



UNIVERSITI TEKNIKAL MALAYSIA MELAKA

**THE EFFECT OF COLOURING AGENT ON THE CORROSION
PROPERTIES OF GLASS COMPOSITE TILES**

This report submitted in accordance with requirement of the Universiti Teknikal
Malaysia Melaka (UTeM) for the Bachelor Degree of Manufacturing Engineering
(Engineering Materials) (Hons.)

by

LIYANA BINTI MUHAMAD ALI

B050910157

910902-11-5134

FACULTY OF MANUFACTURING ENGINEERING

2013



UNIVERSITI TEKNIKAL MALAYSIA MELAKA

BORANG PENGESAHAN STATUS LAPORAN PROJEK SARJANA MUDA

TAJUK: The effect of colouring agent on the corrosion properties of glass composite tile

SESI PENGAJIAN: 2012/2013 Semester 2

Saya **LIYANA BINTI MUHAMAD ALI**

mengaku membenarkan Laporan PSM ini disimpan di Perpustakaan Universiti Teknikal Malaysia Melaka (UTeM) dengan syarat-syarat kegunaan seperti berikut:

1. Laporan PSM adalah hak milik Universiti Teknikal Malaysia Melaka dan penulis.
2. Perpustakaan Universiti Teknikal Malaysia Melaka dibenarkan membuat salinan untuk tujuan pengajian sahaja dengan izin penulis.
3. Perpustakaan dibenarkan membuat salinan laporan PSM ini sebagai bahan pertukaran antara institusi pengajian tinggi.
4. **Sila tandakan (√)

SULIT

(Mengandungi maklumat yang berdarjah keselamatan atau kepentingan Malaysiasebagaimana yang termaktub dalam AKTA RAHSIA RASMI 1972)

TERHAD

(Mengandungi maklumat TERHAD yang telah ditentukan oleh organisasi/badan di mana penyelidikan dijalankan)

TIDAK TERHAD

Disahkan oleh:

Alamat Tetap:

1821, Jalan Sultan Mahmud,

Kuala Ibai, 20400

Kuala Terengganu, Terengganu

Cop Rasmi:

DR. JARIAH BINTI MOHAMAD JUOI
Associate Professor
Faculty of Manufacturing Engineering
Universiti Teknikal Malaysia Melaka
Hang Tuah Jaya
76100 Durian Tunggal, Melaka

Tarikh: 28th JUNE 2013

Tarikh: 3/7/2013

** Jika Laporan PSM ini SULIT atau TERHAD, sila lampirkan surat daripada pihak berkuasa/organisasi berkenaan dengan menyatakan sekali sebab dan tempoh laporan PSM ini perlu dikelaskan sebagai SULIT atau TERHAD.

DECLARATION

I hereby declared this report entitled “The effect of colouring agent on the corrosion properties of glass composite tiles” is the result of my own research except as cited in references.

Signature

: 

Author's name

: LIYANA BINTI MUHAMAD ALI

Date

: 28th JUNE 2013

APPROVAL

This report is submitted to the Faculty of Manufacturing engineering of UTeM as a partial fulfilment of requirements for the degree of Bachelor of Manufacturing Engineering (Engineering Materials) with Honours, the members of supervisory committee is as follow:

.....

PM. DR. JARIAH BINTI MOHAMAD JUOI

(Principal Supervisor)

ABSTRACT

The purpose of this project is to study the effect of colouring agent on the corrosion properties of glass composite tiles. The raw material used is waste soda lime silicate glasses (SLSG) which are transparent and green coloured glass. The glass were milled and sieved to $< 75 \mu\text{m}$ and mixed with ball clay that acts as the filler. Waste SLSG (transparent and green) is mixed with ball clay according to the optimum ratio of 90:10 wt. % for transparent GCM and 85:15 wt.% for green GCM. The mixture are then pressed using uniaxial die pressing and sintered at 850°C with 1 hour holding time. Water absorption, porosity, and density analyses of both GCM produced are analyzed through ASTM C373. Corrosion analyses are carried out using different types of aqueous solution which are low and high concentration of acid and alkali based on Malaysian Standard (Determination of chemical resistance). The main crystalline phase in both GCM identified using X-ray diffraction (XRD) analysis is quartz (SiO_2). Microstructure analysis conducted using scanning electron microscope (SEM) reveals the presence of quartz indicating that crystallise take place during sintering. The physical properties of GCM produced from transparent glass and green coloured glass are 0.06% porosity, 0.03% water absorption, 1.79 g/cm^3 of bulk density; and 0.03% porosity, 0.02% water absorption, 2.05 g/cm^3 of bulk density; respectively. Highest corrosion rate for both transparent and green coloured GCM is observed after been soaked for 5, 12 and 13 days is Lactic acid solution (high concentration acid). The corrosion rates after 13 days of soaking in the Lactic acid solution are 3.84 mpy for transparent GCM and 3.92 mpy for green coloured GCM.

ABSTRAK

Tujuan projek ini adalah untuk mengkaji kesan pewarna pada sifat-sifat kakisan jubin kaca komposit. Bahan mentah yang digunakan adalah sisa soda kapur silikat kaca (SLSG) iaitu kaca telus dan kaca berwarna hijau. Kaca telah hancurkan dan diayak <math><75\ \mu\text{m}</math> dan dicampurkan dengan bebola tanah liat yang bertindak sebagai pemangkin. Kaca SLSG (telus dan hijau) dicampurkan dengan bebola tanah liat mengikut nisbah optimum 90:10 wt. % untuk bahan komposit kaca telus dan 85:15 wt.% untuk bahan komposit kaca berwarna hijau. Pembentukan bahan campuran dijalankan menggunakan kaedah penekanan dan disinter pada suhu 850°C selama 1 jam. Penyerapan air, keliangan, dan ketumpatan dijalankan untuk menganalisis kedua-dua bahan komposit kaca melalui ASTM C373. Analisis hakisan dijalankan dengan menggunakan asid dan alkali berkepekatan rendah dan tinggi berdasarkan Piawaian Malaysia (penentuan rintangan bahan kimia). Fasa utama dalam kedua-dua kristal GCM dikenal pasti menggunakan pembelauan sinar-X (XRD) analisis adalah kuarza (SiO_2). Analisis mikrostruktur dijalankan menggunakan mikroskop imbasan elektron (SEM) menunjukkan kuarza hadir dan membawa kenyataan yang ia berlaku semasa pensinteran. Ciri-ciri fizikal bahan komposit kaca yang dihasilkan daripada kaca telus dan kaca berwarna hijau adalah 0.06% keliangan, 0.03% penyerapan air, $1.79\ \text{g/cm}^3$ ketumpatan pukal dan 0.03% keliangan, 0.02% penyerapan air, $2.05\ \text{g/cm}^3$ ketumpatan pukal. Kadar hakisan tinggi untuk kedua-dua bahan komposit kaca telus dan kaca berwarna hijau di kenal pasti selepas direndam selama 5, 12 dan 13 hari adalah apabila di dalam larutan asid laktik (asid kepekatan yang tinggi). Kadar hakisan selepas 13 hari direndam dalam larutan asid laktik adalah 3.84 mil setahun untuk bahan komposit kaca telus dan 3.92 mil setahun untuk bahan komposit kaca berwarna hijau.

DEDICATION

Dedicated to my beloved parents, family and friends

ACKNOWLEDGEMENT

First of all would like to thank Allah for His blessing to me for giving a good health and thoughts during this project. I would also like to express my deepest gratitude to my supervisor, PM Dr. Jariah Binti Mohamad Juoi, for her source of guidance, assistance, inspiration and concern throughout my research project.

Special thanks to my friends for giving me supports and helps especially for sharing, discussing the knowledge with me and in periods of uncertainties and difficulties. Their support, opinions, and advices will not be forgotten.

Finally, I would like to express my gratitude with highly appreciation and dedication to my parents and family for their love, concerns and encouragements.

TABLE OF CONTENTS

Title	Page
Abstract	i
Abstrak	ii
Dedication	iii
Acknowledgement	iv
Table of Contents	v
List of Tables	ix
List of Figures	xi
List of Abbreviations, Symbols and Nomenclatures	xiii

CHAPTER 1: INTRODUCTION

1.1	Background of study	1
1.2	Problem statement	2
1.3	Objectives	4
1.4	Scope of study	4

CHAPTER 2: LITERATURE REVIEW

2.1	Introduction	6
-----	--------------	---

2.2	Waste	6
	2.2.1 Waste glass	7
2.3	Recycling glass waste	7
	2.3.1 Soda lime silicate glass (SLSG)	10
	2.3.2 Composition and properties of soda lime silicate glass	11
	2.3.3 Glass colouring	14
2.4	Glass composite from waste	14
	2.4.1 Definition	14
	2.4.2 Processing of glass composite	16
	2.4.2.1 Batch composition	17
	2.4.2.2 Forming	18
	2.4.2.3 Sintering	19
	2.4.3 Properties of glass composite from waste	22
	2.4.3.1 Physical properties	23
	2.4.3.2 Corrosion properties	23
	2.4.3.3 The effect of colouring agent on glass composite	24
2.5	Summary of literature review	25

CHAPTER 3: METHODOLOGY

3.1	Introduction	26
3.2	Material preparation	28
	3.2.1 Soda lime silicate glass (transparent and coloured)	28

3.2.2	Ball clay	31
3.2.3	Powder processing	32
3.3	Fabrication of glass ceramic composite	34
3.3.1	Forming (Uniaxial die pressing)	36
3.3.2	Sintering	37
3.4	Material characterization	38
3.4.1	X-ray Diffraction (XRD)	38
3.4.2	Scanning Electron Microscope (SEM) & Energy dispersive X-ray (EDX)	38
3.5	Physical properties analyses	39
3.5.1	Water absorption, porosity, and density analyses	39
3.6	Corrosion test	40
3.6.1	Aqueous solution	41
3.6.2	Preparation/Procedure of glass composite tiles	41
3.6.3	Corrosion rate	42
3.6.4	Corrosion evaluation	43

CHAPTER 4: RESULTS AND DISCUSSIONS

4.1	Introduction	44
4.2	Physical properties analyses	44
4.2.1	Porosity	44
4.2.2	Water absorption	45
4.2.3	Bulk density	46

4.2.4	Discussion on physical properties analysis	47
4.3	Corrosion properties analysis	48
4.3.2	Determination of class	48
4.3.2	Corrosion rate	51
4.3.3	Discussion on corrosion properties analysis	53
4.4	Phase analysis	56
4.5	Microstructure analysis	58
4.6	Elemental analysis	60
CHAPTER 5: CONCLUSIONS AND RECOMMENDATIONS		
5.1	Conclusions	64
5.2	Recommendations for future work	66
REFERENCES		67
APPENDIX		73
	Gantt chart PSM 1	74
	Gantt chart PSM 2	75

LIST OF TABLES

Table	Title	Page
2.1	Summary of main alternatives uses of waste glass	9
2.2	Compositions and Characteristics of Common commercial	12
2.3	Properties of soda lime silicate glass for container	13
2.4	Properties of the glass composite and natural	15
2.5	Optimum batch composition of transparent and green coloured GCM	17
2.6	Feed materials and shapes of green body for common ceramic forming methods	18
3.1	Chemical composition of transparent SLSG	30
3.2	Chemical composition of green coloured SLSG	30
3.3	Chemical composition of ball clay	32
4.1	The determination of class of transparent GCM after corrosion testing	50
4.2	The determination of class of green coloured GCM after corrosion testing	50
4.3	Surface morphology of GCM before and after 13 days of soaking in lactic acid solution	59

4.4	Cross-section morphology of GCM before and after 13 days of soaking in lactic acid solution	59
4.5	SEM micrograph of the surface of transparent GCM before and after 13 days of soaking in lactic acid solution	61
4.6	SEM micrograph of the surface of green coloured GCM before and after 13 days of soaking in lactic acid solution	62
4.7	The element contain in transparent and green coloured GCM before and after corrosion testing	63

LIST OF FIGURES

Figure	Title	Page
2.1	Phase diagram of $\text{Na}_2\text{-CaO-SiO}_2$	10
2.2	Flow chart of the production of glass composite by firing consolidate powders	16
2.3	Schematic representation of the steps in uniaxial die pressing method	19
2.4	Schematic representation of microstructure changes that occur during sintering	22
2.5	Coloured recycle glass bottles	25
3.1	Flow chart of experimental work	27
3.2	Recycle transparent and green coloured glass bottles	29
3.3	Glass bottles after crushing	29
3.4	Ball clay	31
3.5	Process flow of material preparation	33
3.6	Sieve with mesh size of 75 μm	34
3.7	Flow chart of processing ceramic product using die method	35
3.8	Mixed powder in 18 mm \times 18 mm mould is press using uniaxial die pressing	36

3.9	Schematic diagram of common die geometry for dry	36
3.10	Sintering profile	37
4.1	The percentage of porosity of transparent and green coloured GCM	45
4.2	The percentage of water absorption of transparent and green coloured GCM	46
4.3	Bulk density of transparent and green coloured GCM	47
4.4	Corrosion rate for transparent GCM	52
4.5	Corrosion rate for green coloured GCM	53
4.6	XRD pattern of GCM produced from transparent and green SLSG at 850°C	57

LIST OF ABBREVIATIONS, SYMBOLS AND NOMENCLATURE

%	-	Percent
°C	-	Degree Celsius
ρ	-	Density
μm	-	Micrometer
ASTM	-	American Society Testing Material
cm^3	-	Centimetre cube
EDX	-	Energy dispersive X-ray
g	-	Gram
GCM	-	Glass composite materials
GPa	-	Giga Pascal
h	-	Hour
HK	-	Knoop hardness
J	-	Joule
K	-	Kelvin
K_{IC}	-	Critical stress intensity factor
min	-	Minute

MPa	-	Mega Pascal
nm	-	Nanometre
SEM	-	Scanning electron microscope
SLSG	-	Soda lime silicate glass
T _g	-	Glass transition temperature
UV	-	Ultraviolet
wt. %	-	Weight percentage
XRD	-	X-ray diffraction

CHAPTER 1

INTRODUCTION

1.1 Background of study

Waste is an unavoidable by-product of most human activity. According to United Nations Economic and Social Commission for Asia and the Pacific Organization (2013), economic development and rising living standards in the Asian and Pacific Region have led to increases in the quantity and complexity of generated waste. There is a great concern regarding to the increasing amount of industrial wastes such as plastics, glasses, and grog (fired clays). The disposal of these wastes is one of the issues that have received a lot of attention and a high demand for the safety of the environment. One technique used to reduce such wastes is by recycling, which is not only benefits to the environment but also to the economy (Loryuenyong *et. al.* 2009). The success of glass recycling programs has resulted in more post-consumer glass than municipalities can use; there is also a post-industrial glass waste available. The development of alternate products that make use of both types of glass helps in many ways: by reducing landfill, by reducing the need for recycled glass storage, and by supporting the economic viability of recycling programs.

There are different kinds of wastes that have already been recycled in ceramic industry for example waste glasses, which are readily incorporated as an

alternative ceramic raw material or as a fluxing agent in stoneware, tiles, bricks and concrete (Brown and Mackenzie *et. al.* 1982). However, among these, many thousands tones of glass are discarded, much in the form of non-returnable bottles and containers. Glass is an ideal material for recycling. The use of recycled glass in new container helps save of energy. It helps in brick and ceramic manufacture, and it conserves raw materials, reduces energy consumption, and the volume of waste sent to landfill (Turgut and Yahlizadec *et. al.* 2009).

The glasses and ceramics are consist of oxide, fluoride, silicate, borate, phosphate, and related compositions that make up common ceramic materials. These materials are electrically insulating and contain few free carriers, so that chemical attack on their surfaces is mainly by acid-base type of reactions rather than electrochemical corrosion involving redox reactions. Crystalline ceramics and non-crystalline glasses of similar composition often have quite different corrosion behaviour showing that structure at the atomic scale also plays a role. Interest in glass corrosion comes from the necessity of understanding stability of glass containers, sheet glass, and other glasses in the presence of aqueous solutions (White *et. al.* 1981). In general, the solutions are highly under saturated, may be strongly acidic or basic, and the concern is with times in the range of a few years. In contrast, the study on the dissolution (corrosion) of crystalline ceramics is relatively rare because in the environments in which most of these materials are use, attack by aqueous solutions is not a problem to the environment (White *et. al.* 1981).

1.2 Problem statement

Humans have always produced trash and have always disposed of it in some way, so solid waste management is not a new issue. What have changed are the types and amounts of waste produced, the methods of disposal, and the human values and perceptions of what should be done with it. Waste productions are

always increasingly expected to be great, so, the major waste that should be in great concern is the production of glass waste. The production of glass composite materials made from recycling industrial waste can be classified as one of the known technology. According to literature, during the last decade the use of ecologic products has become an important aspect in the ceramic industry, in order to optimize and reduce the consumption of natural resources. Recycled glass can be obtained from several sources such as flat glass, glass bottle, lamps, television screens, etc. Moreover, its amorphous nature and composition (mainly silicon oxide, calcium oxide and sodium oxide) make it an excellent candidate for application in the ceramic industry as a flux agent and/or frits for glazes (Fraga *et. al.* 2011). Many researchers has paid a lot of attention to produce glass, glass ceramic, sintered ceramic materials made from waste glass to make those materials a better production to lead to environment. Waste glass obtained from construction wastes, industrial wastes, and medical wastes have desirable properties to fulfil in most applications such as tiles and roof for constructions, interior facing of containers for chemical/medical industry and as well as road surfacing. For that reason, the recycled glass has to possess compositional homogeneity and must be available in large quantities. The aim of the present work is to study corrosion properties of glass composite materials from recycled glass and ceramic waste as raw materials in the production of ceramic tiles.

Tiles are generally used for covering roofs, floors, walls, showers, or other objects such as tabletops. Alternatively, tile can sometimes refer to similar units made from lightweight materials, typically used for wall and ceiling applications. Tiles are often used to form wall and floor coverings, and can range from simple square tiles to complex mosaics. In many years, tiles are mostly applied in various applications that are highly exposed in different environment. The present study aims to investigate the corrosion properties of glass composite tile made from waste glass. The study involves chemical resistance of unglazed tiles against low concentration acid and alkali, and high concentration acid and alkali.

1.3 Objectives

There are several objectives of this project, which are:

1. To determine the physical properties of glass composite tile made from SLSG (transparent and green coloured) waste glass.
2. To characterize the microstructure and phases present in the produced glass composite samples.
3. To evaluate the corrosion properties of glass composite tile made by different SLSG waste (transparent and green coloured) glass.

1.4 Scope of study

In order to investigate the corrosion properties of glass composite tiles made from waste glass, the scope of the study is tailored to produce glass ceramic samples for structural application and chemical resistance with high corrosion resistance. The fabrication process was carried by common ceramic techniques (e.g. uniaxial pressing method). The study started by preparing recycled glass as raw materials that used in fabrication of glass ceramic samples. The glass powders were prepared by crushing bottles made from glass using hammer until they passed through a sieve of less than 75 μ m to obtain fine particles. The stage of process was followed by mixing the glass powder and ball clays with optimum ratio of 90:10 for transparent SLSG and 85:15 for green coloured SLSG. Ball clays act as fillers to improve the bonding properties between particles and help to obtain dense materials by sticking glass powders together during pressing method (Rozenstrauha *et. al.* 2006).

Forming process was then conducted to form the mixing glass powder into green ceramic article. Green was referred to the unfired ceramic. Preferably uniaxial die pressing is used with 3.5 gram of mixed powder per sample is pressed with 40 MPa (3.5 tons) in a cube shape with measurement 18 mm \times 18 mm. The pressed article is then removed from the die and sintered in a furnace. The

sintering temperature of this study would be the optimum sintering temperature based on previous investigation which is 850°C. For each sintering, the duration of holding time was be one hour. At the end of this study, the sintered glass ceramic samples were then subjected to characterization, physical properties and corrosion properties analyses. The physical properties were analyzed using water absorption, porosity, and bulk density analyses, while the corrosion properties were analyzed through chemical test using weight loss technique. The microstructure analysis of the surface of the fabricated of glass ceramic were observed using the X-ray diffraction (XRD), scanning electron microscope (SEM) and energy dispersive X-ray (EDX). Results on the different waste glass utilized (transparent glass and green glass) were then compared in order to understand the effect of colouring agent on the glass composite corrosion.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

This chapter discussed the review related study on glass composite made from waste glass. The content is focusing on the properties and process preparing the glass composite produced from waste glass.

2.2 Waste

Waste is an unavoidable by-product of most human activity. Shirke (2009) referred waste as rubbish, trash, garbage, or junk is unwanted or unusable material. According to European councils' directive "Waste is any substance or object which the holder discards or intends or is required to discard." There are many source of waste such as residential, industrial, commercial, construction, municipal services, and institutional.

Solid waste can be defined as any solid or semi-solid substance or object resulting from human or animal activities, discarded as useless or unwanted (Shirke *et. al.* 2009). It is an extremely mixed mass of wastes, which may originate from household, commercial, industrial or agricultural activities; according to its contents (organic material, glass, metal, plastic paper etc); or