9A/2010C014

XAFS Analysis of the Structure of Ca in Engineered Barrier Materials (2)

Hiroyuki SAKAMOTO[1*], Kazuko HAGA[1], Hitoshi OWADA[2], Tomoko ISHII[2]
Junichi NISHINO[3], Hiroaki NITANI[3], Masaharu NOMURA[3]
[1]Taiheiyo Consultant Co., Ltd., 2-27-8, Higashinihonbashi, Chuo-ku, Tokyo, 103-0004, Japan
[2]RWMC, 1-15-7, Tsukishima, Chuo-ku, Tokyo, 104-0052, Japan
[3]KEK-PF, Tsukuba, Ibaraki 305-0801, Japan

Introduction

Generation of C-S-H at the boundary of the cementitious materials and bentonite due to their interaction is an important phenomenon to evaluate for more accurate prediction of long-term behaviors of the engineered barriers proposed for the geological disposal of radioactive waste. The C-S-H among secondary minerals had been difficult to analyze, however, the study to date demonstrated quantitative analysis of the C-S-H by using the XAFS measurement. In this study, generation of the C-S-H was analyzed for two types of bentonite aiming at identifying differences in its behavior depending on the difference of the type of the bentonite.

Experiment

Samples

Two types of bentonite were used; Kunigel V1 (Yamagata, Japan) and MX80 (California, USA). The compacted bentonite samples with a dry density of 1.6 g/cm^3 were contacted with ordinary portland cement paste (water to cement ratio of 0.6). The cement-bentonite contacted specimens were immersed in artificial ground water for six years. XAFS analysis specimens were taken from the bentonite part of the immersed specimens at varied distances from the cement-bentonite boundary.

XAFS Measurements

In order to evaluate C-S-H generation at the boundary of the cement and bentonite, Ca *K*-edge (4.04 keV) XAFS measurements were carried out by using BL-9A beamline with a Si(111) double crystal monochromator at the KEK Photon Factory. The measurements were carried out by using transmission mode with ionization chambers. Identification of Ca-containing primary and secondary minerals were carried out by using the pattern fitting with Ca-K XANES spectra to those of standard minerals.

Results and Discussion

According to the pattern fitting analysis of Ca-K XANES spectra, the secondary minerals generated in the bentonite phase by the contact with the cement were C-S-H and calcite. Kunigel V1 had more C-S-H species than MX80. Quantity of the C-S-H tends to decrease with the increase of distance from the boundary with the cement. Compared with Kunigel V1, generation of the C-S-H in MX80 concentrated near the boundary than other regions.

XAFS analysis indicated different quantities and distributions of the C-S-H generation in the bentonite near the boundary depending on the type of the bentonite. Diffusion coefficient of radionuclide estimated by effective montmorillonite density [1] is equal in Kunigel V1 and MX80. Additionally, dissolution rate in montmorillonite is equal in Kunigel V1 and MX80. On the other hand, Calcite amount is different from Kunigel V1 in MX80. Therefore, it is thought that difference of the C-S-H generation is affected by accessory minerals such as Calcedoni, Pragioclace. Study on relations between the C-S-H generation conditions and properties of the bentonite could contribute to achieve more accurate prediction of long-term interaction between bentonite and cementitious materials.

Table I. The weight proportion of Ca of altered samples immersed into artificial groundwater

Type of bentonite	Distance from the boundary (mm)	Results by using XAFS (The mass proportion of the mineral containing Ca to total Ca in the each zone, unit:%)					Ca content of solid phase by using EPMA(%)
		C-S-H	Ca-montmorillonite	Calcite	Dolomite	Plagioclases	
Kunigel V1	0~1	66.4	31.4	2.0	0.2	0.0	5.7
	1~2	65.4	30.5	0.5	3.5	0.0	4.5
	2~3	64.5	18.8	6.2	5.3	5.1	3.5
	4~5	61.1	20.7	6.3	6.1	5.8	3.2
	12~13	33.2	25.3	13.9	15.1	12.5	1.6
MX80	0~1	43.4	48.4	1.3	0.0	7.0	2.3
	1~2	20.7	59.2	11.1	0.1	8.9	1.6
	2~3	23.0	51.2	7.9	1.5	16.4	1.7
	4~5	19.4	57.7	11.4	0.8	10.7	1.5
	12~13	8.3	69.2	13.3	2.4	6.8	1.1

Determined by calculation with REX2000 Ver. 2.5.9

Acknowledgments

This research is a part of the "Evaluation Experiments of Long-Term Performance of Engineered Barriers" under a grant from the Ministry of Economy, Trade and Industry (METI).

References

[1] Japan Atomic Energy Agency, The Federation of Electric Power Companies of Japan: Second Progress Report on Research and Development for TRU Waste Disposal in Japan, JNC TY1400 2005-013 / FEPC TRU-TR2-2005-02(2005).

*Hiroyuki_Sakamoto@grp.taiheiyo-cement.co.jp