氏　　名	青木　和弘
学位の種類	博士（理学）
学位記番号	博甲第１０４４号
学位授与の日付	平成20年3月22日
学位授与の要件	課程博士(学位規則第４条第１項)
学位授与の題目	Microbial activity and material transport of deep groundwater in geological disposal of high-level radioactive wastes （放射性廃棄物処分における深部地下水中の微生物挙動と物質の動態に関する研究）
論文審査委員（主査）	田崎　和江（自然科学研究科・教授）
論文審査委員（副査）	奥野　正幸（自然科学研究科・教授），加藤　道雄（自然科学研究科・教授）， 中西　孝（自然科学研究科・教授），神谷　隆宏（自然科学研究科・教授）

Abstract

Comparative analysis was conducted of groundwater samples taken from boreholes drilled in ten different geological environments throughout Japan. Next, microbial analysis was conducted on samples taken from the Kamaishi Mine. Six groundwater samples were taken from two boreholes from the mine drift in granodiorite, and core samples from bentonite used in the thermo-hydro-mechanical experiment conducted in the drift. Likewise, microbial analysis of groundwater samples taken from boreholes in the underground research laboratory located in Horonobe, Hokkaido enumerated a total number of bacteria in the range of 1.2×10^4 cells/ml to 1.5×10^7 cells/ml.

The above results clearly indicate that microbial activity is universal even in what appears to an anaerobic environment deep underground as long as a certain level of water content, water temperature, reducing conditions and nutritional aspects are found to satisfy the minimum living conditions. In order to analyze the potential microbial impact on the geo-scientific characteristics of the groundwater, using deep groundwater samples from various rocks and different depths collected from throughout Japan, simulation analysis was conducted using the numerical code MINT (Microbial Influence on Nuclide Transport). This code so far has proven to give reliable enumerations for sulphate ion concentration and number of sulphate reducing bacteria, matching well with actual measurements.

Chapter II　Microbial analyses from various geological locations

As part of a series of research conducted to ascertain the existence and extent of microbial activity deep underground, this study analyzed data collected for groundwater and surface water samples taken from eleven boreholes located throughout Japan at depths ranging from 222 to 535 meters below ground level and of varying rock types and age: granite, basalt, ryholite, schist, limestone and sedimentary rocks from Neogene Tertiary to Mesozoic and Paleozoic age. All samples underwent chemical analysis and the results were submitted for qualitative evaluation using a geochemical numerical code. For ten of these samples, microbial analysis was also

conducted, enumerating the total number of bacteria and viable cell counts using conventional culture methods (Figure 1).

The present study confirmed the existence of microorganisms underground as deep as 435 meters below ground level, under the natural state of conditions virtually free of (even before having any) contact with the surface, under expected environmental conditions (i.e., Eh, pH, temperature, pressure, nutrient and energy availability, etc.) and throughout Japan. Further examination in other locations and under other conditions is most likely to reveal the same result. The next step, then, will be to ascertain the size of the microbial population and hence the extent of its activity and impact on the underground structures.

Chapter III Microbial survey at Kamaishi mine

An important part of the near field Performance Assessment of nuclear waste disposal is to evaluate the coupled thermo-hydro-mechanical (T-H-M) phenomena. They include thermal effects on groundwater flow through rock matrix and water seepage into the buffer material, and generation of the swelling pressure of the buffer material and thermal stresses potentially affecting the change in the porosity and fracture aperture of the rock. This chapter compares groundwater samples taken from the surrounding host rock of granodiorite and the bentonite buffer material used for the coupled thermo-hydro-mechanical (T-H-M) experiments at the Kamaishi Mine in Iwate. Analysis is made on microbial activities of the groundwater as well as on the viable heterotrophs and water content of bentonite. This chapter focuses on assessing the survival of the naturally present microbial population in bentonite that has been subjected to compaction upon emplacement and to the subsequent heating and cooling cycles during the T-H-M experiments. Bacterial activity in bentonite buffer materials adjacent to the container could potentially affect microbially influenced corrosion (MIC) and radionuclide migration, but is expected to be limited because of the high temperatures, desiccation and radiation fields associated with the waste container.

The microbial analysis of the T-H-M experiment has shown that the water content in the buffer material is the dominant crucial factor for the survival and activity of viable microbial population (Figure 2). It was found that water content in itself depends on the emission of heat from the waste container. The results from this study suggest that the part of the buffer directly adjacent to the container would be virtually devoid of microbial activity because of the redistribution of the initial water content resulting from high temperature. Assessment of whether microorganisms found in the Kamaishi Mine are aerobic or anaerobic, initially present or repopulated, would require a more expanded analysis incorporating chemical data. A comprehensive T-H-M plus Chemical-Biological study will permit more detailed identification of the microorganisms present and their activity under a variety of conditions which could exist in an actual vault.

Chapter IV Microbial survey at Horonobe Underground Research Center

The Horonobe URL project is devoted to the technological development for a safe geological disposal of high level radioactive wastes over a 20-year period from March 2001 in Horonobe-cho in northern Hokkaido, Japan. This chapter assesses the chemical and microbial analysis conducted on groundwater samples taken from boreholes at the site of the Horonobe Underground Research Laboratory (URL) project. As it was necessary to use drilling mud for excavating the soft sedimentary rock of Neogene Tertiary age, tracer was used to evaluate its influence on the quantitative analysis for possible contamination by the drilling mud.

According to the result of chemical assay and microbial analysis, groundwater is meteoric origin at the shallower part, while at deeper part it is considered mixture of sea water and meteoric water. The boundary of both meteoric and sea water may be around at the 250m-280m for HDB-6 and somewhere deeper than 185m for HDB-8. From the total number of bacteria in the groundwater sample and the tracer concentration, the total number of bacteria in the original groundwater environment is estimated to be in the order of 10^3 to 10^4 cells/ml. The dominance of aerobic heterotroph in the usually anaerobic environment of groundwater infers the effect of contamination in which the water from the well is mostly liked to have mixed into the groundwater. The measurement results of redox potential and confirmation of the existence of nitric acid ion and ammonia ion in the groundwater sample lead to the assumption that the most dominant microorganisms are nitrate reducing bacteria and denitrifying bacteria (Figure 3). Nitrogen cycle can be summarized in Figure 65.

Chapter V Simulation

This chapter aims to predict microbial effects by analyzing simulation results obtained by using the numerical code MINT (Microbial Influence on Nuclide Transport) with input taken from data obtained in Chapter II. The code can model biochemistry and geochemical equilibrium with consideration to solute transport and microbial activity. The goal of the simulation is to develop a model to predict microbial effects on the performance of a high-level radioactive waste repository. It can describe the consumption of organic compounds, generation of reductive environments by consumption of electron acceptors and passive transport of microorganisms suspended in pore water.

Several potential factors that may trigger disturbance to important parameters of Performance Assessment have been proven. One is the corrosion of the drilling rig (H_2 concentration was extremely high singularly for the Kamioka sample). Another is air pollution (high concentration of O_2 was obvious in many samples) as well as dissolution of minerals from the drilling fluid (especially carbonate mineral). Signs of low pH and increased CO_2 production (possibility of microbial interaction/intervention) also are factors to be considered. Analysis of chemical reaction accompanying microbial activity

showed unique characteristics for different microbial activity by site and by varying analytical hypothesis. It is, therefore, important to conduct further analysis of the chemical reaction on mineral surface.

Chapter VI Conclusion

—Geochemical data interpretation to support Performance Assessment—

This chapter examines the importance of gathering geochemical data and how to interpret them in order to actually apply the results in performance assessment. Performance Assessment uses knowledge of natural in-situ chemical conditions. These chemical conditions must be estimated from analytical data that are obtained from site investigations. Usually, these data are obtained from boreholes drilled from the surface and from underground excavations, such as an Underground Research Laboratory.

The quality of any "raw" geochemical data (data obtained directly from site investigations) should be evaluated thoroughly before it is used in any performance assessment. Where determinants are shown to have been perturbed from in-situ conditions, they must be corrected for perturbations; or, if this is not possible, the data need to be rejected and not used. In some cases, it may be necessary to assume a concentration of a determinant, if there is good evidence from the literature or experimental investigations.

学位論文審査結果の要旨

本学位論文に関して、平成19年12月10日に予備審査会を行い、学位論文として提出することを認めた。第一次審査は書面で行った。さらに、平成20年2月5日に行われた口頭発表の後に審査会を開き、協議の結果、以下のように判定した。

本論文は、天然バリア中の深部地下水の微生物調査および人工バリアの構成要素であるベントナイト中での微生物分析結果について論じた。これまで陸域の深部地下水中に存在する微生物に関する報告はほとんどなかった。地質環境が異なる全国１０ヶ所で掘削された試錐孔および幌延深地層研究施設における２孔の試錐孔から採取した地下水を対象に全菌数および特定細菌の生菌数計数を行った。全菌数は、3.2×10^{1} cells/ml から 1.5×10^{7} cells/ml の計数結果が得られた。生菌数としては、前者では硫酸塩還元菌が９ヶ所で、メタン生成菌が３ヶ所で確認された。幌延では硝酸塩還元菌および脱窒菌が卓越していることが確認された。岩手県釜石鉱山の坑道内で実施された熱ー水ー応力連成試験のベントナイト中の従属栄養細菌を分析した結果、水分量が１２％以下では生息しないことが判明した。これらは，極限環境とみなされ微生物活動は乏しいと考えられてきた陸域の地下深部でも、水分量、水温、酸化還元状態、栄養物等の生息条件を満たしていれば微生物活動が普遍的であることを明らかにした日本におけるパイオニア的研究であり、陸域の深部地下水に関する微生物研究に大きく貢献した。以上の研究成果は、博士（理学）に値すると判断した。