Received: 24 December 2022 | Revised: 21 February 2023 | Accepted: 7 March 2023

DOI: 10.1111/jace.19099

Journal of the American Ceramic Society

RESEARCH ARTICLE

Effect of small additions of Li_2O on phase separation and crystallization of barium-silicate glasses

Takato Kajihara[1,2] | **Kosho Akatsuka**[1] | **Hiroyuki Hijiya**[1] | **Satoshi Yoshida**[1] | **Aran Mitsuta**[3] | **Hongye Gao**[4] | **Shigeru Fujino**[5] | **Satoshi Hata**[4,6]

[1]AGC Inc, Materials Integration Laboratories, Kanagawa, Japan

[2]Interdisciplinary Graduate School of Engineering Sciences, Kyushu University, Fukuoka, Japan

[3]Department of Energy Science and Engineering, School of Engineering, Kyushu University, Fukuoka, Japan

[4]The Ultramicroscopy Research Center, Kyushu University, Fukuoka, Japan

[5]Global Innovation Center, Kyushu University, Fukuoka, Japan

[6]Faculty of Engineering Sciences, Kyushu University, Fukuoka, Japan

Correspondence
Takato Kajihara, AGC Inc, Materials Integration Laboratories, Kanagawa, Japan.
Email: takato.kajihara@agc.com

Funding information
Ministry of Education, Culture, Sports, Science and Technology (MEXT), Grant/Award Number: JPMXP1222KU0102

Abstract

It is known that the addition of Li_2O to 33.3BaO-66.7SiO_2 glass, whose composition is the same as $BaSi_2O_5$, promotes crystallization of $BaSi_2O_5$. In this study, in order to clarify the effect of a smaller amount of Li_2O, xLi_2O-(30-x)BaO-70SiO_2[mol%] ($x = 0$, 0.2, 0.5) glasses were prepared. The main crystalline phases in the heat treatments near the maximum crystallization peak temperature, were high-$BaSi_2O_5$ and low-$BaSi_2O_5$ which transformed from high-$BaSi_2O_5$. It is found that the introduction of only 0.2 mol% and 0.5 mol% Li_2O significantly changes the crystallization behavior. In the composition without Li_2O, only high-$BaSi_2O_5$ was formed after heat treatment even for 24 h. For compositions containing Li_2O, low-$BaSi_2O_5$ was formed within 1 h of heat treatment. In these compositions, it is found that the addition of Li_2O enhances phase separation in the early stage of heat treatment, resulting in the formation of Si-rich droplet phases and Ba-rich phases. The composition of the Ba rich glass phase would be close to the stoichiometric composition of $BaSi_2O_5$, suggesting a significant change in crystallization behavior.

KEYWORDS
differential scanning calorimetry, glass ceramics, Li_2O-BaO-SiO_2 glasses, phase separation, Raman spectroscopy, transmission electron microscopy

1 | INTRODUCTION

Glass-ceramics has both glass and crystalline properties, and this is an excellent material in terms of mechanical properties, optical properties, thermophysical properties, and the like. In order to control these properties, it is important to understand effects of glass compositions on the crystallization behavior. For example, Zanotto et al. reported that surface crystallization occurs in many compositions of silicate glass,[1] but 33.3BaO-66.7SiO_2 [mol%] (BaO-2SiO_2: BS2) is the glass compositions in which internal crystal nucleation occurs without any nucleating agent.[2–6] Furthermore, this composition has a high nucleation rate despite their high glass-forming ability.[1,7] Due to this characteristic property, various studies have been conducted to investigate the crystallization mechanism of this glass system. This binary composition is known to have a metastable immiscible region where a phase separation phenomenon occurs below the liquidus temperature when the BaO content is approximately 32 mol% or less.[8–10] For example, the formation of Ba-rich droplets has been observed for BaO from 1 mol% to 6 mol%, and a two-phase entangled structure associated with spinodal decomposition has been observed for BaO from 8 mol% to 12 mol%. Si-rich droplets formation has been observed for BaO between 14 mol% and 30 mol%. In the

