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Biogeochemical changes at early stage after the closure of radioactive waste geological repository in South Korea

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ABSTRACT

Permanent disposal of low- and intermediate-level radioactive wastes in the subterranean environment has been the preferred method of many countries, including Korea. A safety issue after the closure of a geological repository is that biodegradation of organic materials due to microbial activities generates gases that lead to overpressure of the waste containers in the repository and its disintegration with the release of radionuclides. As part of an ongoing large-scale in situ experiment using organic wastes and groundwater to simulate geological radioactive waste repository conditions, we investigated the geochemical alteration and microbial activities at an early stage (~63 days) intended to be representative of the initial period after repository closure. The increased numbers of both aerobes and facultative anaerobes in waste effluents indicate that oxygen content could be the most significant parameter to control biogeochemical conditions at very early periods of reaction (<35 days). Accordingly, the values of dissolved oxygen and redox potential were decreased. The activation of anaerobes after 35 days was supported by the increased concentration to ~50 mg L^{-1} of ethanol. These results suggest that the biogeochemical conditions were rapidly altered to more reducing and anaerobic conditions within the initial 2 months after repository closure. Although no gases were detected during the study, activated anaerobic microbes will play more important role in gas generation over the long term.

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1. Introduction

A major environmental concern in radioactive waste management is the potential release and transport of radionuclides to the biosphere after the closure of underground radioactive waste disposal facilities. Scientists and engineers have been considering numerous scenarios regarding the assessment of the performance of geological repository over a very long time. A starting point of the scenarios is that the repository may be saturated by local groundwater soon after facility closure. This could lead to changes in background geochemical conditions such as pH and oxidation–reduction potential (ORP) in the geological repository. In addition, intrusion of groundwater to the repository could introduce microbial activities, because a variety of indigenous microorganisms are likely present in both groundwater and the wastes as well as organisms introduced during the repository construction and operation phases.

One of the main safety issues in the repository is potential gas generation by microbial activities under saturated groundwater conditions. The generated gas builds up pressure in the repository, which may result in the loss of integrity of the waste containers and the repository, and facilitate radionuclide transport through the groundwater system (Small et al., 2008; Stroes-Gascoyne and West, 1996; Wang and Francis, 2005). The gas can be produced by the increased corrosion of metallic wastes, drums, and/or containers, and organic waste degradation processes by microbial activities. The expected predominant gases are hydrogen (H_2), carbon dioxide (CO_2), and methane (CH_4) (Gillow and Francis, 2011; Small et al., 2008). In particular, low- and Intermediate-Level Wastes (LILWs) could be important for gas production because of

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the high proportion of organic elements that are biodegradable (Gillow and Francis, 2011).

More than 9,00,000 drums (200 L/drum) of LILWs were generated by Korean nuclear facilities, including 23 nuclear power plants, until 2012 (https://www.oecd-nea.org/rwm/profiles). A rock cavern type geological repository (i.e., silo disposal method) has been constructed at Gyeongju, Korea, to dispose of 1,00,000 LILW drums as a first stage of the waste disposal process (Fig. 1). According to the Korea Radioactive Waste Management Corporation (now, Korea Radioactive Waste Agency [KORAD]) (KRMC, 2008), approximately 84% of dry active wastes, which contained 69% of the LILWs generated by the nuclear facilities in Korea, consist of combustible wastes (mostly, organic materials) including rubber, cotton, wood, vinyl, plastic, paper, and activated carbon (Park et al., 2012). The performance of the Gyeongju LILW disposal facilities will be affected by microbial activities, because of the potential intrusion of groundwater to the repository after closure. Therefore, investigations of the biogeochemical effects must be conducted under in situ conditions to evaluate the safety of the Gyeongju repository performance.

Large-scale in situ experiments are under way using simulated LILW package drums placed in a basic disposal concrete container at the Gyeongju repository (Fig. 1). The experimental setup was benchmarked by Finnish experiments because their disposal method is similar to the Korean method (Park et al., 2010). The goal of this study, in part, is to determine the biogeochemical changes brought about by the activities of microorganisms at a comparatively early period after closure of the repository.

2. Materials and methods

2.1. Large-scale in situ experiments

The study area is at the Gyeongju geological repository site in the coastal area of southeastern Korea (Fig. 1), where the bedrock consists of mainly the Cretaceous granite (Choi et al., 2008). Clays (i.e., montmorillonite, zeolite, chlorite, and illite) are frequently observed as secondary minerals in the fractured zone, but not for calcite, and pyrite is widely scattered throughout local geology (Choi et al., 2008).

A large-scale concrete container was installed in the cavern 130 m below sea level in the Gyeongju geological repository. Nine 320-L steel drums were prepared by repackaging 200-L drums compressed under super-high pressure (Park et al., 2012), and were placed in the concrete container (collectively called a 9-Pack). Organic wastes such as vinyl, plastic, and cotton were included in the 320-L drums of the 9-Pack (Fig. 1). Groundwater collected from the Gyeongju geological repository was used to fill the 9-Pack to simulate the saturated environments expected in the LILW repository after its closure.

Two-liter sterile polypropylene containers were used to collect the groundwater, and water samples from the 9-Pack for geochemical and microbial analyses. The groundwater was overfilled into a sampling container, and then it was immediately sealed by a screw cap without headspace to minimize air contact with the water during sampling procedures. Geochemical and microbiological properties in the groundwater were investigated. After the large-scale in situ experiment began, the 9-Pack water samples, which were collected at 7, 21, and 35 days, were used for assessment of geochemical alteration. The types of microorganisms in the 9-Pack water samples collected at 7 and 63 days were determined in order to compare them with the microbes found in the initial groundwater sample. All the water samples including the groundwater sample were stored at 4 °C before the analyses. The generated gases were collected into the Tedlar® gas sampling bags of the online system installed on the 9-Pack container during 63 days.

2.2. Geochemical analyses

Water-quality parameters such as pH, oxidation reduction potential (ORP), dissolved oxygen (DO), and electrical conductivity were measured using multi-probes (Orion 5-Star™ meters; Thermo Scientific Co.). All water samples were filtered using a 0.45-μm polyvinylidene fluoride (PVDF) syringe filter before the chemical analyses. The concentration of anions (F^-, Br^-, Cl^-, NO_3^-, SO_4^{2-}, and PO_4^{3-}) was analyzed using a DX-60 Ion Chromatograph with

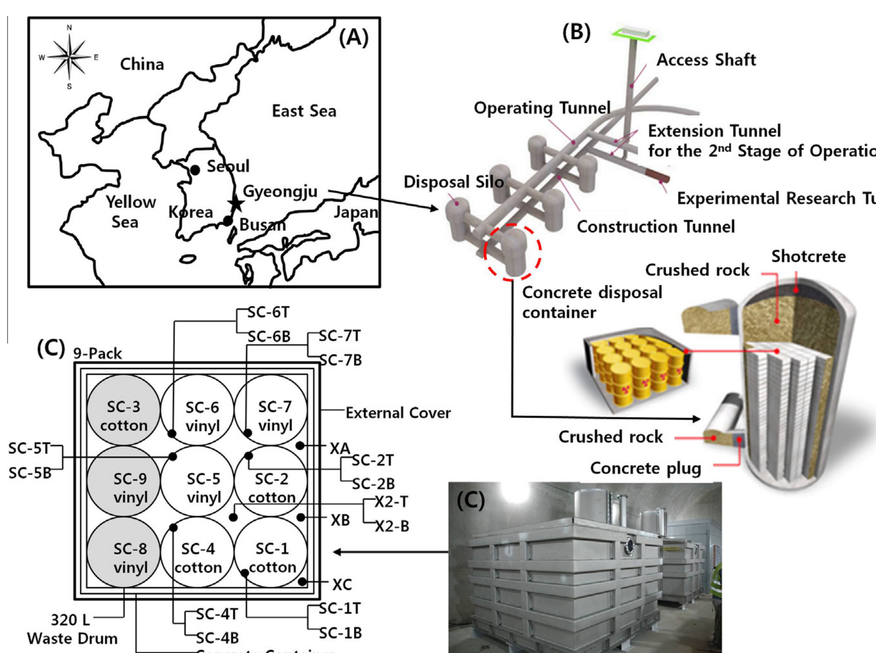

Fig. 1. (A) Location of the LILW disposal site in Korea, (B) schematic layout of disposal silos and unit concrete container and (C) experimental setup (9-Pack) for this study. In the layout of the 9-Pack, the solid points indicate individual sampling locations with sample name and position (top [T] or bottom [B]). The grey-colored waste drums were not used for sampling procedures.