Formation of Cs-compounds by chemical interaction of CsOH and concrete at various temperatures

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To investigate the source of high dose rates at the concrete shield plugs of Unit 2 and 3 at 1F, high temperature tests on the mixture of CsOH and pulverized concrete/main components were conducted. Results showed that both water-soluble and -insoluble phases were formed below 300°C. Namely, Cs$_2$CO$_3$(H$_2$O)$_2$ was formed due to chemical reaction with CaCO$_3$ at room temperature. The authors will discuss the possibility that this might be one of the main trapping mechanisms on shield plugs.

Keywords: Cesium, chemical interaction, CsAlSiO$_4$, concrete shield plug, Fukushima

1. Introduction

High radioactivity (>20PBq) were recently detected in the concrete shield plugs of Units 2 and 3 at the Fukushima Daiichi Nuclear Power Plant (1F) [1], this suggests some trapping effects of released radioactive materials e.g., CsOH and CsI, on concrete surface at high temperatures during severe accident. However, most of current studies on radionuclide contamination mechanisms of concrete are focusing on Cs migration in cementitious materials through aqueous phase, and the study of chemical interaction of Cs and concrete at high temperatures are very scarce. This study aimed to clarify the interaction temperature and the formation of Cs compound by the chemical interaction of CsOH, which is expected to be one of the main radioactive gas or aerosol species, and concrete/main components for understanding the cesium trapping mechanism for the shield plugs.

2. Experimental method

For sample preparation, CsOH.H$_2$O (Sigma Aldrich, 99.5%) was mixed with pulverized concrete (water/cement ratio of 0.55) / its main components i.e., rock-forming aggregate, natural feldspar, natural quartz, amorphous SiO$_2$, and CaCO$_3$. The mixture was then compressed into pellets of size 4mm in diameter under 200MPa. The thermal events of mixed samples were analyzed by using thermogravimetric instrument (Thermo plus EVO2/TG- DTA 8121, Rigaku) under argon gas at a constant flow rate of 200 ml/min and a heating rate of 20 °C/min. To determine the formation of Cs-compounds, mixed samples were heated in the constant argon flow at the same heating rate reaching up to 800°C in maximum, and held for 30min using electric furnace.

3. Results and Discussion

As seen in Fig.1(a), the exothermic peaks were observed in 30wt% CsOH.H$_2$O mixed concrete, aggregate and natural quartz (SiO$_2$) at the temperature range of 100°C-220°C. This suggests that the interaction temperature for CsOH and concrete might occur within this range. Actually, Cs-Al-Si-O phase i.e., CsAlSiO$_4$ was formed due to chemical interaction with aluminosilicate minerals (e.g., KAlSi$_3$O$_8$ and NaAlSi$_3$O$_8$) containing in aggregates from 200°C (as confirmed by XRD, Fig.1(b)), this is much lower than previously reported temperature in similar materials e.g., insulator [2] and soils [3] above 600°C. Furthermore, glass-like product was formed by the interaction with SiO$_2$ above 200°C. Notably, chemical reaction between CsOH and CaCO$_3$ existing in concrete occurred even at room temperature (RT) to form Cs$_2$CO$_3$ and following adsorption of moisture to become Cs$_2$CO$_3$(H$_2$O)$_2$ (Fig.1(b)), this might be one of the main trapping mechanisms of Cs on concrete structure because CaCO$_3$ is commonly formed on concrete surface due to carbonation and the reaction with CsOH might occur at RT.

References


Fig.1. (a) DTA curves of unmixed (dashed curves) and 30wt% CsOH.H$_2$O mixed samples (continuous curves); (b) XRD patterns of representative 30wt% CsOH.H$_2$O mixed concrete heated up to 800°C.