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Microbially mediated sand solidification using calcium phosphate compounds

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ABSTRACT

To evaluate the potential utility of a new calcium-phosphate-compound (CPC)-based biogrout (CPC biogrout), we conducted unconfined compressive strength (UCS) tests and scanning electron microscope (SEM) observations of sand test pieces cemented with CPC biogrout. The CPC biogrout was produced using (1) soil extracts that contained microorganisms derived from one of two soils, which had different pH values, and (2) one of three amino acids or urea as a pH-increasing reactant. A temporal increase in pH was observed in slightly acidic soil by the addition of ammonia sources. On the other hand, there was no significant increase in pH in slightly alkaline soil except for that due to urea. In most cases, the UCS of the test pieces cemented with CPC biogrout produced using soil extracts from acidic soil along with an ammonia source was higher than that of the test pieces cemented with CPC biogrout produced without the addition of ammonia sources. SEM observation of test pieces with UCS of over 50 kPa showed the presence of whisker-like CPC crystals. These results suggest that CPC biogrout affords sufficient strength as a countermeasure for soil liquefaction and that amino acids can be used as new pH-increasing reactants for CPC biogrout. In addition, they suggest that either CPC biogrout or CPC chemical grout alone, or a combination of the two grouts, can be used depending on the various properties of grounds and soils.

1. Introduction

Several major cities in Japan are located on an alluvial plain and a number of settlements in these cities are vulnerable to disasters such as earthquakes. Ever since soil liquefaction was observed in the Niigata earthquake of 1964 (Ohsaki, 1966), the damage caused by soil liquefaction has been confirmed in many earthquakes, such as the great East Japan earthquake of 2011. Hence, there is an urgent need for seismic reinforcement, including countermeasures for soil liquefaction (The Japanese Geotechnical Society, 2011).

In recent years, grout materials have been developed to control ground permeability and to reinforce the ground with bacterially produced cement material (DeJong et al., 2006; Whiffin et al., 2007; Ivanov and Chu, 2008; Hata et al., 2009; Van Paassen et al., 2009; Harkes et al., 2010; Kawasaki et al., 2010; Mukunoki et al., 2010; Van Paassen et al., 2010; Inagaki et al., 2011). These grout materials are called biogrouts, and they are attracting attention as promising successors to chemical grouts because the microbial reaction in biogrouts is expected to be slower than the chemical reaction in chemical grouts; a slower reaction would reduce the solidification speed, allowing the biogrout to spread through a greater volume of soil. A number of mineral formation mechanisms have been considered for biogrout, of which the notable ones are as follows: $CaCO_3$ precipitation using urea and ureolytic bacteria (Harkes et al., 2010); $CaCO_3$ precipitation using glucose and yeast (Kawasaki et al., 2006); siloxane bond formation using glucose and yeast (Terajima et al., 2009); and iron- or manganese-compound precipitation using iron-oxidizing bacteria (Weaver et al., 2011). However, it must be noted that soil and rock vary greatly in their physical, chemical, and biological properties, and therefore, to apply biogrout to various soils and rocks, it is important to develop new mechanisms of cement material precipitation. To this end, we are carrying out fundamental studies on the use of calcium phosphate compounds (CPC) as novel grout materials (Akiyama and Kawasaki, 2012; Figure 1).

Research on CPC precipitation and solidification is also currently underway in the field of medical and dental science. Fernández et al. (1998) reported that the unconfined compressive strength (UCS) of CPC exceeds 10 MPa under normal temperature and pressure conditions. CPC has two unique characteristics: its pH is dependent on its solubility and it exhibits a self-setting mechanism (Tung, 1998). We discovered that grout comprising only CPC (CPC chemical grout) increased the UCS of sand test pieces with time and that the volume of precipitated CPC crystal increased with the pH (Akiyama and Kawasaki, 2012). The results indicated the possibility of developing a CPC biogrout in which an increase in biological pH brings about crystal precipitation of CPC from a low-pH injection solution. Such a CPC biogrout would enable the control of solidification speed; furthermore, the strength of the ground can be increased by using a combination of CPC chemical grout, for rapid solidification, and CPC biogrout, for long-term solidification.

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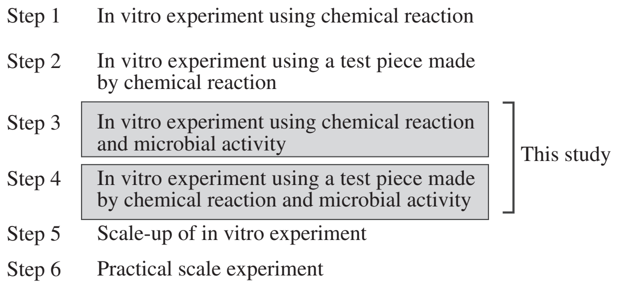

Fig. 1. Flowchart of the entire study. The steps carried out in this study are highlighted in gray.

$CaCO_3$ precipitation using urea and ureolytic bacteria—the most common biogrouting mechanism that involves the use of pH-increasing reactions—occurs as follows: hydrolysis of urea (Eq. (1)), pH elevation by NH_3 production (Eq. (2)), dissolution of CO_2 (Eq. (3)), and $CaCO_3$ precipitation (Eq. (4)) (Whiffin et al., 2007; De Muynck et al., 2010; DeJong et al., 2010; Harkes et al., 2010).

$$(NH_2)_2CO + H_2O \rightarrow 2NH_3 + CO_2 \qquad (1)$$

$$NH_3 + H_2O \rightarrow NH_4^+ + OH^- \qquad (2)$$

$$CO_2 + H_2O \rightarrow 2H^+ + CO_3^{2-} \qquad (3)$$

$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3 \qquad (4)$$

However, injection of foreign ureolytic bacteria (*Sporosarcina pasteurii*) involves many challenges, such as obtaining the approvals and licenses, public acceptance, and the necessity to monitor the microbial ecology for safety (METI Web site, 2005). Although the efficiency of industrial urea production continues to improve, the process nevertheless involves ammonia production, which results in fossil fuel consumption and CO_2 emission (Rafiqul et al., 2005). Moreover, urea is a key raw material for fertilizer production, so urea prices may increase with fertilizer demand; this may render the use of urea for grout production uneconomical. In this study, we investigate a fundamental concept for soil improvement: the activation of indigenous bacteria for soil improvement to preclude the need for injecting foreign microbes. Furthermore, we focus not only on urea but also on novel ammonia sources (amino acids) as pH-increasing reactants to develop adaptable grout materials for a variety of soils and rocks.

In this study, we carried out a fundamental laboratory test of a novel grout called CPC biogrout. A schematic of the study design is shown in Fig. 2. First, we estimated the potential for decomposition of amino acids as candidates for novel ammonia sources by conducting a pH-increasing test. Second, on the basis of these results, we carried out UCS tests to estimate the strength of test pieces cemented by CPC chemical grout and CPC biogrout and observed the test pieces using a scanning electron microscope (SEM). It has been reported that soil liquefaction does not occur during an earthquake if the UCS of the soil and ground is between approximately 50 and 100 kPa (Port and Harbour Institute, 1997; Yamazaki et al., 1998; Matsuda et al., 2008). Therefore, we set 50 to 100 kPa as the target UCS range for the CPC chemical grout and CPC biogrout.

2. Selection of ammonia sources

In this study, we focused on ammonification—the process of amino acid deamination after hydrolysis of protein by heterotrophic bacteria—as the pH-increasing reaction (Galloway, 2005). Amino acids can be produced economically from high-protein organic waste using bacteria. Therefore, we chose amino acids and urea as ammonia sources. In other words, we adopted the pH-increasing mechanism of microbial ammonia production in soil. To the best of our knowledge, no published study has yet reported the use of indigenous microorganisms and ammonia sources other than urea in actual biogrout. The chemicals and enzymes involved in the ammonification and degradation mechanism of amino acids are listed in Tables 1 and 2, respectively.

We identified three amino acids as promising new ammonia sources from among the 20 that mainly constitute proteins (Alberts et al., 2009). First, asparagine (Asn) and glutamine (Gln), each with two amidogens and an initially acidic pH, were chosen for their high efficiency of ammonia production. These amino acids release ammonia according to Eqs. (5) and (6) to produce aspartate (Asp) and glutamate (Glu), respectively, as by-products. These by-products are then taken up by soil microorganisms and the citric acid cycle after conversion to oxaloacetic acid and oxoglutaric acid (Magasanik, 1982).

$$Asn + H_2O \rightarrow Asp + NH_3 \qquad (5)$$

$$Gln + H_2O \rightarrow Glu + NH_3 \qquad (6)$$

Second, we also selected the simplest amino acid glycine (Gly), which has an initially acidic pH and high solubility in water. After it is

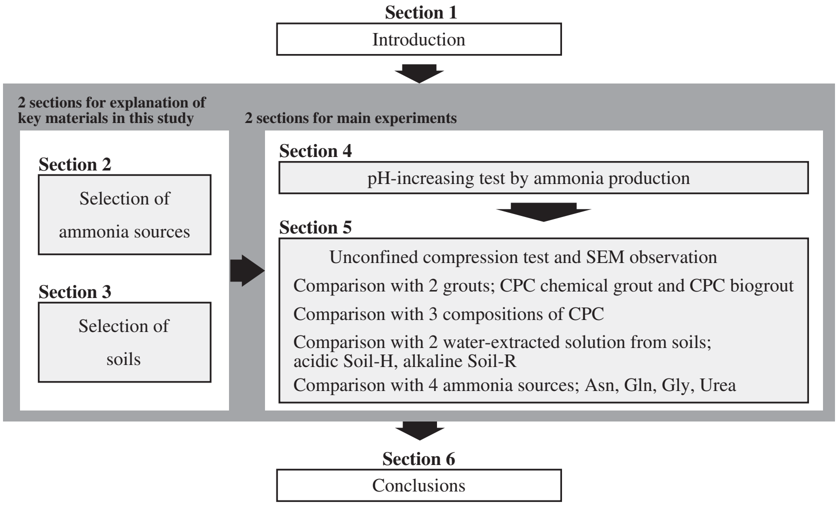

Fig. 2. Design and flow of the present study.