Physico-chemistry of ZrO2 in molten silicates: influence of the melt composition

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Abstract

Due to its refractory properties and to its well-known low solubility in molten silicates, zirconia ZrO2 is often used as a component in refractory bricks and crucibles in high temperatures applications involving contact with molten glass. This work proposes to study the interactions between zirconia and a multi-component soda-lime glass at temperatures comprised between 1200∘C and 1400∘C for times up to 100 h. SEM observation allowed to control the oversaturation of the samples and led to highlight phenomena as coalescence and phase precipitation. In the first minutes of contact, precipitation of zircon ZrSiO4 was detected. Consequently, a similar study was performed on zircon directly in contact with the melt. The zirconium solubility was measured by EPMA (WDS), leading to dissolution kinetics. After a rapid increase, a stabilization of zirconium concentration is observed. Then, the precipitation of more stable zircon leads to a solubility decrease. Long term experiments have shown significant decrease of the solubility, attributed here to the volatilization of sodium from the melt, which consequently implies a change in its composition. Limits of solubility could thus be determined by choosing a compromised time of 2 hours. The influence of the main glass components on zirconium dissolution at 1200∘C was also evaluated via enrichment of the basic composition with 5 and 10 wt.% of network modifiers (*i.e.* Na2O and CaO), a network former (*i.e.* B2O3) and an intermediate (*i.e.* Al2O3). Additions of Na2O and CaO led to the precipitation of new phases in equilibrium with the melt which were Na2ZrSi2O7and Ca2ZrSi4O12 respectively. No phase precipitation was observed when B2O3 or Al2O3 were added. Addition of network former B2O3 did not seem to influence zirconium solubility. However, zirconium solubility decreases significantly in the case of Al2O3 addition, as network forming aluminates [AlO45-] requires charge compensation by Na+ cations.

Keywords: zirconia, molten silicates, dissolution

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