

## The Effect of Boron Oxide on Concentration of Iron Ion Released from $\text{SiO}_2\text{-B}_2\text{O}_3\text{-Na}_2\text{CO}_3\text{-Fe}_2\text{O}_3$ Glass System

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**ABSTRACT.** Iron ion from borosilicate glass can be release in seawater depending on the immersion time by the corrosion process. The effect of boron oxide percentage in the quaternary system of glass was studied. The compositions of the glass system [wt.%, (55-x)%  $\text{SiO}_2$ (15-x)%  $\text{B}_2\text{O}_3$ .25%  $\text{Fe}_2\text{O}_3$ .5%  $\text{Na}_2\text{CO}_3$ ] where  $15\% < x < 40\%$  have been prepared with the different amount of  $\text{B}_2\text{O}_3$ (35-55 wt.%) and  $\text{Fe}_2\text{O}_3$  with  $\text{Na}_2\text{CO}_3$  were fixed. The glass was melting in the alumina crucible for 2 hours soaking time at 1300 °C and cooled to room temperature. The obtained glass was crushed and sieved with yielding a mean size of 2-4 mm. The glass samples in 50 mL synthetic seawater was tested under static condition in 16 weeks to determine the concentration of iron ion and weight loss of the glass samples. The concentration of iron ion was analyzed using UV-Vis Spectrophotometer. The scanning electron microscopy determined the morphology of the sample. From the dissolution studies, it was shown that the concentration of iron ion release was increased with the increasing percentage of  $\text{B}_2\text{O}_3$  content. However, the addition of silica oxide was increased the chemical durability of glass system. The percentage boron oxide at 55 wt.% of boron gives the optimum rate of iron ion released in seawater, which is 1.11ppm.

**Keywords:** Boron oxide, Ion released, Iron ion;

*Received:* 15.10.2017, *Revised:* 15.12.2017, *Accepted:* 30.02.2018, and *Online:* 20.03.2018

**DOI:** 10.30967/ijcrset.1.S1.2018.130-134

*Selection and/or Peer-review under responsibility of Advanced Materials Characterization Techniques (AMCT 2017), Malaysia.*

### 1. INTRODUCTION

Pure borate glasses have a poor chemical durability when it comes to application and it is important in development of new materials such as oxides glass [1]. Bengisu in 2016 reported that, borate glass has high affinity to water but when it is combined with other oxide such as  $\text{SiO}_2$  or  $\text{Al}_2\text{O}_3$ , the chemical durability can be improved by changing the structural [2]. The Controlled Release Glass (CRG) is a class of materials that completely dissolve in aqueous media. Their degradation of corrosion controlled process that follows zero order release over the life of the material, for example the release rate is constant and independent of time and concentration [3].

The glass structure is depends on two factors; network former and network modifier. Glass former or known as network formers consists of oxide such as  $\text{SiO}_2$ , and  $\text{B}_2\text{O}_3$ , which are indispensable in the formation of the glass. Meanwhile, network modifier acts as chemical additional in a glass system, which change the boron coordination from trigonal to tetrahedral [4] and also break the framework and produce non-bridging

oxygen [5]. The chemical durability, which is the resistance offered by a glass towards attack by aqueous solution and atmospheric agents, the releasing ion from the synthetic seawater will be affected whether slow or controlled release. It is very important for possible reaction between the glass surface and acid or aqueous medium [6].

Iron oxide ( $\text{Fe}_2\text{O}_3$ ) was known as fertilizer, which is promoting the algae growth and also have a good effects of chemical durability in phosphate glass [6]. Previous studies show that transition metals such as  $\text{Al}_2\text{O}_3$ ,  $\text{ZrO}_2$ ,  $\text{MnO}_2$  and  $\text{TiO}_2$  have low softening point and tend to promote the chemical durability of the glass [7-13]. The presence of  $\text{Fe}_2\text{O}_3$  will cause the  $\text{Fe}^{3+}$  ion substituted at the silicon sites in the  $\text{SiO}_2$ - $\text{Na}_2\text{O}$ - $\text{Al}_2\text{O}_3$ - $\text{B}_2\text{O}_3$  glass system [14].

The aim of this work is to study the effect of  $\text{B}_2\text{O}_3$  percentage on the iron ion released from the  $\text{SiO}_2$ - $\text{B}_2\text{O}_3$ - $\text{Na}_2\text{CO}_3$ - $\text{Fe}_2\text{O}_3$  glass system. Indeed, the  $\text{SiO}_2$  percentage is added along the  $\text{B}_2\text{O}_3$  percentage with the total is 70 wt.%. Therefore, glass dissolution test method was used to determine the concentration of iron ion with different percentage of boron oxide. Then, the morphology of glasses was evaluated from the scanning electron microscopy (SEM).

## 2. MATERIALS AND METHODS

**2.1 Synthetic Seawater Preparation.** 41.95 g of sea salt (Sigma-Aldrich) was weighed and mixed with 1 liter of deionized water using 1 L volumetric flask. Then, the solution was adjusted with 0.1 N HCl or NaOH to obtain pH 8.2

**2.2 Glass Preparation.** The chemicals compositions of glasses were shown in Table 1. The mixing reagent grade raw materials, such as  $\text{SiO}_2$ ,  $\text{B}_2\text{O}_3$ ,  $\text{Na}_2\text{CO}_3$ , and  $\text{Fe}_2\text{O}_3$  were prepared for 100 g. The glass has been prepared by melting in the alumina crucible within 2 hours of soaking time at 1300 °C in the furnace and cooled to room temperature. The ion released and weight loss is determined through the immersion of the glass samples in 50 mL synthetic seawater for 16 weeks under the static condition. The morphology of sample were determined by the Scanning Electron Microscopy (SEM) (JOEL JDM-6360LA) was used in this study.

**Table 1** The chemicals compositions of glass (wt.%)

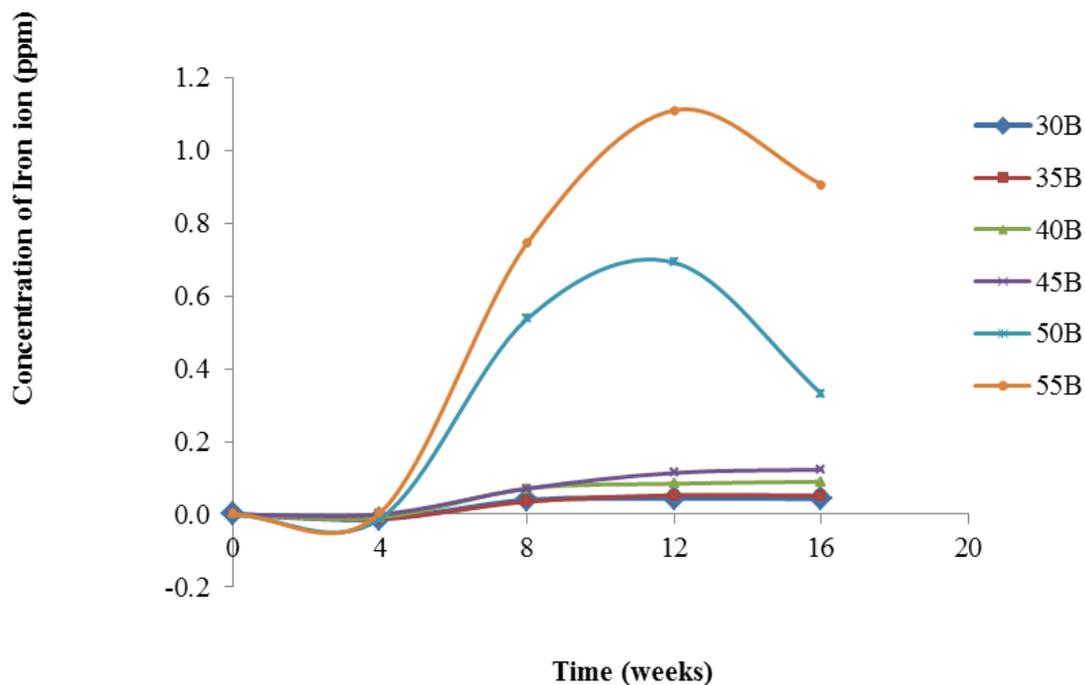
Glass Coding	Compositions (wt.%)			
	$\text{SiO}_2$	$\text{B}_2\text{O}_3$	$\text{Na}_2\text{CO}_3$	$\text{Fe}_2\text{O}_3$
30B	40	30	5	25
35B	35	35	5	25
40B	30	40	5	25
45B	25	45	5	25
50B	20	50	5	25
55B	15	55	5	25

**2.3 Glass Dissolution.** The chemical durability of glasses was determined by measuring the weight loss of the samples immersion. From the measurement, 1.0 g of glass samples were crushed in size of 2.0-4.0 mm was immersed in 50 ml of synthetic seawater under static condition for 16 weeks (112 days). The concentration of iron ion was analyzed using UV-Vis Spectrophotometer (UV 1800) with phenanthroline method [15] and weight loss of glasses was measured.

**2.4 Weight loss % experiment.** In 100 ml polyethylene bottles with covers, 1 g of glass grains (2.00-4.0 mm) was immersed in 50 ml of synthetic seawater for 16 weeks at room temperature. The weight loss (%) was calculated based on weight of sample before and after immersion.

### 3. RESULTS AND DISCUSSION

The effect of  $B_2O_3$  on the iron ion release from the synthetic seawater was studied for six types of glass with different  $B_2O_3$  content in a range of 30-55 wt.%. The iron ion released from synthetic seawater is illustrated in Fig. 1. It is show that the iron ion concentration increased linearly with the time but started to decrease at 12 weeks. This is due to the chemical durability of the glass in synthetic seawater [16]. Previous studies show that, normally  $B_2O_3$  substituted for  $SiO_2$  in network former sites in borosilicate glasses decreases the chemical durability of a glass and also increased the ion released [17].



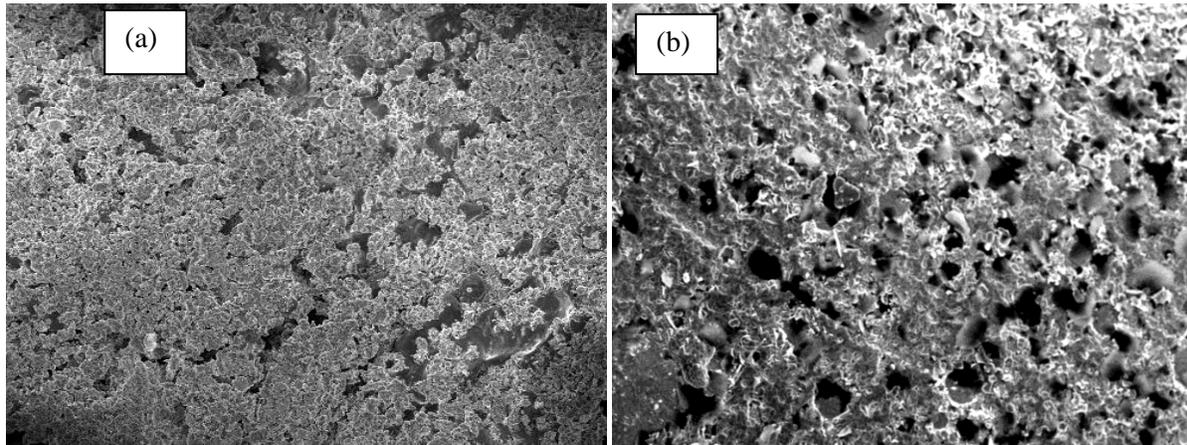
**Fig. 1** Concentrations of iron ion released from 0-16 weeks

The Fig. 1 shows the concentration of iron ion increased with increasing percentage of boron oxide content. From the results it can be seen that, the glass samples for 30B40Si, 35B35Si, 40B30Si and 45B25Si showed low concentration of iron ion. Since, the less of iron ion released, so that the pH of seawater tent to increased (alkaline solution) which is slow down the corrosion process [18]. Increase the amount of boron oxide and decrease the content of silica, the concentration iron ion increased. This is due to the solubility of the glass increased [19].

The iron ion released is depends on the glass composition. The boron anomaly in the borosilicate glass changed the coordination numbers from three to four and back to three depends on the addition of network modifier as alkalis or alkaline earth oxides. So that, when the glass is reacted in the acidic medium, the modifier cation can be exchanged with  $H^+$  in liquid medium ( $OH^-$ ).The ferrous ion was released when the content of the boron oxide increased [20].

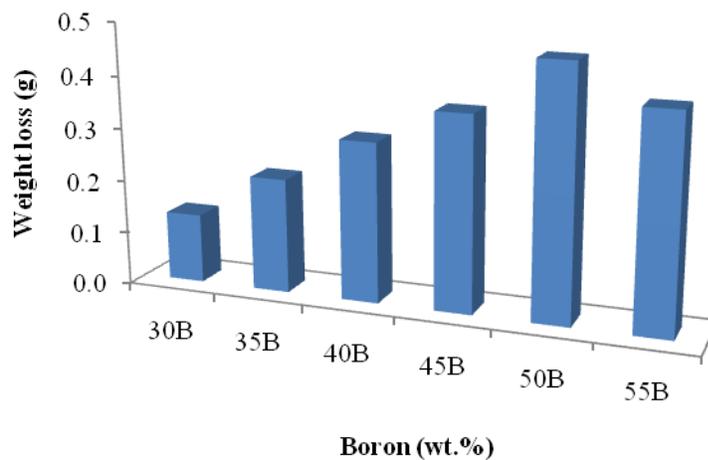
SEM was conducted to further investigate on the surface morphology of glasses and also the size of pores. Fig. 2 show the SEM phase of 55B for (a) before and (b) after immersion for 16 weeks. The observation before

the immersion, there is less pores on the surface of glass. Meanwhile, after the immersion at 16 weeks, there is more pores appeared. This is owing to the iron ion was released out from the glass.



**Fig. 2** SEM morphology for 55B glass (a) Before immersion and (b) After immersion for 16 weeks

The types of ions in a glass structure generally tend to release out of the glass surface when reacted in an aqueous solution. The ion released rate is depends on the glass composition and the state of the ions in the glass structure. From the Fig. 3, it is shown that the weight loss with different percentage of boron oxide content (wt.%). As percentage of boron oxide content increased, the weight loss of the glass also increased (0.13-0.47 g). The 50B20Si shows the highest value of weight loss, which is 0.471 g. This is due to the corrosion process when the glasses make contact with the aqueous medium [21] and the solubility of water increased [14].



**Fig. 3** The weight loss of different contents of boron oxide (wt.%)

#### 4. SUMMARY

In summary, this is clearly observed, the decreasing amounts of  $\text{SiO}_2$  give the effect of chemical durability of the glass system, which is low and less stable. The concentration of iron ion was increased with the

increasing percentage of B<sub>2</sub>O<sub>3</sub> content. The percentage boron oxide at 55 wt.% of boron gives the optimum rate of iron ion released in seawater, which the concentration is 1.11 ppm.

#### ACKNOWLEDGEMENT

The authors are grateful for the financial support from research grant FRGS 55095 and also thanks to Universiti Malaysia Terengganu for the facilities.

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