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SYNTHESIS AND CHARACTERISATION OF GLASS IONOMER CEMENT FROM SODA LIME SILICA WASTE GLASS OR ANADARA GRANOSA L. SHELL

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By

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Chairman : Norhazlin Zainuddin, PhD

Faculty : Science

Glass ionomer cements (GIC) are produced via acid-base reaction between calcium-fluoroaluminosilicate glass powders and freeze-dried polyacrylic acid (PAA) powder. Soda lime silica (SLS) waste glass, mainly consist of silica dioxide (SiO_2) and *Anadara granosa* (cockle shells), mainly consist of calcium carbonate ($CaCO_3$), have been utilised in this study as the prime source of SiO_2 and $CaCO_3$ by incorporating it as part of glass components for production of GIC. Therefore, the main objectives of this research were to study the potential of SLS waste glass and *Anadara granosa* shells as raw materials for synthesis of GIC and investigate the effect of the waste materials towards setting reaction and compressive strength of GIC. GIC were produced by introducing distilled water into the pre-mixed glass and PAA powders in a powder: liquid weight ratio of 4:1. Energy dispersive X-ray spectrometry (EDX), Thermogravimetric Analysis (TGA), X-Ray Diffraction (XRD), Solid State Nuclear Magnetic Resonance (SS NMR), Fourier-Transform Infrared spectroscopy (FTIR), and compressive strength evaluation were employed for characterisations of waste materials, glasses and GIC. Result from EDX showed that SLS waste glass contained more than 70% of SiO_2 and result from TGA and EDX showed that the percentage of $CaCO_3$ in *Anadara granosa* was more than 98 %. This suggested that both SLS waste glass and *Anadara granosa* shell were ideal natural sources of SiO_2 and $CaCO_3$ and suitable to be utilised in GIC production. Both FTIR and XRD analyses implied that calcite was the only polymorph found in *Anadara granosa* shell. The absence of sharp diffraction peaks in XRD pattern suggested that all glass types produced namely GWX 1 (using high purity raw material), GWX 2 (using SLS waste glass as source of SiO_2), and GWX 3 (using *Anadara granosa* as source of $CaCO_3$), were completely amorphous. From SS NMR results, the chemical shift of ^{29}Si suggested that all glasses have Q^3 (4Al) structure. When Na content is higher, the chemical shift of ^{31}P chemical shift shifted to a more positive value. The chemical shift of ^{27}Al shifted to a more positive value when Al:P ratio has increase. FTIR result showed that setting reaction of all glass types involved the occurrence of cross-linking between polyacrylate chain and metal ions from glass given by gradual conversion of COOH absorption band at 1690-1700 cm^{-1} into COO^-M^{n+} absorption

band at 1550-1600 cm^{-1} and hardening of GICs given by gradual conversion of Si-O-H absorption band at 950-969 cm^{-1} into Si-O-Si absorption band at 1040-1056 cm^{-1}. GIC 2 cements exhibited slower setting rate at initial stage but completely established cross-linking at 1 day ageing whereas GIC 3 cements showed cross-linking completely at 1 hour ageing time which were similar as GIC 1 cement. All cements maintained their compressive strengths throughout the ageing time. At 1 day ageing time, GIC 2 and GIC 3 cements exhibited lower compressive strength of 59.15 MPa and 62.70 MPa, respectively in relative to 69.39 MPa for GIC 1 cements. However, GIC 2 and GIC 3 cements showed increment in compressive strength and achieved slightly higher values than GIC 1 after 7 days and 28 days of ageing time. Sodium content in both GIC 2 and GIC 3 was found to be dominant factor of slow setting rate and low compressive strength at initial stage. Based on analyses conducted in this study, both SLS waste glass and *Anadara granosa* can be potentially utilised for glass synthesis in GIC production as GIC 2 and GIC 3 cement exhibited similar setting reaction and compressive strength as GIC 1 cement.

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

SINTESIS DAN PENCIRIAN SIMEN KACA IONOMER DARIPADA KACA BUANGAN KAPUR SODA SILIKA ATAU CENGKERANG *ANADARA GRANOSA* L.

Oleh

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Simen kaca ionomer (GIC) dihasilkan melalui tindak balas asid-bes antara serbuk kaca kalsium-fluoroaluminosilikat dan serbuk beku-kering poliakrilik asid (PAA). Kaca buangan kapur soda silika (SLS) yang sebahagian besarnya mengandungi silika dioksida (SiO_2) dan cengkerang *Anadara granosa* (cengkerang kerang) yang sebahagian besarnya mengandungi kalsium karbonat ($CaCO_3$), telah digunakan dalam kajian ini sebagai sumber utama silika dioksida (SiO_2) dan kalsium karbonat ($CaCO_3$) sebagai sebahagian daripada bahan komponen untuk penghasilan GIC. Oleh itu, objektif utama projek ini adalah untuk mengkaji potensi kaca buangan SLS dan cengkerang *Anadara granosa* sebagai bahan mentah bagi sintesis GIC dan mengkaji kesan bahan buangan tersebut terhadap tindak balas mengeras dan juga kekuatan mampatan GIC. GIC telah dihasilkan dengan mencampurkan air suling ke dalam pra-campuran kaca dan serbuk PAA dalam nisbah berat serbuk: cecair 4:1. Spektroskopi Serakan Tenaga Sinar X (EDX), Analisis Termogravimetrik (TGA), Pembelauan Sinar-X (XRD), Spektroskopi Resonans Magnet Nukleus Keadaan Pepejal (SS NMR), Spektroskopi Inframerah Pengubah Fourier (FTIR), dan penilaian kekuatan mampatan telah digunakan bagi pencirian bahan buangan, kaca dan GIC. Keputusan daripada EDX menunjukkan SLS mengandungi lebih daripada 70% SiO_2 dan keputusan daripada TGA dan EDX menunjukkan bahawa peratusan $CaCO_3$ dalam cengkerang *Anadara granosa* adalah melebihi 98%. Ini mencadangkan bahawa kedua-dua kaca buangan SLS dan cengkerang *Anadara granosa* adalah sumber semula jadi ideal bagi SiO_2 dan $CaCO_3$ dan sesuai digunakan untuk penghasilan GIC. Kedua-dua analisis FTIR dan XRD menunjukkan kalsit merupakan satu-satunya polimof yang terdapat dalam cengkerang *Anadara granosa*. Ketiadaan puncak pembelauan yang tajam dalam XRD mencadangkan bahawa semua jenis kaca yang dihasilkan bernama GWX 1 (menggunakan sumber bahan yang tinggi ketulenannya), GWX 2 (mengunakan kaca buangan SLS sebagai sumber SiO_2) dan GWX 3 (menggunakan cengkerang *Anadara granosa* sebagai sumber $CaCO_3$), adalah amorfus sepenuhnya. Daripada keputusan SS NMR, anjakan kimia ^{29}Si mencadangkan bahawa semua kaca mempunyai struktur Q^3 (4Al). Apabila kandungan Na lebih tinggi, anjakan kimia ^{31}P telah menganjak ke nilai yang lebih

positif. Anjakan kimia ^{27}Al telah menganjak ke nilai yang lebih positif apabila nisbah Al:P telah bertambah. Keputusan FTIR menunjukkan tindak balas mengeras bagi semua jenis kaca melibatkan berlakunya tindak balas silang antara rangkaian poliakrilat dan ion-ion logam dari kaca berdasarkan penukaran jalur penyerapan COOH pada 1690-1700 cm^{-1} kepada jalur penyerapan $COO^{-}M^{n+}$ pada 1550-1600 cm^{-1} dan pengerasan GIC berdasarkan penukaran jalur penyerapan Si-OH pada 950-969 cm^{-1} kepada jalur penyerapan Si-O-Si pada 1040-1056 cm^{-1}. Simen GIC 2 mempamerkan kadar pengerasan yang lebih perlahan pada peringkat awal tetapi menunjukkan pembentukan tindak balas silang sepenuhnya pada penuaan 1 hari manakala simen GIC 3 menunjukkan tindak balas silang sepenuhnya pada penuaan 1 jam sama seperti simen GIC 1. Semua simen mengekalkan kekuatan mampatan sepanjang tempoh penuaan. Pada penuaan 1 hari, simen GIC 2 dan GIC 3 menunjukkan kekuatan mampatan yang lebih rendah 59.15 MPa dan 62.70 MPa, masing-masing berbanding dengan 69.39 MPa bagi GIC 1. Walau bagaimanapun, simen GIC 2 dan GIC 3 menunjukkan peningkatan kekuatan mampatan dan telah mencapai kekuatan mampatan yang lebih sedikit berbanding dengan simen GIC 1 selepas 7 hari dan 28 hari penuaan. Kandungan natrium dalam GIC 2 dan GIC 3 merupakan faktor utama bagi kadar mengeras yang perlahan dan kekuatan mampatan yang lebih rendah pada peringkat awal. Berdasarkan beberapa analisis yang dijalankan dalam kajian ini, kaca buangan SLS dan cengkerang *Anadara granosa* adalah berpotensi digunakan bagi sintesis kaca dalam penghasilan GIC sebab simen GIC 2 and GIC 3 mempamerkan tindak balas pengerasan dan kekuatan mampatan yang serupa dengan simen GIC 1.

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FRANCIS THOO VOON WAI

2014

I certify that a Thesis Examination Committee has met on 12th December 2014 to conduct the final examination of Francis Thoo Voon Wai on his thesis entitled Synthesis and Characterisation of Glass Ionomer Cement from Soda Lime Silica Waste Glass And *Anadara Granosa* L. Shell in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the degree of Master of Science.

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LIST OF ABBREVIATIONS

GIC	Glass Ionomer Cement
SLS	Soda Lime Silica Glass
PAA	Poly(acrylic acid)
FTIR	Fourier Transform Infrared Spectroscopy
ISO	International Organisation For Standarisation
DCL	Degree of Cross-Linking
ASPA	Aluminosilicate Polyacrylic Acid
SS NMR	Solid State Nuclear Magnetic Resonance
XRD	X-Ray Diffraction
t_f	Firing time
ppm	Part per million

CHAPTER 1

INTRODUCTION

1.1 Background of glass ionomer cement

Oral health is important in giving us a quality life. Oral health problems would affect speech and the ability to consume food. Besides, unhealthy oral condition affects our diet, social interaction, psychological status, appearance and many more (John *et al.*, 2004; Qureshi *et al.*, 2014; Steele *et al.*, 2004). Thus, researches in improving dental restorative materials are crucial.

So far, people commonly use gold and ceramics as dental restorative materials. Until late seventies, amalgam became popular dental restorative material (Friberg *et al.*, 1995). Anyway, the use of amalgam in dental restorative materials has been widely discussed due to its mercury release and toxicity to human body (Roulet, 1997). Due to this reason, a variety of alternatives to amalgam has been studied and marketed.

Amalgam has a long clinical history as a reliable, easily handle and inexpensive dental restorative material. However, the controversy of the mercury and poor aesthetics are the disadvantages of amalgam. Other than that, amalgam also has disadvantages such as aesthetically undesirable, exhibits allergic and toxic potential upon mercury release (Lohbauer, 2009; Nicholson & Anstice, 1999) and requires undercut cavity preparation due to its non-adhesive property (Jones, 1995). Other common dental filling materials are resin composites, dental casting alloys, and glass ionomer cement. Dental casting alloys give excellent physical properties but the production is expensive and some composition in the alloys will cause allergic to certain group of people (Davidson & Mjör, 1999). Besides of these disadvantages, both amalgam and dental casting alloys do not have same colour as tooth colour and the demand is increasing for a better appearance of dental materials. Resin composites, which have similar colour as tooth give good physical properties but allergic problems have arisen. Concern on the estrogenic effects of bisphenol A as an environmental hormone has been pointed out (Fleisch *et al.*, 2010; Olea *et al.*, 1996; Qureshi *et al.*, 2014).

In the early of 70s, scientists have invented glass ionomer cements (GIC), which is made from silicate with modified SiO_2/Al_2O_3 ratio to improve the appearance of the cement (Kent *et al.*, 1973). Multiple advantages of GIC hence become popular and commonly accepted by the dental community. The ability to modify physical properties by manipulating powder to liquid ratio, exhibit anticariogenic potential with fluorine uptake and release by enamel and dentin, good biocompatibility, good chemical adhesion to hydroxyapatite phase in dentin, enamel, bone and mineralised tissue, low toxicity and tooth colour replication are among the advantages (Davidson & Mjör, 1999). Other than this, wide range of clinical applications such as GIC as liner, base, luting cement, sealant, surface restoratives, anterior and posterior fillings indicate the versatility of GIC. The applicability of GIC in biomedical application have been studied, for instance, substitutes for bone replacement (Brook & Hatton, 1998), skull defect sealing (Geyer *et al.*, 1998), and bone cements (Hatton *et al.*, 2006).

In this study, water settable GIC was employed to improve handling properties of GIC. Water settable GIC is a form of the dental cement in, which freeze-dried PAA is added to glass powder, which later activated by distilled water. In general, setting reaction of GIC involves the neutralisation of polymeric acid (in this case PAA) by basic glass to form metal polyacrylate as showed in Figure 1.1 (Lohbauer, 2009).

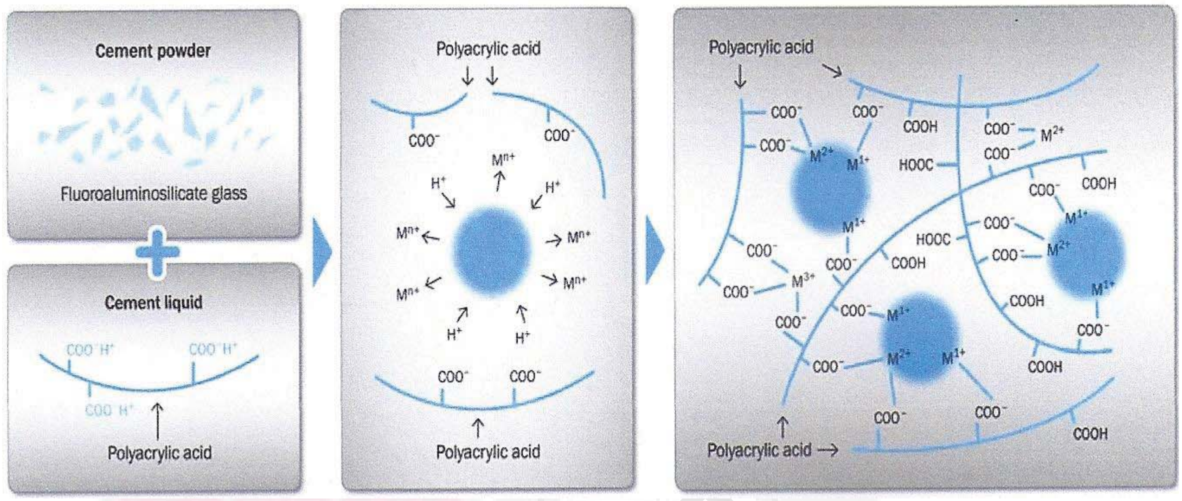

Figure 1.1 : Schematic diagram of setting reaction in GIC (Lohbauer, 2009).

Every year industry and household produce a huge number of waste glass, for instance those glass containers for food and beverage as well as the commodity items. One of the biggest potions of waste glasses is soda lime silica (SLS) waste glass. SLS is suitable to substitute SiO_2 in the glass composition due to its compositions mainly SiO_2 (73.9 wt%) and CaO (11.2 wt%) (Jong, 1989). According to reports, SLS waste glass has been applied in other field of studies and it gave good physical properties, which include mechanical strength. For example in 2004, Tucci et al have reported the use of SLS waste glass in stoneware tile to increase the compressive strength (Tucci *et al.*, 2004). Other than that, SLS waste glass has also been used in producing vitreous ceramics according to a finding from previous studies (Marinoni *et al.*, 2013).

Another raw material used in this research was *Anadara granosa* shell, which commonly known as blood cockle, is a bivalve mollusc, which is found in a large quantity in muddy area of coastal region of South East Asian countries, particularly in Malaysia, Indonesia and Thailand (Sahari & Mijan, 2011). Generally, Malaysia is the main producer for blood cockles. In 2007, 260 farmers have been working on 6250.09 hectares of area for blood cockle cultivation, this number has increased to 10740.20 hectares work 1004 farmer in year 2012 (Izura & Hooi, 2008). In addition, in year 2011 Malaysia had produced 75568.91 metric tonnes of blood cockles. Cockle shells is a good candidate to substitute $CaCO_3$ in the glass composition because previous study found that 98.7% of the content is $CaCO_3$ (Awang *et al.*, 2007).

1.2 Problem statements

Among all the dental filling materials, GIC have attracted much attention and been widely used due to their excellent features compared to some other conventional dental filling materials like amalgam. Furthermore, GIC is widely use because of its low toxicity, tooth colour replication, biocompability and so much more. The possibility to control the physical properties of dental material also an added advantage.

Various materials can be used in making GIC, however using waste as the raw material in producing GIC is still limited and little information is available. Malaysia produces tonnes of waste of *Anadara granosa* shell (blood cockle) and SLS waste glass which can be used in producing the GIC using these two wastes. *Anadara granosa* shell and SLS waste glass are rich in $CaCO_3$ and SiO_2 respectively, and both are main composition in producing the glass for GIC formation. Hence study on the potential of these two waste in producing dental filling materials were carried out in this research project and the results of the study are presented in this thesis.

1.3 Significance of the study

Dental caries is one of the worldwide chronic diseases in, which individuals are susceptible to it throughout their lifetime. It is the primary cause or oral pain and tooth loss and it can progress until the tooth is destroyed. Tooth restoration has been applied over years as a treatment for tooth decay. In fact, a person's appearance, speech and eating habits may be affected without having tooth restoration over dental caries.

As mentioned earlier, GIC is one of the best dental restoratives which provide anticariogenic effect, good adhesion with dentin and enamel, biocompatibility and so forth. In addition, GIC is widely applicable as various forms of restorations in dentistry field and also in other fields. Nowadays, more researchers begin to investigate for SLS and blood cockle shells applicability. The two waste materials used in this study have now gradually become value-added material in various fields. In spite of disposing SLS waste glass and blood cockle shells in landfills, the potential of both of them can be exploited through waste recovery and utilization of recovered material and become raw materials. As a result, the pollution caused by the accumulation of the wastes especially SLS waste glass can be reduced.

From previous studies, SLS waste glass and cockle shells contain high content of SiO_2 and $CaCO_3$, respectively. They are both suitable to substitute common natural SiO_2 source from quartz sand and calcium carbonate source from limestone.

The use of waste gasifier slag in producing glass ionomer cements has been used in previous study. Four gasifier slags used in this study were Drayton, British Gas, Newlands, and E1 Cerrejon. All of the gasifier slags were successfully mixed and made into GIC except for Newlands, which is too reactive to be mixed into GIC. The finding of this research is significant as GIC, which is hydrolytic were successfully formed from the waste and the advantage using these wastes is the Si: Al ratio in them gave a better compressive strength. (Sullivan *et al.*, 2000)

In this study, the raw materials for glass making were substituted to waste materials, which were SLS and clamshell. The glass composition used for the synthesis of glass phase comprised of 26% SiO_2, 19.5% Al_2O_3, 20% P_2O_5, 19.5% $CaCO_3$, and 15% CaF_2 in weight percentage. There were three glass types being synthesised and studied. GWX 1, GWX 2 and GWX 3, which differed by their source of SiO_2 (GWX

2 from SLS waste glasses), $CaCO_3$ (GWX 3 from waste *Anadara granosa* shell) respectively, while GWX 1 from analytical grade reagents.

1.4 Objectives of the study

The objectives of this study were:

1. To synthesise and characterise GIC from SLS waste glass and *Anadara granosa* shells.
2. To investigate the effects of SLS waste glass and *Anadara granosa* shells utilization towards setting reaction and compressive strength of GIC.

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