B., Chevallier, J., Salehi, H., Cuisinier, F., Isidor, F., & Løvschall, H. (2016). Apatite precipitation on a novel fast-setting calcium silicate cement containing fluoride. *Acta Biomaterialia Odontologica Scandinavica*, 2(1), 68-78.

Raya, I., Mayasari, E., Yahya, A., Syahrul, M., & Latunra, A. I. (2015). Synthesis and characterizations of calcium hydroxyapatite derived from crabs shells (Portunus pelagicus) and its potency in safeguard against to dental demineralizations. *International Journal of Biomaterials*, 2015, 1-8.

Rey, C. (1998). Calcium phosphates for medical applications. In Z. Amjad (Ed.), *Calcium Phosphates in Biological and Industrial Systems* (pp. 217-251). Boston: Springer.

Rezaie, H. R., Bakhtiari, L., & Öchsner, A. (2015). *Biomaterials and Their Applications*. Berlin: Springer International Publishing.

Rodríguez-Lorenzo, L. M., Hart, J. N., & Gross, K. A. (2003). Structural and chemical analysis of well-crystallized hydroxyfluorapatites. *The Journal of Physical Chemistry B*, 107(33), 8316-8320.

Rošin-Grget, K., & Linčir, I. (2001). Current concept on the anticaries fluoride mechanism of the action. Collegium Antropologicum, 25(2), 703-712.

Rošin-Grget, K., Peroš, K., & Šutej, I. (2013). The cariostatic mechanisms of fluoride. *Acta Medica Academica*, 42(2), 179-188.

Roscoe, H. E., & Schorlemmer, C. (1878). A Treatise on Chemistry: The non-metallic elements. *Analyst*, 2(19), 115-117).

Rölla, G., & Ekstrand, J. (1996). Fluoride in oral fluids and dental plaque. In B .A. Burt (Ed.), *Fluoride in Dentistry* (pp. 215-229). Copenhagen: Munksgaard.

Rujitanapanich, S., Kumpapan, P., & Wanjanoi, P. (2014). Synthesis of hydroxyapatite from oyster shell via precipitation. *Energy Procedia*, 56, 112-117.

Saddeek, Y. B. (2004). Ultrasonic study and physical properties of some borate glasses. *Materials Chemistry and Physics*, 83(2-3), 222-228.

Saji, G. S., Shamnadh, M., Varma, S., Amanulla, S., Bharath, R. S., & Oommen, C. (2018). Experimental evaluation of compressive strength of pmma-seashell based biocomposites for orthopedic applications. *Materials Today: Proceedings*, 5(8), 16509-16515.

Salama, S. N., Salman, S. M., & Darwish, H. (2002). Effect of nucleation catalysts on crystallisation characteristics of aluminosilicate glasses. *Ceramics-Silikáty*, 46(1), 15-23.

Souza, G. P., Rambaldi, E., Tucci, A., Esposito, L., & Lee, W. E. (2004). Microstructural variation in porcelain stoneware as a function of flux system. *Journal of the American Ceramic Society*, 87(10), 1959-1966.

Sehgal, J., & Ito, S. (1998). A new low-brittleness glass in the soda-lime-silica glass family. *Journal of the American Ceramic Society*, 81(9), 2485-2488.

Seppa, L., Forss, H., & Øgaard, B. (1993). The effect of fluoride application on fluoride release and the antibacterial action of glass lonomers. *Journal of Dental Research*, 72(9), 1310-1314.

Shackelford, J. F. (1996). *Introduction to Materials Science for Engineers*, New Jersey: Pearson.

Shafiu Kamba, A., Ismail, M., Tengku Ibrahim, T. A., & Zakaria, Z. A. B. (2013). Synthesis and characterisation of calcium carbonate aragonite nanocrystals from cockle shell powder (Anadara granosa). *Journal of Nanomaterials*, 2013, 1-9.

Sheng, J., Kadono, K., Utagawa, Y., & Yazawa, T. (2002). X-ray irradiation on the soda-lime container glass. *Applied Radiation and Isotopes*, 56(4), 621-626.

Shelby, J. E. (2005). *Introduction to Glass Science and Technology*. Cambridge: The Royal Society of Chemistry.

Shah, F. A. (2016). Fluoride-containing bioactive glasses: glass design, structure, bioactivity, cellular interactions, and recent developments. *Materials Science and Engineering: C*, 58, 1279-1289.

Shih, P. K., & Chang, W. L. (2015). The effect of water purification by oyster shell contact bed. *Ecological Engineering*, 77, 382-390.

Shrivastava, A. (2018). *Introduction to Plastics Engineering: Plastic Properties and Testing*. Oxford: William Andrew.

Skallevold, H. E., Rokaya, D., Khurshid, Z., & Zafar, M. S. (2019). Bioactive glass applications in dentistry. *International Journal of Molecular Sciences*, 20(23), 5960.

Small, H., Stevens, T. S., & Bauman, W. C. (1975). Novel ion exchange chromatographic method using conductimetric detection. *Analytical Chemistry*, 47(11), 1801-1809.

Srinivasan, K. K., Adhikari, A. V., & Satapathy, L. N. (2015). Evaluation of calcium fluoroaluminosilicate based glass ionomer luting cements processed both by conventional and microwave assisted methods. *Technologies*, 3(2), 58-73.

Silva, T. H., Mesquita-Guimarães, J., Henriques, B., Silva, F. S., & Fredel, M. C. (2019). The potential use of oyster shell waste in new value-added by-product. *Resources*, 8(1), 13-28.

Singh, A. (2012). Hydroxyapatite, a biomaterial: its chemical synthesis, characterization and study of biocompatibility prepared from shell of garden snail, Helix aspersa. *Bulletin of Materials Science*, 35(6), 1031-1038.

Singh, A., & Purohit, K. M. (2011). Chemical synthesis, characterization and bioactivity evaluation of hydroxyapatite prepared from garden snail (helix aspersa). *Journal of Biotechnology & Biomaterials*, 1, 104-112.

Sinton, C. W., & LaCourse, W. C. (2001). Experimental survey of the chemical durability of commercial soda-lime-silicate glasses. *Materials Research Bulletin*, 36(13), 2471-2479.

Smith, D. C. (1998). Development of glass-ionomer cement systems. *Biomaterials*, 19(6), 467-478.

Stamboulis, A., Hill, R. G., Law, R. V., & Matsuya, S. (2004). A MAS NMR study of the crystallisation process of apatite-mullite glass-ceramics. *In Key Engineering Materials*, 254, 99-102.

Stan, C. V., Beavers, C. M., Kunz, M., & Tamura, N. (2018). X-ray diffraction under extreme conditions at the Advanced Light Source. *Quantum Beam Science*, 2(1), 4-33.

Stanton, K., & Hill, R. (2000). The role of fluorine in the devitrification of $SiO_2 \cdot Al_2O_3 \cdot P_2O_5 \cdot CaO \cdot CaF_2$ glasses. *Journal of Materials Science*, 35(8), 1911-1916.

Stevenson, P. (2012). *Foam engineering: Fundamentals and applications*. West Sussex: JohnWiley & Son.

Sunder S. (2019 July 1), Glassmaking trends in Malaysia. *Glass Worldwide*, 32-36. Retrieved from https://www.glassworldwide.co.uk/sites/default/ files/afgm-articles/Malaysia.

Suryanarayana, C., & Norton, M. G. (1998). Practical aspects of X-ray diffraction. In C. Suryanarayana & M Grant-Norton (Eds.), *X-Ray Diffraction* (pp. 63-94). Boston: Springer.

Suteu, D., Bilba, D., Aflori, M., Doroftei, F., Lisa, G., Badeanu, M., & Malutan, T. (2012). The seashell wastes as biosorbent for reactive dye removal from textile effluents. *Clean–Soil, Air, Water*, 40(2), 198-205.

Talal, A., Tanner, K. E., Billington, R., & Pearson, G. J. (2009). Effect of ultrasound on the setting characteristics of glass ionomer cements studied by Fourier Transform Infrared Spectroscopy. *Journal of Materials Science: Materials in Medicine*, 20(1), 405-411.

Tanaka, K., Ohta, K., Haddad, P. R., Fritz, J. S., Miyanaga, A., Hu, W., Hasebe, K., Lee, K.P. & Sarzanini, C. (2001). High-performance ion-exclusion/ cation-exchange chromatography of anions and cations in acid rain waters on a weakly acidic cation-exchange resin. *Journal of Chromatography A*, 920(1-2), 239-245.

Taruta, S., Mukoyama, K., Suzuki, S. S., Kitajima, K., & Takusagawa, N. (2001). Crystallization process and some properties of calcium mica–apatite glass-ceramics. *Journal of Non-Crystalline Solids*, 296(3), 201-211.

Tarvornpanich, T., & Souza, G. P. (2007). Use of soda-lime-silica waste glass as an alternative flux in traditional ceramics. *International Conference on Geology of Thailand: Towards Sustainable Development and Sufficiency Economy*, 7(1), 46-53.

Tarvornpanich, T., Souza, G. P., & Lee, W. E. (2005). Microstructural evolution on firing soda–lime–silica glass fluxed whitewares. *Journal of the American Ceramic Society*, 88(5), 1302-1308.

Tilocca, A., & Cormack, A. N. (2007). Structural effects of phosphorus inclusion in bioactive silicate glasses. *The Journal of Physical Chemistry B*, 111(51), 14256-14264.

Tomlinson, S. K., Ghita, O. R., Hooper, R. M., & Evans, K. E. (2007). Investigation of the dual setting mechanism of a novel dental cement using infrared spectroscopy. *Vibrational Spectroscopy*, 45(1), 10-17.

Tung, M. S. (1998). Calcium phosphates: structure, composition, solubility, and stability. In Z. Amjad (Ed.), *Calcium phosphates in biological and industrial systems* (pp. 1-19). Boston: Springer.

Tredwin, C. J., Young, A. M., Georgiou, G., Shin, S. H., Kim, H. W., & Knowles, J. C. (2013). Hydroxyapatite, fluor-hydroxyapatite and fluorapatite produced via the sol–gel method. Optimisation, characterisation and rheology. *Dental Materials*, 29(2), 166-173.

Uddin, M. K. H. (2018). *Introduction And Development of Aluminium Ion Free Silicate Based Bioactive Glass Ionomer Cement Based on Glass Flakes and Setting Modifier (Citrate) For Direct Posterior Dental Restorations.* (Doctoral dissertation). University of Sheffield, United Kindom.

Van Noort, R., & Barbour, M. (2014*). Introduction to Dental Materials-E-Book*. Elsevier Health Sciences.

Vermeersch, G., Leloup, G., Delmee, M., & Vreven, J. (2005). Antibacterial activity of glass–ionomer cements, compomers and resin composites: relationship between acidity and material setting phase. *Journal of Oral Rehabilitation*, 32(5), 368-374.

Wah, L. C. (2012*). Effects Of Heat Treatment on Structure and Thermal Diffusivities of* SiO_2*-*Na_2O*-*Al_2O_3*-Cao-*CaF_2 *Glass-Ceramics from Waste Materials.* (Master dissertation). Universiti Putra Malaysia, Malaysia.

Wang, Y., Lin, S., & Suzuki, Y. (2007). Study of limestone calcination with CO_2 capture: Decomposition behavior in a CO_2 atmosphere. *Energy & Fuels*, 21(6), 3317-3321.

Wang, C., Zhao, J., Zhao, X., Bala, H., & Wang, Z. (2006). Synthesis of nanosized calcium carbonate (aragonite) via a polyacrylamide inducing process. *Powder Technology*, 163(3), 134-138.

Wasson, E. A., & Nicholson, J. W. (1991). Studies on the setting chemistry of glass-ionomer cements. *Clinical Materials*, 7(4), 289-293.

Wasson, E. A., & Nicholson, J. W. (1993). New aspects of the setting of glass-ionomer cements. *Journal of Dental Research*, 72(2), 481-483.

Wei, M., Evans, J. H., Bostrom, T., & Grøndahl, L. (2003). Synthesis and characterization of hydroxyapatite, fluoride-substituted hydroxyapatite and fluorapatite. *Journal of Materials Science: Materials in Medicine*, 14(4), 311-320.

Wilson A. D., McLean J.W. (1988). *Glass-ionomer Cement*. Chicago: Quintessence Publishing.

Wilson, A. D., & Kent, B. E. (1971). The glass-ionomer cement, a new translucent dental filling material. *Journal of Applied Chemistry and Biotechnology*, 21(11), 313-313.

Wilson A.D. and McLean J., (1988). *Glass-Ionomer Cement*. Chicago: Quintessence Publishing.

Wilson, A. D. (1978). The chemistry of dental cements. *Chemical Society Reviews*, 7(2), 265-296.

Wilson, A. D., & McLean, J. W. (1988). *Scientific and Clinical Development in Glass Ionomer Cement*. Chicago: Quintessence Publishing.

Wilson, A. D., Hill, R. G., Warrens, C. P., & Lewis, B. G. (1989). The influence of polyacid molecular weight on some properties of glass-ionomer cements. *Journal of Dental Research*, 68(2), 89-94.

Wilson, J., Clark, A. E., Douek, E., Krieger, J., Smith, W. K., & Zamet, J. S. (1994). Clinical applications of bioglass implants. In Ö. H. Andersson, R. P. Happonen & A. Yli-Urpo (Eds.), *Bioceramics* (pp. 415-422). Turku: Elsevier.

Wilson, A. D., & Prosser, H. J. (1982). Biocompatibility of the glass ionomer cement. *The Journal of the Dental Association of South Africa*, 37(12), 872-879.

Williams, D. F. (1987). Advanced applications for materials implanted within the human body. *Materials Science and Technology*, 3(10), 797-806.

Williams, J. A., Briggs, E., Billington, R. W., & Pearson, G. J. (2003). The effects of adding fluoride compounds to a fluoride-free glass ionomer cement on subsequent fluoride and sodium release. *Biomaterials*, 24(7), 1301-1308.

Wiegand, A., Buchalla, W., & Attin, T. (2007). Review on fluoride-releasing restorative materials—fluoride release and uptake characteristics, antibacterial activity and influence on caries formation. *Dental Materials*, 23(3), 343-362.

Weiss, J., Reinhard, S., Pohl, C., Saini, C., & Narayaran, L. (1995). Stationary phase for the determination of fluoride and other inorganic anions. *Journal of Chromatography A*, 706(1-2), 81-92.

Wen, G., Zheng, X., & Song, L. (2007). Effects of P_2O_5 and sintering temperature on microstructure and mechanical properties of lithium disilicate glass-ceramics. *Acta Materialia*, 55(10), 3583-3591.

Wu, W., Xie, D., Puckett, A., & Mays, J. W. (2003). Synthesis of amino acid-containing polyacids and their application in self-cured glass-ionomer cement. *European Polymer Journal*, 39(5), 959-968.

Wu, S. C., Hsu, H. C., Hsu, S. K., Tseng, C. P., & Ho, W. F. (2017). Preparation and characterization of hydroxyapatite synthesized from oyster shell powders. *Advanced Powder Technology*, 28(4), 1154-1158.

Xie, D., Culbertson, B. M., & Kao, E. C. (1993). New glass-ionomer polymers containing N-acryloyl-6-aminocaproic acid. *Journal of Dental Research*, 72(385), l993.

Xiang, Q., Liu, Y., Sheng, X., & Dan, X. (2007). Preparation of mica-based glass-ceramics with needle-like fluorapatite. *Dental Materials*, 23(2), 251-258.

Xiong, X., Chen, H., & Zhu, R. (2014). Oyster shell waste supported CuCl2 for aldehyde-alkyne-amine coupling reaction to propargylamines. *Chinese Journal of Catalysis*, 35(12), 2006-2013.

Yamakami, S. A., Ubaldini, A. L. M., Sato, F., Medina Neto, A., Pascotto, R. C., & Baesso, M. L. (2018). Study of the chemical interaction between a high-viscosity glass ionomer cement and dentin. *Journal of Applied Oral Science*, 26.

Yamamuro T. (2004). Bioceramics. In D. G. Poitout (Ed.), *Biomechanics and Biomaterials in Orthopedics* (pp. 21-33). London: Springer.

Yli-Urpo, H., Lassila, L. V., Närhi, T., & Vallittu, P. K. (2005). Compressive strength and surface characterization of glass ionomer cements modified by particles of bioactive glass. *Dental Materials*, 21(3), 201-209.

Yvon, H. J. (2017). Raman spectroscopy for analysis and monitoring. *Raman Data and Analysis*, 1, 2.

Yadav, A. K., & Singh, P. (2015). A review of the structures of oxide glasses by Raman spectroscopy. *RSC Advances*, 5(83), 67583-67609.

Youness, R. A., Taha, M. A., & Ibrahim, M. (2018). In vitro bioactivity, physical and mechanical properties of carbonated-fluoroapatite during mechanochemical synthesis. *Ceramics International*, 44(17), 21323-21329.

Young, A. M., Sherpa, A., Pearson, G., Schottlander, B., & Waters, D. N. (2000). Use of Raman spectroscopy in the characterisation of the acid–base reaction in glass-ionomer cements. *Biomaterials*, 21(19), 1971-1979.

Yu, H., Zheng, M., Chen, R., & Cheng, H. (2014). Proper selection of contemporary dental cements. *Oral Health Dental Management*, 13(1), 54-59.

Zahra, V. N., Kohen, S. G., & Macchi, R. L. (2011). Powder-liquid ratio and properties of two restorative glass ionomer cements. *Acta Odontológica Latinoamericana*, 24(2), 200-204.

Zaid, M. H. M., Matori, K. A., Wah, L. C., Sidek, H. A. A., Halimah, M. K., Wahab, Z. A., & Azmi, B. Z. (2011). Elastic moduli prediction and correlation in soda lime silicate glasses containing ZnO. *International Journal of Physical Sciences*, 6(6), 1404-1410.

Zaid, M. H. M., Matori, K. A., Sidek, H. A. A., Halimah, M. K., Wahab, Z. A., & Samsudin, N. F. (2016). Fabrication and crystallization of ZnO-SLS glass derived willemite glass-ceramics as a potential material for optics applications. *Journal of Spectroscopy*, 2016, 1-7.

Zaid, M. H. M., Matori, K. A., Ab, A. S. H., Wahab, Z. A., & Rashid, S. S. A. (2017). Effect of sintering on crystallization and structural properties of soda lime silica glass. *Science of Sintering*, 49(4), 409-417.

Zarifah, N. A., Lim, W. F., Matori, K. A., Sidek, H. A. A., Wahab, Z. A., Zainuddin, N., Fadilah, B. N., Fauzana, A. N. & Salleh, M. A. (2015). An elucidating study on physical and structural properties of 45S5 glass at different sintering temperatures. *Journal of Non-Crystalline Solids*, 412, 24-29.

Zagrajczuk, B., Dziadek, M., Olejniczak, Z., Sulikowski, B., Cholewa-Kowalska, K., & Laczka, M. (2018). Structural investigation of gel-derived materials from the $SiO_2Al_2O_3$ system. *Journal of Molecular Structure*, 1167, 23-32.

Zeng, H., Wang, L., Ye, F., Yang, B., Chen, J., Chen, G., & Sun, L. (2016). Mechanical–structural investigation of chemical strengthening aluminosilicate glass through introducing phosphorus pentoxide. *Frontiers in Materials*, 53(3), 1-7.

Zhou, Y. Y., Li, X. X., & Chen, Z. X. (2012). Rapid synthesis of well-ordered mesoporous silica from sodium silicate. *Powder Technology*, 226, 239-245.

Zhao, J., Dong, X., Bian, M., Zhao, J., Zhang, Y., Sun, Y., Chen, J. & Wang, X. (2014). Solution combustion method for synthesis of nanostructured hydroxyapatite, fluorapatite and chlorapatite. *Applied Surface Science*, 314, 1026-1033.

Zhang, X., & Vecchio, K. S. (2006). Creation of dense hydroxyapatite (synthetic bone) by hydrothermal conversion of seashells. *Materials Science and Engineering: C*, 26(8), 1445-1450.

Zhang, S., Stamboulis, A., & Ni, W. (2019). The effect of boron substitution for aluminium on the microstructure of calcium fluoro-aluminosilicate glasses and glass-ceramics. *Journal of the European Ceramic Society*, 39(5), 1918-1924.

Zoergiebel, J., & Ilie, N. (2013). Evaluation of a conventional glass ionomer cement with new zinc formulation: effect of coating, aging and storage agents. *Clinical Oral Investigations*, 17(2), 619-626.

BIODATA OF STUDENT

Mohammad Zulhasif bin Ahmad Khiri was born in Pontian, Johor, Malaysia in 30th March 1991. He received his primary education at Sekolah Tengku Mahmood Iskandar (1), Pontian, Johor (1998-2002) and then, continued his secondary education school at Sekolah Menengah Kebangsaan Dato' Ali Haji Ahmad, Pontian, Johor. In 2009, he furthered his study in Sciences in Physics at Johor Matriculation College (2009-2010) and received his first degree in Bachelor of Science (Honours) in Major Physics in 2014 from Universiti Putra Malaysia, Serdang,Selangor. Then, in 2015, he continued his study in Advanced Materials field under supervision of Assoc. Prof. Dr. Khamirul Amin Matori at Institute of Advanced Technology (ITMA), Universiti Putra Malaysia, Serdang, Selangor and then, graduated his Master of Sciences degree in years of 2017.

LIST OF PUBLICATIONS

Khiri, M. Z. A., Matori, K. A., Zainuddin, N., Abdullah, C. A. C., Alassan, Z. N., Baharuddin, N. F., & Zaid, M. H. M. (2016). The usability of ark clam shell (Anadara granosa) as calcium precursor to produce hydroxyapatite nanoparticle via wet chemical precipitate method in various sintering temperature. *SpringerPlus*, 5(1), 1-15.

Khiri, M. Z. A., Matori, K. A., Zaid, M. H. M., Abdullah, C. A. C., Zainuddin, N., Alibe, I. M., & Effendy, N. (2019). Crystallization behavior of low-cost biphasic hydroxyapatite/β-tricalcium phosphate ceramic at high sintering temperatures derived from high potential calcium waste sources. *Results in Physics*, 12, 638-644.

Khiri, M. Z. A., Matori, K. A., Zaid, M. H. M., Abdullah, C. A. C., Zainuddin, N., Alibe, I. M., Rahman, N. A.A., & Wahab, S. A. A. (2019). The effect of the Ph values and sintering temperatures on the physical, structural and mechanical properties of nano hydroxyapatite derived from ark clam shells (Anadara granosa) prepared via the wet chemical precipitate method. *Ceramics–Silikáty*, 63(2), 194-203.

Khiri, M. Z. A., Matori, K. A., Zaid, M. H. M., Abdullah, A. C., Zainuddin, N., Jusoh, W. N. W., Rahman, N.A.A., & Effendy, N. (2020). Soda lime silicate glass and clam Shell act as precursor in synthesize calcium fluoroaluminosilicate glass to fabricate glass ionomer cement with different ageing time. *Journal of Materials Research and Technology*, 9(3), 6125-6134

Jalil, R. A., Matori, K. A., Zaid, M. H. M., Zainuddin, N., Khiri, M. Z. A., Rahman, N. A. A., Jusoh, W. N. W., & Kul, E. (2020). A Study of Fluoride-Containing Bioglass System for Dental Materials Derived from Clam Shell and Soda Lime Silica Glass. *Journal of Spectroscopy*, 2020, 1-9.

Jusoh, W. N. W., Matori, K. A., Zaid, M. H. M., Zainuddin, N., Khiri, M. Z. A., Rahman, N. A. A., Jalil, R. A., & Kul, E. (2020). Influence of different CaF_2 contents and heat treatment temperatures on apatite-mullite glass ceramics derived from waste materials. *Ceramics–Silikáty*, 64(4), 447-459.

Rahman, N. A. A., Matori, K. A., Zaid, M. H. M., Zainuddin, N., Ab Aziz, S., Khiri, M. Z. A., Jalil, R. A., & Jusoh, W. N. W. (2019). Fabrication of Alumino-Silicate-Fluoride based bioglass derived from waste clam shell and soda lime silica glasses. *Results in Physics*, 12, 743-747.

Jusoh, W. N. W., Matori, K. A., Zaid, M. H. M., Zainuddin, N., Khiri, M. Z. A., Rahman, N. A. A., Jalil, R. A., & Kul, E. (2019). Effect of sintering temperature on physical and structural properties of Alumino-Silicate-Fluoride glass ceramics fabricated from clam shell and soda lime silicate glass. *Results in Physics*, 12, 1909-1914.

UNIVERSITI PUTRA MALAYSIA

STATUS CONFIRMATION FOR THESIS / PROJECT REPORT AND COPYRIGHT

ACADEMIC SESSION : ______________________

TITLE OF THESIS / PROJECT REPORT :

EFFECT OF WATER RATIO AND AGING TIME ON GLASS IONOMER CEMENT DERIVED FROM CALCIUM FLUOROALUMINOSILICATE-BASED GLASS

NAME OF STUDENT: MOHAMMAD ZULHASIF AHMAD KHIRI

I acknowledge that the copyright and other intellectual property in the thesis/project report belonged to Universiti Putra Malaysia and I agree to allow this thesis/project report to be placed at the library under the following terms:

1. This thesis/project report is the property of Universiti Putra Malaysia.
2. The library of Universiti Putra Malaysia has the right to make copies for educational purposes only.
3. The library of Universiti Putra Malaysia is allowed to make copies of this thesis for academic exchange.

I declare that this thesis is classified as :

*Please tick (√)

☐	**CONFIDENTIAL**	(Contain confidential information under Official Secret Act 1972).
☐	**RESTRICTED**	(Contains restricted information as specified by the organization/institution where research was done).
☐	**OPEN ACCESS**	I agree that my thesis/project report to be published as hard copy or online open access.

This thesis is submitted for :

☐	**PATENT**	Embargo from ____________ (date) until ____________ (date)

Approved by:

(Signature of Student) New IC No/ Passport No.:	(Signature of Chairman of Supervisory Committee) Name:
Date :	Date :

[Note : If the thesis is CONFIDENTIAL or RESTRICTED, please attach with the letter from the organization/institution with period and reasons for confidentially or restricted.]