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Chemical corrosion and gamma-ray attenuation properties of Zr and Ti containing lead silicate glasses

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ABSTRACT

Lead silicate glasses (LSGs) have high gamma-ray attenuation but low chemical durability properties. In this work, LSGs with (55.5–68.5 wt%) PbO content containing ZrO_2 and TiO_2 additions were produced. The chemical corrosion of various produced LSGs in 0.5 N HNO_3 aqueous solution and determination of their gamma-ray attenuation coefficients for ^{60}Co and ^{137}Cs sources were investigated. The weight loss measurements, the SEM micrographs, the EDS analysis of the sample surfaces and the ICP analysis of solution were used to characterize the dissolution process. The effects of PbO content, ZrO_2 and TiO_2 additives on chemical corrosion, and also the effect of PbO on gamma-ray attenuation coefficient, glass transition temperature (Tg), and density of LSG glasses were determined. The results showed that by increasing the lead content of glass the gamma-ray attenuation coefficient, chemical corrosion and density were increased, but the Tg decreased. One of the samples with PbO contents of 65.4 wt% and SiO_2 content of 26.9 wt% showed a very low chemical corrosion behavior and good gamma-ray absorption property.

1. Introduction

Addition of lead in silicate glass reduces the melting point and network connectivity, but increases the density, refractive index, chemical corrosion, electrical conductivity, and radiation attenuation coefficient of X- and gamma-rays [1–7].

The chemical corrosion of LSG glasses have been the object of several investigations [5–7]. In low lead content (<50 mol% PbO) LSG glasses, Pb has the network modifying role [1,3,4,6] which generates non-bonding oxygen atoms. These atoms are the most efficient in increasing the chemical corrosion of glass. When a piece of LSG glass is placed inside the aqueous acid solution, two distinct reactions occur. First, the ion-exchange is started by the reaction (1):

$$\equiv Si{-}O{-}Pb{-}O{-}Si\equiv + 2H^+ \leftrightarrow 2(\equiv Si{-}O{-}H) + Pb^{2+}. \quad (1)$$

The rate of this reaction is dependent on the lead content and the mass transport of lead and proton through Pb-depleted hydrated silica layer on glass surface [6–8]. The mass transport is reduced in presence of small amount of higher field strength cations such as Mg^{2+}, Ca^{2+}, Al^{3+}, Ti^{4+}, and Zr^{4+} in glass composition containing other modifiers [9–13].

Secondly, by taking place of the ion-exchange reaction which results in the production of Pb-depleted layer, the water diffusion and the water surface contact with sample are increased. Therefore, the water molecules penetrate through this layer and react with silica network by the hydrolysis reaction (2):

$$\equiv Si{-}O{-}Si\equiv + H_2O \leftrightarrow 2(\equiv Si{-}O{-}H). \quad (2)$$

This reaction, in more open silica network, occurs at a faster rate, breaks down the Si–O–Si bonds and leads to glass dissolution.

On the other hand, in physical point of view, the high amount of lead in these glasses makes them highly effective for attenuation of gamma-rays due to high atomic number and density of lead. Therefore, for radiation protection applications, investigation on determination of gamma-ray attenuation coefficients of these glasses is very important.

In this study, the effect of TiO_2 and ZrO_2 additives and the ratio of (Pb + Na + K)/Si of composition on chemical corrosion of LSGs samples were investigated. Also the density, glass transition temperature (Tg) and gamma-ray attenuation coefficients of these glasses were determined.

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