

Role of redox reaction for long-term barrier function of geological environment

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Redox fronts are created at the boundary between two rock/groundwater systems with different oxidation environments. The development of redox fronts in the geological environments, e.g. in the near-field and far-fields, of high level radioactive waste (HLW) repositories of all designs is usually unavoidable. Particularly, the introduction of air and oxidizing waters into a HLW repository during its excavation and construction will take place oxidation of the exposed rock surfaces, and a redox front will be established. In the case that the excavated repository is left open for any length of time during the operation, the continuous supply of oxidants will cause the redox front to migrate from the excavation walls into the host rock. There will also be a possibility of further front migration even after the closure of repository. The understanding of long-term behavior of redox front migration into the host rock, therefore, is one of the key issues to characterize in order to evaluate the barrier function of geological environment and to develop an appropriate process model for the near-field geochemistry.

As an analogous example of the long-term redox front migration, a redox front generated up to several meters depth from the ground surface into the Tertiary sedimentary rock distributed in central Japan has been investigated. Geology and the feature of distribution pattern of the redox front show that the redox reaction has been progressed approximately within the last several hundreds ka after the development of present ground surface structure over the Quaternary covers. Detailed microscopic observation and the geochemical analysis with XRF, EPMA, ICP-MS and SEM-EDS show that the redox front has accumulated the major and minor elements at least ten times more than the background level. Particularly, transition elements such as Zn, Co and Ni as well as HREE are co-precipitated within the high concentration band of Fe and Mn oxide at the front. Microscopically, the aggregations of spheroidal ferric iron grain with the diameter of few micrometers are identified in the pore spaces of sedimentary rock. This suggests that the migration of redox front might be caused by the movement of those grains through the rock matrix scavenging the dissolved elements during migration probably due to the effective surface area of ionic adsorption. This function will also be effective for the radionuclide retardation in the near field environment. The ratio profile of ferrous and ferric iron from the oxidizing part to reducing part through the front also reveals that the oxidizing part of host rock has recovered as reducing environment after passing by the redox front. This is being suggested by the supply of reducing pore water through the surface organic rich soil, and the mobile morphological feature of oxidants might also be considered as a reason of redox front movement.

In this paper, a long-term near-field geochemical process with the detailed morphological and geochemical nature of oxidants in the redox front generating in the sedimentary rock expected in the near-field environment of HLW repository will be presented.