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Leaching of Pb^{2+} and Te^{4+} in Yb^{3+} doped lead tellurite glasses in aqueous solution

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ABSTRACT

A series of Yb doped lead oxychloride tellurite glasses with composition of (80-x)TeO_2-10PbO-10$PbCl_2$-xYb_2O_3 were prepared using conventional melt quenching technique. These glasses were then immersed in distilled water (pH 4 and pH 9) for 10 days. The quantitative analysis of Pb^{2+} and Te^{4+} were made using induced coupled plasma-mass spectrometer (ICP-MS). The results showed that Te^{4+} and Pb^{2+} were highly leached in acidic solution or at lower pH compared to alkaline solution and the addition of Yb^{3+} in the glass system had reduced or maintained the leaching of Te^{4+} and Pb^{2+} from the glasses.

| Tellurite Glasses| Leaching of Te and Pb | Solution pH | Yb doped |

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1. INTRODUCTION

Since the discovery of tellurite as glass former when fused with small amount of LiO_2 [1], there were lot of research efforts dedicated on these glasses due to their potential applications for optical devices and optical amplifiers. Tellurite glasses possess high refractive index, large third-order nonlinear susceptibility and low phonon energy, which are capable of reducing the nonradiative loss due to multi-phonon relaxation [1]. These glasses are also known as very good host for rare earth ions which makes their properties to favourably change and also useful for practical laser application [2].

Durability of glass is one of the important factors that need to be considered in material selection for technological implementation. Glass corrosion occurs when it is contacted directly to water or aqueous solution. There are two types of corrosion that occurs in glass which are the preferential corrosion where only some components are dissolved into the leaching media, while the rest remain to form a stable protective film and full corrosion where all the composition can enter into the leaching media which obviously shows that the glass with preferential corrosion is of good chemical durability and the glass with full corrosion is of poor chemical durability since there is no protective film is formed [3].

Previous studies showed that tellurite glass has a reasonable chemical durability [4]. The objective of this paper is to find out the influence of Yb^{3+} on the leaching of Te^{4+} and Pb^{2+} in aqueous solution.

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2. EXPERIMENTAL

2.1 Samples preparation

A Series of Yb doped lead oxychloride tellurite glasses having composition of (80-x)TeO_2-10PbO-10$PbCl_2$-xYb_2O_3 with ($0.0 \le x \le 3.0$) were prepared using conventional melt quenching technique. The powdered materials were first weighed and mixed on an electronic analytical balance and milled before they were placed in a platinum crucible.

The mixtures were then placed in an electric furnace for melting process at 900°C for about 20 m inutes. The mixtures were rotated from time to time during the melting process to ensure the homogeneity of the samples. The homogenized melts were poured onto a stainless steel plates and were annealed at 250°C.

Table 1: Composition of prepared samples

No.	Sample	TeO_2 (mol%)	PbO (mol%)	$PbCl_2$ (mol%)	Yb_2O_3 (mol%)
1	S1	80.0	10	10	0.0
2	S2	79.5	10	10	0.5
3	S3	79.0	10	10	1.0
4	S4	78.5	10	10	1.5
5	S5	78.0	10	10	2.0
6	S6	77.5	10	10	2.5
7	S7	77.0	10	10	3.0

2.2 Corrosion test

The successfully prepared glasses were grinded and polished to eliminate the surface defects that may influence the results for corrosion test. After ensuring the flatness of the glasses, the samples were cut into specific size of 2 mm x 10 mm x 10 mm and then immersed in distilled water and aqueous solution of pH 4 and pH 9 under static condition for 10 days.

The solution was placed in a 250 ml beaker and was sealed by aluminium foil to avoid inclusion of dust of insects. After 10 days, the samples were removed from the pH solutions and distilled water. The leaching of Te^{4+} and Pb^{2+} into the solution was measured using inductively coupled plasma mass spectrometer (ICP-MS) model ELAN 6100 PERKIN ELMER.

3. RESULTS & DISCUSSION

3.1 Sample preparation

The samples were visually stable in air. The X-ray diffraction measurement was carried out using Siemens Diffractometer D5000 to determine the amorphous nature of the samples. The XRD spectrum for S1 is showed in Figure 1. It can be seen that a broad halo shaped hump appears in the spectrum instead of sharp peaks which confirmed the amorphous nature of the samples. Every other sample also showed the same shape of XRD spectra which also conclude the amorphous nature of the glasses.

3.2 Leaching behaviour

A plot of leached Pb^{2+} and Te^{4+} versus Yb content was produced and the result is presented in Figure 2. From the figure 2 (a), it can be seen that the leaching of Pb^{2+} in solution of pH 4 is higher then the leaching of Te^{4+}. This is because the glass corrosion mechanism in acidic solution acts differently then in alkali solution. The acidic solution attacks the alkali ions. By dissolving the alkali ions in the glass system, a porous surface was left which consists of the network of the glass host (tellurite) with holes due to the removal of alkali ions by acid.

Figure 2 (b) shows the comparison of leached Pb^{2+} with Te^{4+} versus Yb content in distilled water. It can be seen that the amount of leached Pb^{2+} and Te4+ is slightly the same. The previous study reported that the corrosion mechanism of glasses in distilled water is the same as the mechanism in acidic solution but so much slower which explains the low amount of Pb^{2+} and Te^{4+} leached in the solution.

Figure 2 (c) on the other hand shows the opposite where the amount of Te^{4+} leached is higher than Pb^{2+}. This is because unlike acidic solution, alkali solution attacks directly to the surface of the glass. When the glass was contacted with the alkali solution, the attack acts directly to the glass surface which is rich with tellurite. This explains the high amount of Te^{4+} leached compared to the amount of Pb^{2+} leached in the alkali solution.

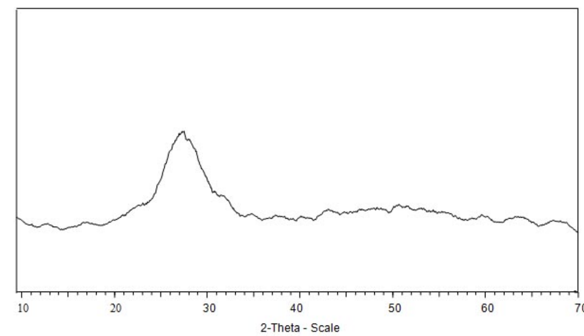

Fig. 1 The X – ray diffraction spectra of a sample

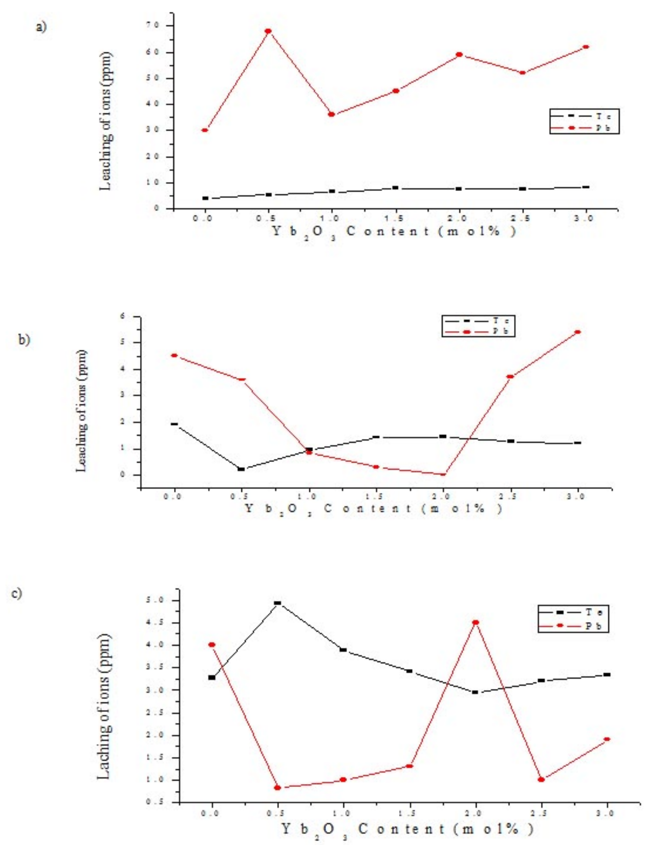

Fig. 2 Comparison of leached amount of Te^{4+} and Pb^{2+} versus Yb^{3+}content after immersed in a) pH 4, b) distilled water and c) pH 9 for 10 days

As the glass surface is in contact with the solution, the protons or hydroxonium ions will diffuse into the glass network and leads to the ion exchange between proton and alkali network modifier [5-7]. The water species will reacts with the glass network so that the alkali is freed and hydrogen ions replace the alkali's place. The possible reactions of these mechanisms are as follows:

$Te\text{-}OPb_{(gl)} + H_3O^+_{(s)} \longrightarrow Te\text{-}OH_{(gl)} + Pb^{2+}_{(s)} + H_2O$ (1)

$Te\text{-}OPb_{(gl)} + H_2O_{(s)} \longrightarrow Te\text{-}OH_{(gl)} + Pb^{2+}_{(gl)} + OH^-$ (2)

Firstly, the Pb^{2+} is leached out from the glass matrix into the solution as an exchange process with the H^+ from the solution. Thus forms the Te-OH network in the glass matrix and Pb^{2+} in the solution and water molecules. The water molecules are then reacts with the Te-OH network thus repeated the same process with the glass surface. Meanwhile, for the Te^{4+}, the corrosion process that takes place could be in the form of:

$$Te\text{-}OH + HO\text{-}Te \longrightarrow Te\text{-}O\text{-}Te + H_2O \quad (3)$$

Leaching occurred rapidly at the surface and slowed down as the leach size grew. This was perhaps a diffusion controlled process and explained why the leached amount of Te^{4+} was smaller than the leached amount of Pb^{2+} in the aqueous solution [5].

Figure 3 shows the leached amount of Te^{4+} and Pb^{2+} in various pH of solution versus the Yb_2O_3 content in the glass system. Figure 3a) shows that the amount of leached Te^{4+} while Figure 3 b) shows the amount of leached Pb^{2+}

Figure 3 a) showed that the amount of leached Te^{4+} in the solution of pH 4 is the highest followed by the amount in pH 9 and distilled water. however, the differences is not really big compare to the amount of leached Pb^{2+} in Figure 3 b) where the amount of leached Pb^{2+} in pH 4 is really high and the leached amount in pH 9 and distilled water was really low.

This is because, in pH 4, acidic solution attacks the alkali ions, unlike the attacks in pH 9 where the attacks occurs directly to the surface of the glass. However, due to the relatively high acidic condition, the rate of attacks occurs quite fast which cause the high leached amount of Te^{4+}.

This mechanism also explains the large differences of leached amount of Pb^{2+} in Figure 3 b). In this figure, it can be seen that the leached amount of Pb^{2+} was really high in acidic solution compared to the leached amount in pH 9 and distilled water. This is because in acidic solution, the highly leached of Pb^{2+} occurs because the attacks occurred directly to Pb^{2+} ions while in pH 9, the attacks occurred directly to the surface where Te^{4+} leached more in the surface.

The attacks of distilled water act similarly with the attacks of acidic solution. However, the rate was so much slower which cause the leached amount to be so much lower compared to the attacks of solution of pH 4 and pH 9.

Figure 3 also showed the influences of Yb^{3+} content in the glass network to the leaching of Te^{4+} and Pb^{2+} from the glass system. From Figure 3 a), it can be seen that the leaching amount of Te^{4+} increases with the addition of Yb^{3+} in the glass network for the glass immersed in pH 4 solution. However, as the Yb^{3+} content acceding 1.5 mol%, the leaching amount decreases. The leaching amount of Te^{4+} in solution of pH 9 and distilled water on the other hand, showed that the addition of Yb^{3+} content in the glass network helped in maintaining the leached amount of Te^{4+} from the glass.

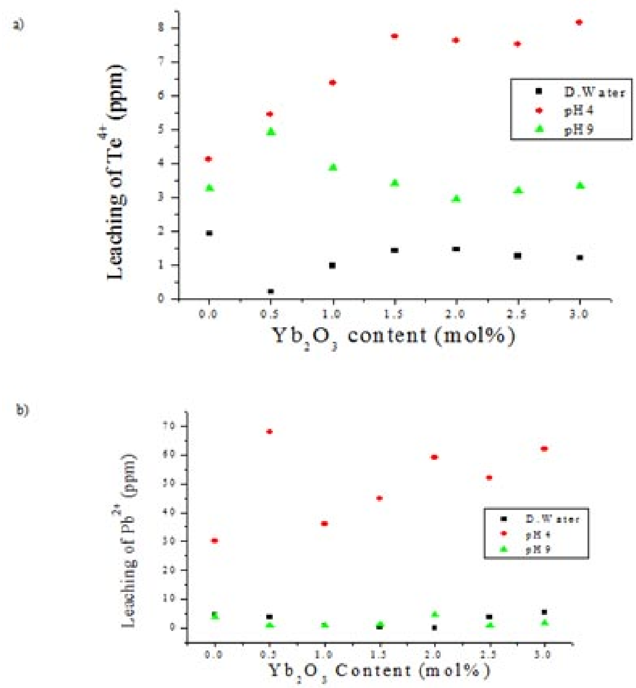

Fig. 3 Leaching of a) Te^{4+} and b) Pb^{2+} after immersed in aqueous solution of pH 4, pH 9 and distilled water for 10 days

Figure 3 b) also showed the same phenomenon for the leached of Pb^{2+}. From the figure, it can be seen that the amount of Yb^{3+} in the glass network. Although the glass immersed in pH 4 lower the leached amount of Pb^{2+} from the glass with the addition of Yb^{3+} in lower amount, but as it exceeds 2 mol%, the leached amount of Pb^{2+} seems to be maintained.

The plot of glass immersed in distilled water and pH 9 on the other hand, showed a significant result on how the amount of Yb^{3+} in the glass system helped in lowering the leached amount of Pb^{2+} from the glass.

Previous research had reported that the addition of rare earth in the glass network can control the corrosion of glass in aqueous solution. As the Yb^{3+} is introduces into the glass network, the Yb^{3+} will enter the glass network because of its larger radius. This will result in the increasing of compactness of a glass structure and inhibits the diffusion of water into the glass matrix [8].

It is clearly seen that the leaching of Pb^{2+} is higher than those of Te^{4+}. It was believed that the leaching process is due to the ion exchange mechanism. In this mechanism, the result is the release of freed ions and glass network constituent into the solution [9].

4. CONCLUSION

The leaching amount of Te^{4+} and Pb^{2+} were observed by ICP-MS. The measurement showed that the leached ions occurred in acidic solution is higher than in alkaline solution and in distilled water due to differences of attacking mechanism of acidic and alkali solution to the glass where the acid attacks the alkali ions from the glass while alkali solution attacks directly to the glass surface. The attacks of distilled water acts similarly with the acidic solution only the mechanism was so much slower. It was found that as the Yb^{3+} content was increased, the leaching amount was maintained. The leaching of Pb^{2+} was ion-exchange controlled while the leaching of Te^{4+} was diffusion controlled mechanism.

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