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主 論 文 の 要 旨

論文題目 Microbial $CaCO_3$ precipitation for decreasing the hydraulic conductivity of porous media
(多孔質媒体の透水係数を減少させる微生物炭酸カルシウム沈殿)

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論 文 内 容 の 要 旨

Contamination of groundwater due to leakage from landfills or from paddy fields that have high hydraulic conductivity is a significant problem throughout the world. Because leachate, which can be defined as a highly contaminated liquid that appears after the degradation of the organic fraction of the wastes in combination with percolating rainwater, in landfills and fertilizers and pesticides in paddy fields affect drinking water resources. Therefore, it is desirable for the hydraulic conductivity of a landfill with 50cm of thickness of clay with lower than 10^{-6} cm/s in order to prevent migration of contaminated fluid from the landfill to groundwater and for the hydraulic conductivity of a paddy field lower than 10^{-5} cm/s in order to maintain flooding water levels and to achieve higher efficiencies of fertilizers and pesticides with respect to longer retention. Application of clay minerals has been proposed as a method to decrease the hydraulic conductivity of soil. One of the clay minerals recommended is bentonite, which has a high specific surface area and swelling potential. Clay liners, whose essential material is bentonite, also have been reported to decrease the hydraulic conductivity of landfills. However, the long-term durability of landfill liners is still arguable. Landfill waste degradation is a long-term process, and stabilization of waste to inert state continues for 20-30 years after landfill completion. Several studies have indicated the permeation of inorganic salt solutions and seasonal drying, which leads shrinkage of the clay and development of cracks, can cause the increase in hydraulic conductivity. Thus, clay minerals may not provide confinement well in the long-term in landfills and paddy fields.

It is important to develop other maintenance techniques. Microbial clogging, which results in production of insoluble organic and inorganic compounds by microbes and occurs via many pathways, such as photosynthesis, urea hydrolysis, and sulfate reduction, is considered as one such maintenance method used for hazardous waste confinement. Microbial clogging is expected to decrease the hydraulic conductivity of porous media. However, the effect of $CaCO_3$ precipitation on reducing hydraulic conductivity has not been extensively studied, except for using in sealing cracks, for increasing the resistance of cementitious materials and for achieving the co-precipitation of Sr. Therefore the main purposes of this study can be sorted as to understand quantitatively the effect of microbial clogging on the hydraulic conductivity of glass beads as porous media in the presence of *Sporosarcina pasteurii*, to determine the effects of bentonite suspension and yeast extract as nutrient source on decrease in hydraulic conductivity of glass beads as porous media in the presence of *Sporosarcina pasteurii* and to examine the effect of *in situ* microbial clogging in a paddy field with a high hydraulic conductivity.

$CaCO_3$ precipitation induced by resting *Sporosarcina pasteurii* (ATCC 11859) was investigated using continuous-flow of a precipitation solution composed of 0.5 M $CaCl_2$ and 0.5 M urea through columns containing glass beads. It was shown that the subsequent formation of $CaCO_3$ precipitation reduced hydraulic conductivity from between 8.38×10^{-1} and 3.27×10^{-4} cm/s to between 3.70×10^{-1} and 3.07×10^{-5} cm/s. Analysis using the Kozeny–Carman equation suggested that the effect of microbially induced $CaCO_3$ precipitation on hydraulic conductivity was not due to the formation of individual $CaCO_3$ crystals but instead that the precipitate aggregated with the glass beads.

The effects of bentonite suspension and yeast extract as nutrient source on reduction in hydraulic conductivity of columns containing glass beads in the presence of *Sporosarcina pasteurii* (ATCC 11859) was examined. Even though, introduction of only bentonite suspension caused small decrease in hydraulic conductivity, and introduction of cell suspension and precipitation solution including yeast extract caused relatively high decrease in hydraulic conductivity, the combination of introduction of bentonite suspension, cell suspension and precipitation solution including yeast extract caused the highest decrease in hydraulic conductivity. In other words, the lowest hydraulic conductivity, which was reduced from between 8.38×10^{-1} and 4.10×10^{-3} cm/s to between 9.91×10^{-4} and 2.11×10^{-6} cm/s, was observed with introduction of bentonite suspension, cell suspension and a precipitation solution composed of 0.5 M $CaCl_2$, 0.5 M urea, and 2% w/v yeast extract.

The microbial clogging was applied to a paddy field in Nagoya University Farm. The hydraulic conductivity of the paddy field was reduced by *in situ* microbial calcium carbonate precipitation through the addition of bentonite, $CaCl_2$, urea, and corn steep liquor. Biocalcification treatment decreased the hydraulic conductivity of the treated plots in the field from an average of 10^{-3} cm/s to a range of 10^{-5} to 10^{-7} cm/s. Laboratory experiments demonstrated that addition of corn steep liquor enhanced the urease activity of soil, even at 4°C, as indicated by a decrease in urease activation energy temperature sensitivity. These experimental results resemble the gradual decrease in hydraulic conductivity observed in the field when the average daily temperature was 7°C.

The reduction in hydraulic conductivity of glass beads was explained by the amount of $CaCO_3$ precipitate and the number of deposited resting cells. Analysis using the Kozeny–Carman equation indicated that the mechanism underlying the reduction in hydraulic conductivity was the aggregation of the glass beads with the $CaCO_3$ precipitate, which increased the beads' effective size. The reduction in hydraulic conductivity of glass beads was improved by deposition of bentonite and enhancement of urease activity which caused more $CaCO_3$ precipitation, with respect to yeast extract that promoted growth of bacterial population. Achievement of 10^{-6} cm/s of hydraulic conductivity indicated the applicability of the technology to the environment for confinement.

The hydraulic conductivity of the paddy field was reduced significantly. Factors affecting the efficiency of decreasing the hydraulic conductivity can be sorted as $CaCO_3$ formation amount, temperature, and nutrients. Activity in low temperatures increases the applicability of the technology to the field.