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## 博士論文題名

Novel polymerized bentonites and their application as the core of GCL used in aggressive environment

(新たに開発した重合ベントナイトと侵食性環境に 使用される GCL への応用)

To increase the swell potential and maintain lower permeability of bentonite in aggressive environments (high concentration cation solutions, very high or low pH solutions, etc.), novel polymerized bentonites (PBs) were produced, and their potential use as the core material of geosynthetic clay liner (GCL) to be used in the aggressive environments were evaluated.

(1) Polymerization method and suitable conditions. Novel PBs were produced using natural sodium bentonite (UB) and two monomers, acrylic acid (M<sub>1</sub>) and acrylamide (M<sub>2</sub>), using free radical polymerization method. The initiator (I) used was potassium persulfate, the deionized water (DI-W) as solvent. Using free swelling index (FSI) in 0.6 M NaCl solution as an index, the suitable polymerization conditions identified were: pH of 6,  $I/(M_1 + M_2) = 0.5\%$ , and  $M_1/M_2 = 0.5$ . Further, during the polymerization process, instead of using nitrogen gas to remove oxygen, a method of using vacuum pressure was established.

(2) Microstructures of novel PBs. Designating PB with polymer content of 10% as 0.1PB and so on, the results of X-ray diffraction (XRD) pattern shows that d-spacing of bentonite crystal of 0.1PB is the same as that of UB and the product was a microcomposite. For 0.2PB, its dspacing was increased and the products was a nanocomposite. The SEM images of swelled PBs show large amount of polymer like net structures between bentonite particles. These polymers will have an important role for increasing the resistance of PBs to the aggressive cation solutions.

(3) Swelling and consolidation properties and permeabilities of the PBs. The properties of 0.1PB and 0.2PB were evaluated by a series of experiment test, i.e., FSI tests, swelling pressure tests, consolidation tests and permeability tests. The results from FSI tests and the swelling pressure tests show that the PBs have higher swelling capacity than those of UB in DI-W and cation solutions (i.e., 0.6 M NaCl and 0.03-0.06 M CaCl<sub>2</sub>). With 0.6 M NaCl solution, the FSI of 0.1PB is approximately 29 mL/2g. which exceed the requirement to be used as the core of GCLs (> 24 mL/2g). The mechanism that PBs had higher swelling capacity is that the novel PBs have two hydrophilic groups, -CONH<sub>2</sub> and -COONa. Due to both groups can be connected with exchangeable cations directly or indirectly by physical interaction, which contributes to reduce the amount of cations entering the diffuse double layers of bentonite particles. The results of the consolidation tests show that for all liquids tested, the compression indexes (Cc) of PBs are higher than that of UB. For a given void ratio, the order of permeability calculated from the consolidation test results is  $k_{UB} >$  $k_{0.1PB} > k_{0.2PB}$  (subscripts indicate the corresponding materials). For void ratios up to 5 for 0.1PB, the value of k is still smaller than 10<sup>-10</sup> m/s in the 0.6 M NaCl and 0.03-0.06 M CaCl<sub>2</sub> solutions. The directly measured values of permeability from the flow rate test are comparable with that from the consolidation test results. Therefore, it is suggested that the novel PBs have potential to be used as a barrier material under high Na<sup>+</sup> concentration environments (e.g., sea water).

(4) Behavior GCLs with 0.1PB as core material (PB-GCLs). Leakage rate test results show that for the PB-

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GCL, with DI-W, a circular damage-hole up to 100 mm in diameter, and with 0.6 M NaCl solution, a damage-hole up to 15 mm in diameter were self-healed. In comparison, for UB-GCL in DI-W, a damage hole up to 60 mm in diameter and in 0.6 M NaCl solution up to 5 mm were self-healed. Therefore, PB-GCL had higher self-healing capacity. Further, the methods for predicting self-healing capacity of UB-GCL as well as PB-GCL with a circular damage-hole have been established. Good agreement between the measured and predicted self-healing ratios (healed area divided by the total damage area) was obtained. The prediction methods are useful for selecting a suitable GCL to be used under a given environment.

In summary, this research developed a new method for polymerizing bentonite and produced PBs which had higher swelling capacity and lower permeabilities with cation solutions. In addition, the leakage rate test results indicate that PB-GCLs (0.1PB) had very lower permeability and higher self-healing capacity. It is suggested that the novel PBs are potentially to be used as a barrier material under aggressive environments (e.g., seawater).