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Microbial Gas Generation and Implications of Low- and Intermediate- Level Radioactive Waste (L/ILW) Disposal in Korea

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1. Introduction

Low- and intermediate- level radioactive wastes (L/ILW) generated from nuclear power plants, industries, hospitals, and research institutions contain a variety of radionuclides and a large fraction of biodegradable organic materials such as contaminated paper, rubber gloves, shoes, cotton, spent resins, cartridge filters, etc. These wastes are typically packaged in 55- gallon steel drums and stored or disposed of in the subsurface facility. Degradation of organic matter by microbes in L/ILW and corrosion of steel drums can generate gas which has the potential to compromise the integrity of waste drums and release the radionuclides and other contaminants into the environment. Microbial activity for gas generation can be achieved by a series of microbial processes involving enzymatic oxidation-reduction reactions by various species of microbes. Microbial activity is limited by the availability of electron donors and acceptors, moisture, and nutrients (specifically nitrogen); and affected by temperature, pH, Eh, radiation and toxic compounds. Here we review the gas generation studies conducted at L/ILW in Finland, USA, Sweden, and Switzerland. We assess the microbial gas generation from L/ILW to be disposed of at Korea and the implication on the radionuclide mobility.

2. Information of L/ILW

Low- and intermediate- level radioactive wastes produced in Korea by industry, hospitals, and nuclear power-plants (NPPs), contain organic materials with short-lived radionuclides, whereas intermediate-level radioactive waste contains higher concentrations of radionuclides, and these wastes are packaged in steel drums or containers' The backfill materials used are cement, bentonite or crushed rock as in the case of Korea. The wastes will be disposed of in underground silos (25 m width and 50 m height) at 130 m below sea level at the Wolsong L/ILW disposal facility under construction at Bonggil-ri, Yangbuk-myeon, Gyeongju. The composition of L/ILW that will be stored in Wolsong repository site is summarized in Table. 1 [1].

3. Site environment

The rock composition of Wolsong site consists of trachyte, rhyolite, granite, granodiorite which has igneous characteristics, and ion concentration of groundwater in Wolsong site is dominated by Ca, Na, Cl_2, SO_4 and HCO_3 which can affect pH [1]. The values of DO and Eh decreased with depth, suggesting that anaerobic and reducing condition existed in deep groundwater, and the concentration of Fe and Mn was high, indicating that redox condition in groundwater was controlled by these oxides. Although there is some indirect evidence of microbial activity but no systematic studies were undertaken to determine microbiological characteristics of this site.

Table 1: Type and quantity of L/ILW

Source	Quantity (drums)	Type
Nuclear power-plants	~ 70,000	Miscellaneous radioactive solid waste, concentrates, spent resins, cartridge filters
Research Institutes	~ 16,300	Same as NPPs included decommissioning wastes
Nuclear fuel manufacturi ng facilities	~ 5,600	Metal, wood, lime deposit, glass, concrete, composite, etc.
Radioisotop es	~ 6,100	Unsealed source (combustibles, incombustibles, organic or inorganic matters, etc.), sealed source.

4. Microbial activity

Microorganisms have been detected in L/ILW sites, deep geological formations, and waste repository sites designated for disposal of high level waste. Indigenous microorganisms and those present in L/ILW can metabolize organic compounds from L/ILW as an energy source aerobically and under anaerobic conditions use nitrate and sulfate as an electron acceptors. Such bacteria can produce H_2, N_2, CO_2, and CH_4 gas as a result of anaerobic respiration. The sequence of metabolic pathway and the reaction mechanisms catalyzed by microorganisms that can be found in a repository site is summarized in Table. 2 [2].

Table 2: Chemical reactions catalyzed by microorganisms

Process	Reactions
Cellulose degradation	$[C_6H_{10}O_5]n + nH_2O \rightarrow nC_6H_{12}O_6$
Aerobic degradation	$C_6H_{12}O_6 + 6O_2 \rightarrow 6\mathbf{CO_2} + 6H_2O$
Nitrate reduction [Denitrification]	$5C_6H_{12}O_6 + 24HNO_3 \rightarrow 30\mathbf{CO_2} + 12\mathbf{N_2} + 42H_2O$
Anaerobic [Fermentation]	$C_6H_{12}O_6 + H_2O \rightarrow 2\mathbf{CO_2} + 2\mathbf{H_2} + C_2H_5OH + CH_3COOH$
Anaerobic acetogenesis	$C_2H_5OH + H_2O \rightarrow CH_3COOH + 2\mathbf{H_2}$
Sulphate reduction [Acetoclastic]	$CH_3COOH + H_2SO_4 \rightarrow \mathbf{H_2S} + 2\mathbf{CO_2} + 2H_2O$
Sulphate reduction [H_2-utilizing]	$4H_2 + H_2SO_4 \rightarrow \mathbf{H_2S} + 4H_2O$
Methanogenesis [Acetoclastic]	$CH_3COOH \rightarrow \mathbf{CO_2} + \mathbf{CH_4}$
Methanogenesis [H_2-utilizing]	$CO_2 + 4H_2 \rightarrow \mathbf{CH_4} + 2H_2O$

Table 3: Summary of Gas generation studies at various sites

Site	Study	Experimental and modeling results
Olkiluoto Finland	Experimental & Modeling	H_2 is generated due to metal corrosion, and H_2, CH_4, CO_2, O_2, N_2 are generated due to microbial activity
WIPP U.S.A	Experimental	H_2, CH_4, CO_2, O_2, N_2, N_2O are generated due to microbial activity
SFL 3-5, Sweden	Modeling	H_2 is generated due to metal corrosion, and H_2, CH_4, CO_2 are generated due to microbial activity
Opalinus clay, Swiss	Modeling	H_2 is generated due to metal corrosion, and CH_4, CO_2 are generated due to microbial activity

5. Case studies

A large-scale gas generation experiment was conducted at Olkiluoto, Finland for 9 years to determine the microbial gas generation from LLW in waste drums disposed of in the operational VLJ repository [3]. Gas was generated in association with the degradation processes with the potential to overpressure the repository, which can promote transport of groundwater and gas and consequently radionuclides. In another study, at Waste Isolation Pilot Plant (WIPP), U.S.A. gas generation with simulated waste was investigated under aerobic and anaerobic humid and inundated conditions in the presence and absence of nutrients for ~11 years [4]. The main purpose of the experiment at WIPP was determine the rate and extent of gas generated from the microbial degradation of organic constituents of transuranic (TRU) waste under conditions expected in the WIPP site. Modeling of gas generation has been reported from L/ILW in Sweden, Switzerland and Germany. The experimental and modeling results at several sites are summarized in Table. 3 [3, 4, 5, 6].

6. Conclusions

Microorganisms in L/ILW play an important role in degradation of organic materials, corrosion of steels and generate gas. Long-term experimental and modeling studies conducted with simulated wastes in Finland and USA confirm that various gases produced by microorganisms can affect the performance of the waste repository sites. Consequently mitigative measures have been undertaken or proposed to sequester CO_2 produced. Because of the waste heterogeneity and the site specific environmental conditions, it is difficult to predict rate and extent of microbial gas generation. However, it is suggested that gas generation experiments should include actual waste forms and reflect the local site characteristics and the environmental conditions.

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