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Desaturation via Biogenic Gas Formation as a Ground Improvement Technique

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Abstract

Desaturation by biogenic gas formation can significantly affect the hydro-mechanical behaviour of soil. The high compressibility of the gas dampens pore pressure build up during both monotonic and cyclic undrained loading. Stimulating biogenic gas production therefore has potential as a ground improvement method to mitigate the risk of both static liquefaction and earthquake induced liquefaction. However, gas generated below the ground water table at shallow depth may also constitute a hazard for offshore foundations and terrestrial deposits, as a sudden release of trapped gas may cause instability. In order to evaluate the potential use of biogenic gas for geotechnical applications it is essential to be able to predict gas production and assess its effect on the hydro-mechanical behaviour of a soil. A basic theoretical framework to estimate the volume of gas produced by a biogenic process and the related degree of saturation, experimental results on the rate of gas generation, and its impact on soil behavior are presented herein.

INTRODUCTION

This paper provides an overview of recent developments regarding the potential use of biogenic gas formation for geotechnical applications. Desaturation due to biogenic gas formation can significantly affect the hydro-mechanical behaviour of soils. A small fraction of gas is sufficient to reduce the pore fluid bulk stiffness (Biot, 1941; Skempton, 1956) and dampen pore pressure build up during monotonic and cyclic undrained loading (Yang et al. 2004; Yegian et al. 2007, He and Chu 2014). Desaturation by stimulation of biogenic gas formation has been suggested as a means of mitigating both static liquefaction (He and Chu 2014; Pham et al. 2016) and earthquake-induced liquefaction (Rebata-Landa and Santamarina 2012; He et al. 2013; Kavazanjian et al. 2015). However, when a large amount of trapped gas suddenly escapes in a seafloor sediment or a terrestrial deposit with a shallow water table it may trigger instability. Particularly for offshore foundations, shallow gas is considered a major hazard (Grozic et al. 1998) as the sudden release of gas can cause a bearing capacity failure. The presence of gas can also significantly reduce the hydraulic conductivity of soils, even if it fills up a small fraction of the pore space (Ronen et al., 1989; Baird & Maldron, 2003; Mahabadi and Jang, 2014; Mahabadi et al. 2016). In order to evaluate the potential use of desaturation via biogenic gas for geotechnical applications, it is essential to be able to predict the amount of produced gas and estimate its effect on the hydro-mechanical behaviour of the treated sediments (Pham, 2017). This paper provides an overview of recent experimental work regarding the impact of desaturation via biogenic gas formation on the behaviour of soil and presents a basic theoretical framework to estimate the volume of produced gas and the related degree of saturation.

MICROBIALLY INDUCED DESATURATION BY NITRATE REDUCTION

The most common biogenic gases that are formed in the subsurface are methane (CH_4), nitrogen (N_2), hydrogen sulphide (H_2S), and carbon dioxide (CO_2). These gases are the product of metabolic processes of microorganisms. As nitrogen gas has a low solubility and is neither toxic nor a greenhouse gas, biogenic production of nitrogen gas seems to be the most appropriate candidate for ground improvement via biogenic gas generation. Several recent studies have investigated the potential use of biogenic nitrogen gas for ground improvement (He et al. 2013; He and Chu 2014; Kavazanjian et al. 2015, Pham et al. 2016).

Nitrogen gas is formed in the subsurface when denitrifying microorganisms are provided with a solution containing nitrate and dissolved organic matter. This process may be referred to as dissimilatory reduction of nitrate, or denitrification. Denitrification is a natural process in the global nitrogen cycle. In denitrification, organic matter is oxidized to inorganic carbon and nitrate is reduced to nitrogen gas. The reduction of nitrate (NO_3^-) to nitrogen gas (N_2) goes through several intermediate reactions, which involves specific enzymes and the formation of intermediate nitrogen compounds: nitrite (NO_2^-), nitrous oxide (N_2O), and nitric oxide (NO) (Rebata-Landa and Santamarina, 2012). When employing denitrification for engineering purposes, accumulation of these intermediates should be avoided as nitrite and nitric oxide are both toxic and inhibit microbial growth and nitrous oxide is a very strong greenhouse (Almeida, Julio et al. 1995; Chung and Chung 2000; Zumft, 1997; Madigan et al. 2012). The microorganisms that mediate denitrification (i.e., dentrifiers) are ubiquitous in the subsurface. Hence, denitrification can be induced in most soils by stimulation of indigenous microorganisms.

To promote the efficient and full reduction of nitrate to nitrogen gas, selecting the right substrate composition is essential (O’Donnell 2016, Pham et al. 2016). Too much nitrate may lead to accumulation of intermediate compounds, while leaving a large excess of organic substrate would be inefficient. Various organic substrates can be used, but many studies used a solution containing calcium acetate and calcium nitrate (Van Paassen 2009; Van Paassen et al. 2010; Van der Star et al., 2012; Kavazanjian et al., 2015, Hamdan et al. 2016; Pham et al. 2016). The advantage of using a calcium salt as substrate is that the inorganic carbon produced by the microbial metabolism will precipitate with the dissolved calcium to form calcium carbonate minerals. Calcium carbonate ($CaCO_3$) precipitation helps to buffer the pH as it consumes the alkalinity produced by reduction of the nitrate (i.e. bicarbonate (HCO_3^-) and hydroxide (OH^-)). Maintaining a stable pH stimulates microbial growth and helps to prevent the accumulation of toxic intermediate nitrogen compounds (Pham et al., 2016). $CaCO_3$ precipitation may also increase strength, stiffness and dilatancy of soils by coating the soil particles and creating cementing bonds at the particle contacts, a process referred to as Microbially Induced Carbonate Precipitation (MICP). While MICP through denitrification is a time-intensive process (Martin et al. 2013; Van Paassen et al. 2010), the associated solid mineral phase is considered to provide a more durable improvement. However, the required amount of substrates for significant cementation is much higher than the amount which is required to significantly desaturate a soil. Hence, the combined process of Microbial Induced Desaturation and Precipitation (MIDP) can be considered a two-stage ground improvement method: in the short term mitigating liquefaction by gas formation and in the long-term by calcium carbonate precipitation (Kavazanjian et al. 2015; Khodadadi et al. 2017; O'Donnell 2016).

THEORETICAL FRAMEWORK TO ESTIMATE THE GAS VOLUME

The amount and rate of gas production depends on the substrate availability, metabolic stoichiometry and conversion rate, and the ambient conditions (temperature and pressure). The stoichiometry of the metabolic reaction consists of two parts: a catabolic reaction and anabolic reaction. With acetate ($C_2H_3O_2^-$) as carbon source and electron donor and assuming complete reduction of nitrate to nitrogen gas, the stoichiometry of the catabolic reaction determined by solving the mass and electron balance is shown in Eq. 1:

$$C_2O_3H_2^- + 1.6NO_3^- + 0.4H_2O \rightarrow 0.8N_2 + 2HCO_3^- + 0.6OH^- \quad (1)$$

The catabolic reaction generates energy, which is required to grow biomass ($CH_{1.8}O_{0.5}N_{0.2}$) in the anabolic reaction shown by Eq. 2:

$$0.73C_2O_3H_2^- + 0.2NO_3^- + 0.28H_2O \rightarrow CH_{1.8}O_{0.5}N_{0.2} + 0.45HCO_3^- + 0.48OH \quad (2)$$

The stoichiometry of complete metabolic reaction depends on the growth rate of the denitrifying bacteria. The stoichiometry at maximum growth, determined using a thermodynamic approach suggested by Kleerebezem and Heijnen (2010), is shown by Eq. 3:

$$1.21C_2H_3O_2^- + 0.97NO_3^- + 0.17H_2O \rightarrow CH_{1.8}O_{0.5}N_{0.2} + 0.39N_2 +$$

$$1.41HCO_3^- + 0.76OH^- \tag{3}$$

In reality, the microbial growth rate is often less than the maximum growth rate. It can be limited by a variety of factors, such as inhibition due to accumulation of intermediate nitrite or limited availability of substrate, nutrients or trace elements. As a result of these inhibition mechanisms, the actual metabolic reaction stoichiometry varies between conditions of maximum growth (Eq. 3) and zero growth (Eq. 1). Consequently, the yield of nitrogen gas over nitrate (N_2/NO_3^-) ranges from 0.4 to 0.5.

Upon initial formation, the dinitrogen (N_2) produced by the denitrifying organisms is in a dissolved state. Once the solution gets sufficiently supersaturated (saturated beyond a threshold concentration), small gas bubbles will form and rapidly grow until the nitrogen concentration in aqueous solution, c_a [in mol L^{-1}], is in equilibrium with the partial pressure in the gas phase, p_g [in atm], according to Henry's law:

$$k_H = p_g / c_a \tag{4}$$

in which k_H is Henry's constant for nitrogen, 1542.6 [atm L mol^{-1}] (Yaws, 2012). When the gas is present in the soil as small spherical air bubbles, the gas pressure is related to the radius of the bubble. The capillary pressure, which is the difference between the gas pressure, p_g, and water pressure, p_w, is expressed by the Young-Laplace equation (Blander and Katz 1975):

$$p_g - p_w = 2\ T_s / r \tag{5}$$

in which T_s is the surface tension of the liquid and *r* is the radius of the bubble in [m]. For water at 20°C, T_s is approximately 0.072 [N m^{-1}] (Rebata-Landa & Santamarina, 2012). Hence, for small bubbles the pressure in the gas phase can be significantly higher than the water pressure, but as the bubbles grow larger or agglomerate the pressure difference decreases and the gas pressure may eventually become equal to the pore water pressure. Assuming N_2 is the only gas being formed, the amount of gas, n_g, in equilibrium with the dissolved concentration of nitrogen, c_a, can be calculated from the total concentration of gas formed in the metabolic reaction, c_t:

$$n_g = (c_t - c_a)\ V_L \tag{6}$$

where V_L is the liquid volume [in L]. Neglecting potential capillary pressure effects which can occur in very small gas bubbles, the partial gas pressure, p_g, is assumed to be equal to the hydraulic pressure, u, in the pore fluid. In this case the volume of gas, V_g, can be determined from the amount of gas, n_g, (in moles) at the given pressure and temperature using the ideal gas law.

$$V_g = n_g\ R\ T / p_g \tag{7}$$

where, T is the temperature in the system, which is assumed at 293 [K] and R is the gas constant, i.e. $8.205\ 10^{-2}$ [L atm K^{-1} mol^{-1}]. The gas saturation, S_g, is defined as the ratio between the gas volume and the total pore volume, V_p:

$$S_g = V_g/V_p \tag{8}$$

Using this theoretical approach and assuming that all nitrate in the catabolic reaction is fully converted to nitrogen gas, the degree of saturation can be calculated as a function of pore water pressure (or the corresponding water depth under hydrostatic conditions) (Pham et al., 2016). Results of this calculation for the two extreme metabolic stoichiometry scenarios of zero and maximal microbial growth for three different nitrate (NO_3^-) concentrations are presented in Figure 1.

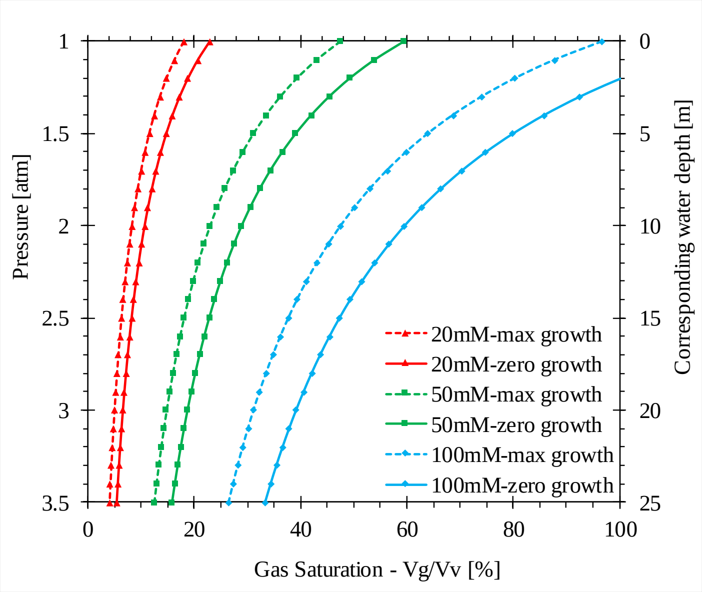

Figure 1. Gas saturation induced by denitrification as a function of pore pressure for maximal growth (dashed lines) and zero growth (continuous lines) for three different concentrations of consumed NO_3 (from Pham 2017)

As shown in Figure 1 the resulting gas saturation strongly depends on the amount of converted nitrate and the pore water pressure, but is relatively insensitive to the metabolic stoichiometry. With increasing water depth the volume of gas gets smaller due to both the higher water pressure and to a higher solubility. Considering that a gas saturation of about 5% has been shown to be enough to significantly increase liquefaction resistance (He and Chu 2014, O'Donnell 2016), it seems that one pore volume of substrate solution with a nitrate concentration of 20 mmol L^{-1} generating about 0.14 g L^{-1} nitrogen gas may be sufficient to mitigate the potential for liquefaction for treatment depth of up to 100 ft below the water table (pore water pressure up to approximately 3 atm, or 6250 lb/ft^2).

GAS MIGRATION AND STABILITY

The calculated gas saturation in Figure 1 is expected to be higher than the actual gas saturation based on the assumptions made and due to convective and diffusive transport mechanisms. First, the dissolved concentration may be higher than the equilibrium concentration. In order to form gas bubbles the solution needs to be sufficiently supersaturated to exceed the gas nucleation threshold. Second, when bubbles are very small the pressure inside the bubble is higher than the water pressure ($p_g > p_w$). Hence, following the ideal gas law (Eq. 7), more moles of gas can be stored in the same gas volume. Finally, when the bubbles are small enough to pass through the pores, or when the gas pressure is high enough, the gas may migrate.

Okamura et al. (2006) observed that the gas phase in a soil can remain stable for several years. Once the bubbles are trapped, the gas phase may be redistributed through Ostwald ripening, a phenomenon in which small bubbles tend to dissolve, diffuse, and agglomerate into the larger bubbles (Schmelzer and Schweitzer, 1987). Dissolution of the gas phase may also occur if the dissolved concentration in the surrounding water is below the equilibrium concentration. When performing MIDP experiments inside a triaxial cell, Pham et al. (2016) found that after the gas formed inside the sample the gas volume slowly reduced in time. They attributed this phenomenon to dissolution and diffusion through the latex membrane. O'Donnell et al. (2016, 2017) noted the same effect in samples formed in acrylic columns.

When bubbles grow bigger than the pores in which they are trapped, they can only migrate when the gas pressure exceeds the required capillary pressure to squeeze through the pore throats (Haines 1930, Brooks and Corey 1964). The required gas pressure can still be calculated using Eq. 5, assuming that the contact angle between the gas-liquid interface and the solid is zero and the bubble radius is equal to the radius of the largest connected pore throat. Once the air entry value of the soil is exceeded, the gas squeezes through the pore throat and migrates to the next pore, while the gas volume expands and the gas pressure drops. This uneven displacement of gas bubbles is often referred to as Haines jumps (e.g., Armstrong et al. 2015). Once the gas forms a network of connected gas filled pores, rapid upward percolation of the gas may occur through vertical pathways in the soil (Soares et al. 1988, Istok et al. 2007). The degree of saturation at which this rapid upward percolation occurs is defined as the gas percolation threshold. Any further gas production after reaching the gas percolation threshold will irregularly vent out of the pores. Pham (2017) showed that the gas percolation threshold for poorly graded fine sands ranged from 20 to 25%.

The percolation of gas is affected by the soil intrinsic permeability. During the upward migration of gas, bubbles may get stuck below layers with a low intrinsic permeability such as clay or silt layers. In this case, the gas pressure may not be able to exceed the air entry pressure of the layers above and consequently the gas may spread laterally to form a gas pocket. The intrinsic permeability of the soil may also be affected by the formation of the other products of the metabolic conversion such as biomass or solid minerals (Baveye et al., 1998; Thulner et al., 2002; Pham et al., 2017).

EFFECT OF BIOGENIC GAS ON MECHANICAL BEHAVIOUR

The effect of biogenic gas on mechanical behaviour of soils depends on whether the gas is present as a continuous phase in contact with the atmosphere or occluded as disconnected bubbles, pockets or lenses (Leroueil et al., 2015). For liquefaction mitigation the biogenic gas should be in an occluded state. When the gas fraction is small and distributed as bubbles which are smaller than the typical pore size, the effective stress of the unsaturated soil can be calculated in a similar way as for a fully saturated soil. However, due to the large compressibility of the gas phase, the compressibility of the pore fluid increases significantly (Biot, 1941; Tsukamoto et al. 2002; Ishihara et al., 2004). As a result, small levels of desaturation (e.g., 1 percent) can increase small strain stiffness during undrained loading and increase cyclic resistance significantly (Ishihara and Tsukamoto, 2004; Okamura and Soga, 2006).

He and Chu (2014) performed consolidated undrained compression tests on sand which was desaturated by stimulating biogenic nitrogen gas production. Test were performed at similar initial void ratio (e_0), but with various degree of saturation (S_r) as represented by Skempton's B-value. The results showed that a reduced degree of saturation dampens pore pressure build-up during undrained monotonic loading, which results in a higher deviatoric stress and higher stiffness at relatively small strains, as illustrated in Figure 2. Pham et al. (2016, 2017) obtained similar results. O'Donnell et al. (2016, 2017) performed undrained cyclic loading tests on sand which was desaturated by denitrifying bacteria and, like Ishihara and Tsukamoto (2004), they found that a small reduction in water saturation can significantly increase the undrained shear strength of sand subject to cyclic loading, as shown in Figure 3.

When a large fraction of gas gets trapped forming gas pockets, the upward buoyancy force counteracts the overburden pressure, reducing the effective stress on the underlying strata. As the overburden pressure transfers to the gas phase and the water phase underneath, the gas pressure and water pressure below the gas phase will increase. If the gas pressure in a gas pocket becomes equal to or greater than the overburden pressure but does not exceed the capillary pressure required to squeeze through the pores of the overlying soil ($p_{ov} = p_g < p_c$), a gas pocket may generate a fracture and lift up the overlying soil. As a result the effective stress on the underlying layers may reduce to zero (Sobkowicz and Morgenstern, 1984; Grozic, 1998; Leroueil et al. 2015).

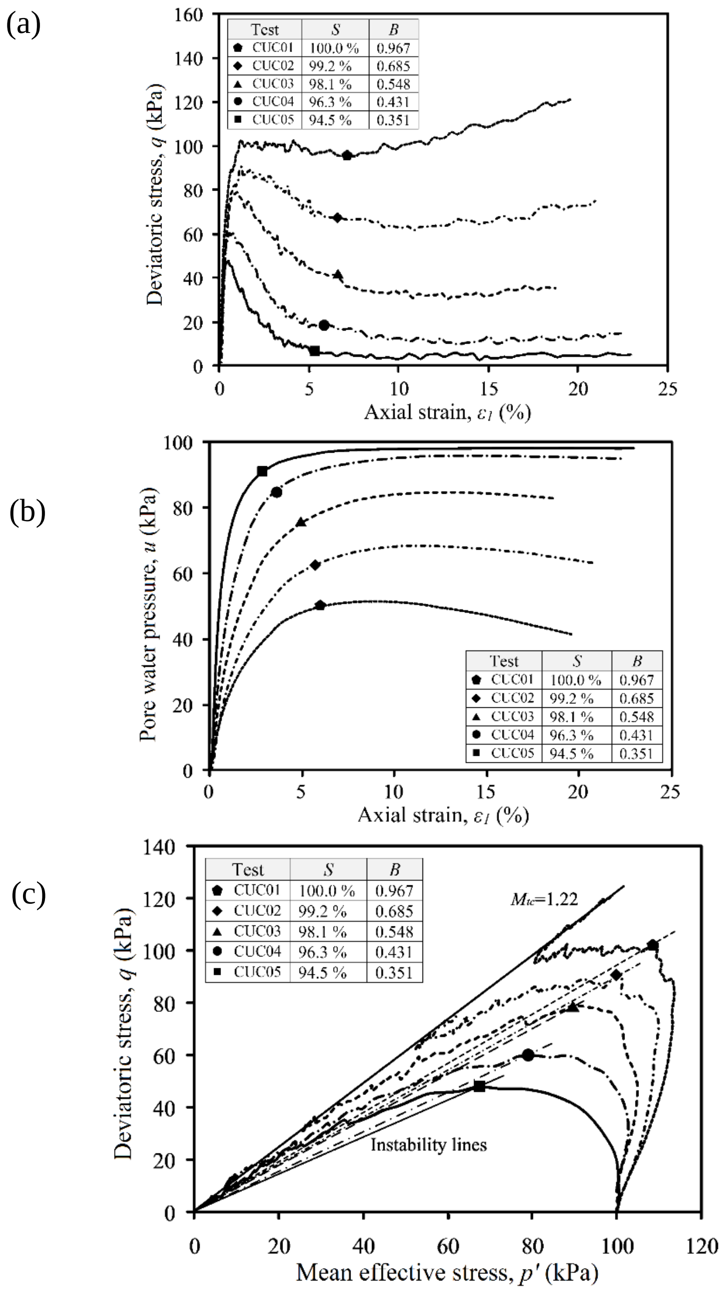

Figure 2 Mechanical response of a partially saturated sand under undrained monotonic loading. (a) stress-strain response, (b) pore pressure response and (c) effective stress paths (from He and Chu 2014 with permission of ASCE).

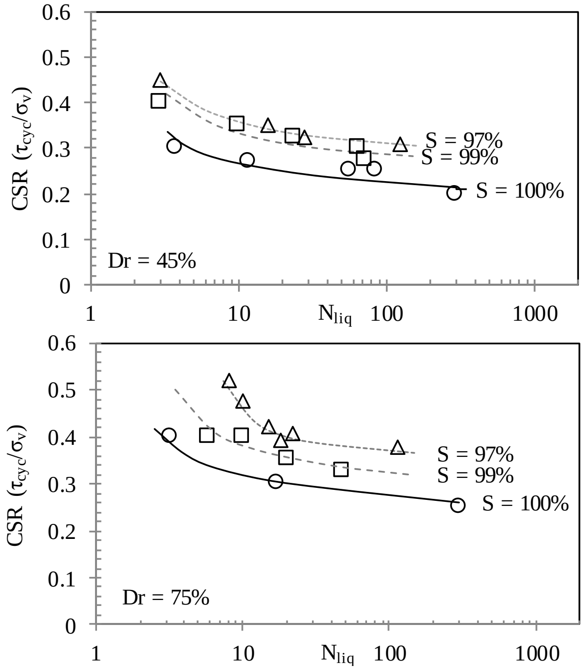

Figure 3 Response of partially saturated sand under undrained cyclic loading (O'Donnell 2016)

During an experiment in which a sand column was flushed from bottom to top with a substrate solution stimulating production of nitrogen gas through microbial denitritification, occluded gas lenses appeared in the upper part of the column and the partly saturated sand above the gas lenses lifted up about 15 cm, as illustrated in Figure 4. Similarly, Torres and Larrahondo (2017) showed significant vertical expansion of coarse grained sand when they mixed the sand with organic matter and exposed the organic matter to chemical oxidation using hydrogen peroxide.

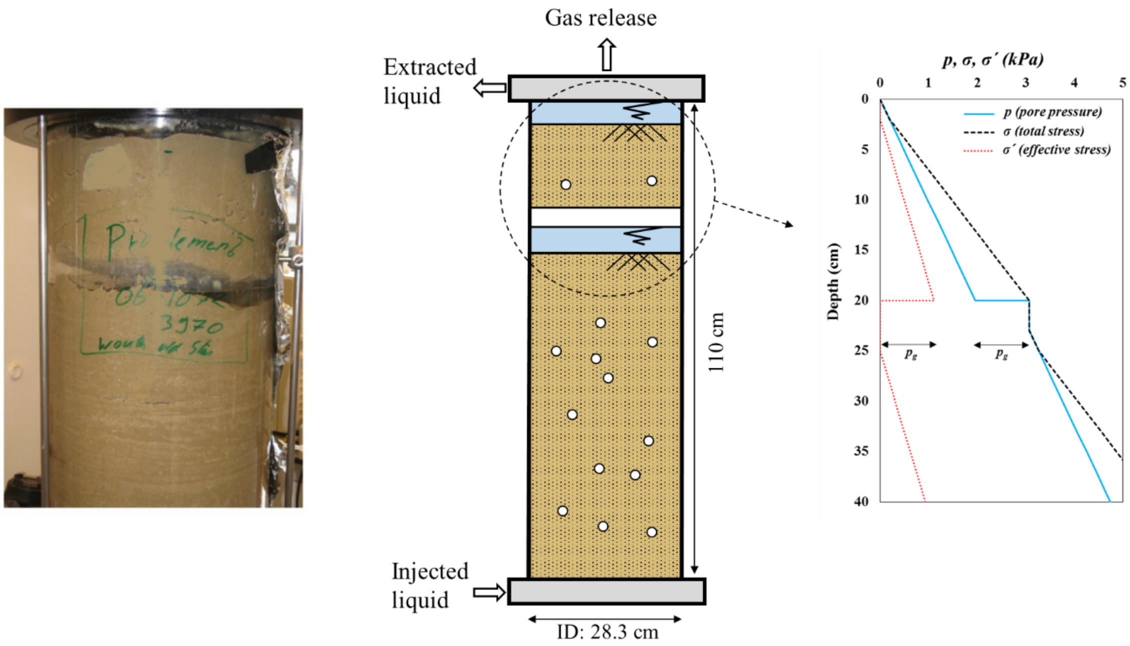

Figure 4: Occluded gas lenses appeared in the upper part of a sand column, lifting up the overburden, increasing the pore pressure and reducing the effective stress in the underlying soil (Van Paassen, 2009)

Gas filled pockets cause volumetric expansion of the sand and typically occur under conditions of low confinement (e.g. Leroueil et al. 2015). However, in various natural and man-made soil profiles including as fine-grained marine sediments or mine tailings, such low confinement conditions can extend hundreds of meters below the surface. Semi-horizontal trapped gas pockets and irregular gas venting has been observed in natural methane hydrate deposits (e.g. Suess et al., 1999) and is postulated as the mechanism behind mud volcanos in various marine and terrestrial soil profiles (e.g. Dimitrov, 2002). When gas pockets or gas-filled fissures reach the surface or a zone with higher permeability, the accumulated gas may escape suddenly, causing instability (Grozic, 1998). If the gas suddenly escapes in this manner, fissures close and the gas saturation will drop. However, some of the structure of the soil may be permanently altered, as cavities may remain once the gas is relieved due to bridging sand particles (Torres & Larrahondo, 2017).

CONCLUSION

Biogenic gas production has potential to be used as a relatively rapid ground improvement method, e.g. to mitigate earthquake-induced liquefaction. As nitrogen gas has a low solubility and is neither toxic nor a greenhouse gas, stimulating biogenic production of nitrogen gas through nitrate reduction (denitrification) is considered the most appropriate candidate for ground improvement applications. Due to gas compressibility, even a small fraction of gas is sufficient to reduce pore fluid bulk stiffness and increase cyclic resistance.

However, when there is a large amount of gas formation, upward migrating gas can become trapped below low permeable soil layers, which can reduce the effective stress to zero and cause potential instability under low confinement conditions. In order to estimate the expected gas saturation and related mechanical response due to biogenic gas, a theoretical model is presented based on a thermodynamic approach, metabolic stoichiometry, basic laws of physics, and equilibrium chemistry. This approach shows that gas saturation depends on pressure conditions and converted amount substrates. Further investigation is required to validate the proposed model, evaluate the mechanisms of production, migration, and retention of biogenic gas, and optimize the treatment procedures for varying soil types and pressure conditions.

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