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Review Article

The Impact of Calcium Chloride in Cementation Solution on Microbial Induced Calcite Precipitation: A Systematic Review

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ABSTRACT

This review aims to quantify the impact of calcium chloride in cementation solutions on Microbial Induced Calcite Precipitation (MICP). Specific soil strength properties, such as the Unconfined Compressive Strength (UCS) test, permeability (k) and calcium carbonate content of the soil, form the basis of quantifying the test results. Relevant articles from various online databases such as Scopus, Science Direct, ProQuest Dissertations and Theses Global (PQDT), Mendeley and Google Scholar are obtained with search strings of suitable keywords. The Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) were used to screen and select related articles based on exclusion and inclusion characteristics. This review shows a positive correlation between calcium concentrations and soil strength properties, where higher concentrations of calcium solutions induce stronger bonding between soil particles due to better calcite precipitation. However, we also note a reversed correlation when the concentration of calcium solutions is higher than 1 M. This review also verifies that the MICP process enhances soil strength using optimum calcium chloride concentration to avoid soil brittleness. This result benefits other fields, such as agricultural and soil engineering.

Keywords: Calcite, calcium chloride, permeability, polymorph, unconfined compressive strength, vaterite

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INTRODUCTION

Immense awareness about sustainability in most aspects of life in recent times has influenced the curiosity of researchers worldwide regarding microbial-induced calcite precipitation (MICP) and the bio-cementation process (Chuo et al., 2020). Microbial activity induces and regulates chemical reactions in loose granular soil, solidifying its structure with incorporated tensile strength and a greater density (Duo et al., 2018). The crucial components for MICP application are bacteria, urea and calcium chloride. The bacteria present in the soil secrete specific enzymes for reactions, such as carbonic anhydrase and urease (Wei et al., 2015), to convert specific chemical substances into carbonate ions for MICP. Al Qabany et al. (2012) report that passive precipitation is the most preferred type of MICP technique used in research, where the pH value of the soil system is regularly changed as the hydrolysis of urea is influenced by bacterial activity.

According to Chahal et al. (2011), calcium chloride ($CaCl_2$) acts as a calcium source for the growth of respective bacteria, which are added to the media for calcium carbonate ($CaCO_3$) precipitation. The existence of calcium ions in the soil system induces the formation of $CaCO_3$, which is established by the presence of $CaCl_2$ in the media (Golovkina et al., 2020; Lapierre et al., 2020). However, the use of commercial $CaCl_2$ in MICP applications can have disadvantages as (i) it is not cost-efficient, (ii) it can lead to a decrement in chemical efficiency, and (iii) it showcases corrosive characteristics (DeJong et al., 2006; Khadim & Zheng, 2017). Chemical efficiency in MICP is defined as the amount of precipitated calcite compared to the amount of pure chemical substances like urea and $CaCl_2$ percolated into the soil. However, Al Qabany et al. (2012) report that the best input rate of chemical efficiency with the use of pure urea and analytical grade $CaCl_2$ in creating MICP condition can reach up to 0:084 mol/L/h. However, it can drop to half the rate even within similar conditions. These findings indicate that there is no guarantee of high efficiencies of MICP even with commercial $CaCl_2$.

There are still questions with inconclusive answers in MICP research studies, specifically on the impact of calcium chloride in the cementation solution in MICP. This systematic review thus intends to answer related questions by searching for alternative calcium sources such as mollusc shells, limestone powder, seawater and eggshells. This systematic literature review aims to explore more sustainable methods for MICP to be used in the future by setting the objectives to compare the impact of $CaCl_2$ on two properties of MICP, namely, (i) permeability values test and (ii) the Unconfined Compressive Strength (UCS).

MATERIALS AND METHODS

Literature Search Strategy

This study is focused on research on MICP processes that induce soil strength properties. The primary literature search was done using Scopus, Science Direct, Mendeley, Google Scholar, ProQuest Dissertations and Theses Global (PQDT) online databases. Search strings for terms in the title, keywords, or abstract were employed using the Boolean operators "AND" or "OR," as shown in Table 1. The most effective search strategy that minimises irrelevant article results was found to be limiting search results to titles and abstracts. Publications published between 2011 and early 2021 were found using this search strategy, which also maintained data accuracy.

Table 1

Search strategy and search string terms used in PRISM

Num.	Subject	Search string
1.	Microbial Induced Calcite Precipitation (MICP)	'Microbial induced calcite precipitation' 'Microbial induced calcium carbonate precipitation' 'Biocementation' 'Biomineralization'
2.	Calcium	'Calcium' 'Calcium chloride influence' 'Calcium chloride factor' 'Calcium shell' 'Calcium powder'
3.	Soil Strength Properties	'Soil strength properties' 'Mechanical properties' 'Hydraulic properties' 'Unconfined Compressive Strength (UCS)' 'Permeability'

Screening Process

Inclusion criteria were used to finalise the research articles to fulfil the objectives of this study. Hence, published studies that did not meet the criteria from previous studies and review papers were excluded. A total of 300 published studies were screened by title, abstract and full-text evaluation. Briefly, 98 % of the screened English-language articles were rejected in the screening phase, and only six were included in the final screening stage. The search results were filtered based on the inclusion criteria throughout the screening process. The published articles were included if they (i) used the MICP mechanism, (ii) used different calcium concentrations, (iii) used alternative calcium sources, (iv) reported Unconfined Compressive Strength (UCS) test data, (v) reported permeability test data, (vi) reported $CaCO_3$ content data, and (vii) reported polymorph produced. The PRISMA is shown in Figure 1, where all summaries of steps and exclusion explanations used to limit the search results further are included.

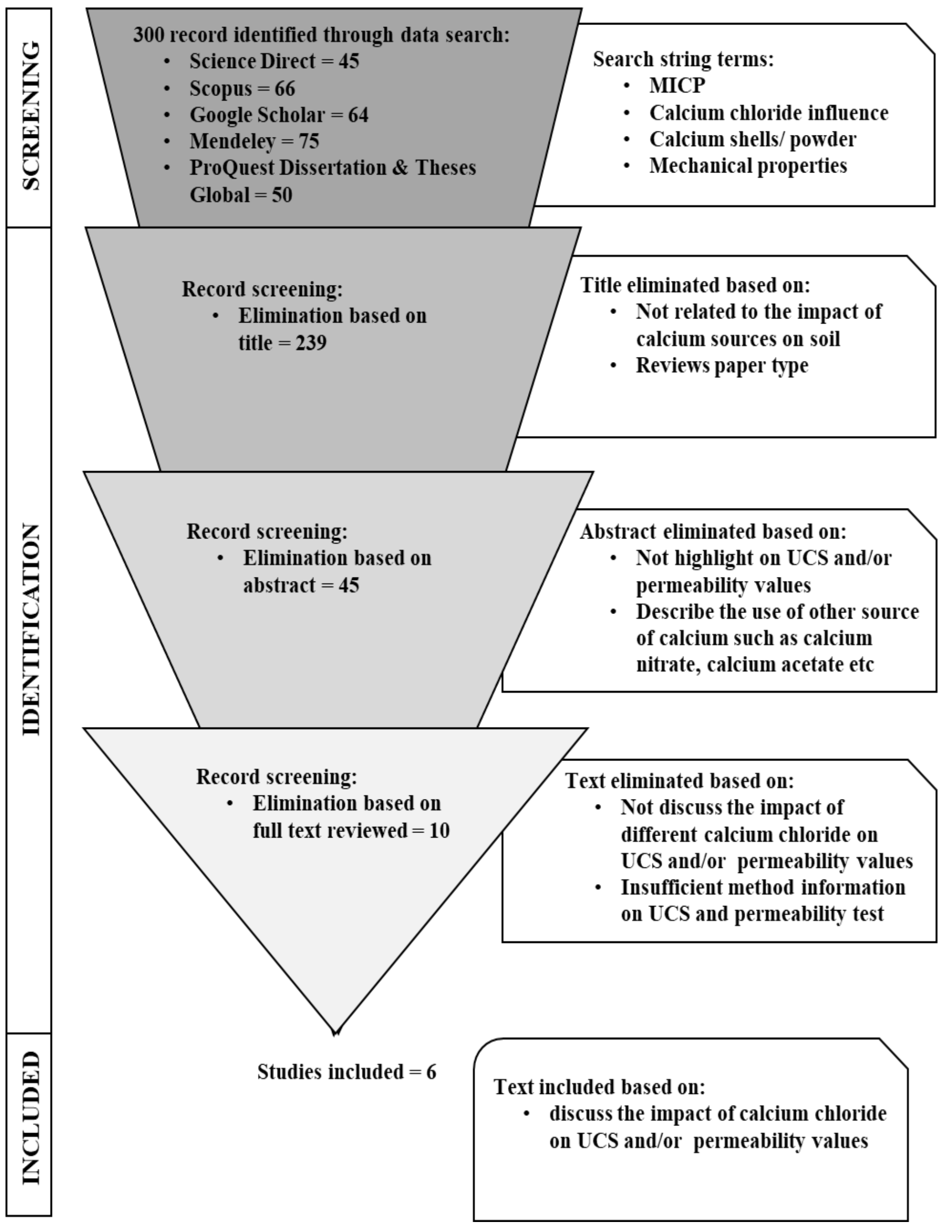

Figure 1. Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) for review on the impact of calcium chloride on MICP in soil

Data Extraction

A total of 6 publications that met the requirements were published in the last decade. All of these were used to evaluate $CaCl_2$'s impact on soil permeability, and the rest were further used to assess the impact of $CaCl_2$ on the Unconfined Compressive Strength. As a result, these articles have qualitative and quantitative data that may be used for further evaluation.

RESULTS AND DISCUSSION

A total of 300 articles were found using primary search string terms in five online databases. The screening process was based on titles, abstracts, and full text. In this process, 239 articles were rejected upon title screening, 45 were rejected upon abstract screening, and ten were rejected upon full-text screening, resulting in only six studies being included in this review. The oldest research article in this study was published in 2013, while the newest one was published in 2019. Four out of the final six articles used *Sporosarcina pasteurii* in their MICP studies, except for Soon et al. (2014) and Cheng et al. (2014), which used *Bacillus megaterium* and *Bacillus* sp., respectively (Table 2). Most of the included studies produced the most stable polymorph of $CaCO_3$, known as calcite, and only two articles reported the production of another polymorph of $CaCO_3$, vaterite, by *Sporosarcina pasteurii* (Liang et al., 2020; Al Qabany & Soga, 2013).

$CaCl_2$ Impact on Soil Permeability

Different concentrations of $CaCl_2$ influence soil particles in MICP treatment differently in terms of soil strength properties. This review extracted and analysed data on specific soil strength properties based on the UCS and permeability tests. Table 2 summarises the permeability test data extracted from selected studies.

Table 2

Types of calcifying bacteria, $CaCl_2$ polymorph, permeability and Unconfined Compressive Strength test values produced in MICP in the soil samples treated with different concentrations of calcium chloride

Studies	Bacteria	$CaCO_3$ Polymorph	Calcium Chloride Concentration (M)	Permeability (m/s)	Unconfined Compressive Strength (kPa)
Soon et al. (2014)	*Bacillus megaterium*	Calcite	1 0.5 0.25	5.1×10^{-8} 0.5×10^{-8} 1.4×10^{-8}	78 130 120
Liang et al. (2020)	*Sporosarcina pasteurii*	Vaterite	0.4	1.12×10^{-4} 1.7×10^{-4} 1.5×10^{-4}	1454 852 984

Table 2 *(Continue)*

Studies	Bacteria	$CaCO_3$ Polymorph	Calcium Chloride Concentration (M)	Permeability (m/s)	Unconfined Compressive Strength (kPa)
Al Qabany and Soga (2013)	*Sporosarcina pasteurii*	Vaterite	1 0.5 0.25	1.7×10^{-5} 5.7×10^{-6} 1.9×10^{-6}	822 1659 1413
Choi et al. (2017)	*Sporosarcina pasteurii*	Calcite	0.3	6.0×10^{-6}	1110
Cheng et al. (2014)	*Bacillus* sp.	Calcite	0.01 M	5.7×10^{-5}	227
Shahrokhi-Shahraki et al. (2014)	*Sporosarcina pasteurii*	Calcite	1 0.5 0.25 0.1	2.6×10^{-5} 1.7×10^{-4} 1.8×10^{-4} 2.1×10^{-4}	240 80 75 50

Generally, the main factors contributing to the permeability coefficient are the soil type, porosity, density, and soil composition (Koestel et al., 2018). The permeability test is one of the most important assessments used to determine the ability of MICP treatment to bind soil particles together. Despite the fact that each paper reported on a different experimental setup and type of sand, all data show that permeability is inversely proportional to $CaCl_2$ concentration up to 0.5 M (Table 2). The lowest measured permeability test value at 0.5 M $CaCl_2$ is 0.5 x 10-8 m/s (Soon et al., 2014), and the highest measured permeability test value at 0.5 M $CaCl_2$ is 5.7.1 x 10-4 m/s (Al Qabany & Soga, 2013). Nevertheless, it depends on the type of bacteria, concentration of $CaCl_2$ and type of $CaCO_3$ polymorph. The gap between adjacent soil structures is reduced at higher concentrations of cementation solution (1.5–2.5 mol/L) based on the $CaCO_3$ that bonds to the exterior of soil particles and clogged pores. More $CaCO_3$ is involved in the consolidating and interconnecting processes, which improves the structural rigidity of the soil system (Duo et al., 2018).

The reduction in permeability value is attributed to $CaCO_3$ polymorph distribution, particularly calcite precipitations at particle-particle interactions, which cause the opening of pores that inhibit water flow (DeJong et al., 2010; Duo et al., 2018). Even though there are no changes in the specimens' extrinsic appearance when MICP is used, the calcite precipitation distribution pattern at the pore level can produce significant permeability values compared to vaterite (Al Qabany & Soga, 2013). Because calcite and vaterite are different solid-state phases of $CaCO_3$, the impact of MICP on permeability varies. Calcite is a thermodynamically stable $CaCO_3$ polymorph that can withstand soil grain pressure

(Ganendra et al., 2014), while vaterite is a minor, metastable, and in a transitional phase to form a much stable polymorph, the calcite (Hua et al., 2007).

Soon et al. (2014) and Al Qabany and Soga (2013) report that the permeability data trend is violated when it reaches a threshold of $CaCl_2$ concentration, specifically at 1 M, and report permeability values of 5.1 x 10^{-8} ms^{-1} and 1.7 x 10^{-5} ms^{-1}, respectively. The permeability data increased as the calcium concentration increased from this threshold value of 1 M of $CaCl_2$. These two articles prove the concept of an optimum cementation solution concentration in enhancing the strength of MICP-treated soil specimens, with the ideal cementation solution concentration ranging between 0.5–1 M of $CaCl_2$. A higher $CaCl_2$ concentration solution led to less homogenous precipitation at both the micro and macro scales (Soon et al., 2014). Although the reported samples were not entirely cemented, the early decline pattern seen in the observed permeability data of the 1 M samples is associated with localised clogging rather than an overall loss in permeability (Al Qabany & Soga, 2013). The use of a greater concentration of cementation solution did not only lead to denser calcite structures but also resulted in a rapid reduction of bacterial activity. The reduction in bacterial activity s explained by the urea compound becoming less abundant in the encapsulated bacteria to catalyse hydrolysis (Al Qabany et al., 2012).

A similar negative impact of calcium concentration beyond the optimum range on bacterial growth was also reported by Chunxiang et al. (2009); whereas calcium ions surround the cell membrane, bacterial enzyme digestion and the consequent changes in the properties of the $CaCO_3$ layer are inhibited with restriction of urea passage. The same trend was reported by Nemati et al. (2005), where an increase in the concentration of $CaCl_2$ led to greater conversion to $CaCO_3$, with the highest conversion of 99 % observed in cultures containing 25 and 30 g/L $CaCl_2$. A concentration of 40 g/L $CaCl_2$ resulted in a lag phase in $CaCO_3$ synthesis with a decreased conversion rate of 80 %. It demonstrates that concentrations of $CaCl_2$ higher than the threshold have an impeding effect on bacterial activity. High quantities of urea hinder bacterial growth, suggesting the vitality of an optimum $CaCl_2$ concentration, as aforementioned in the MICP technique, to improve soil structure (Nemati et al., 2005).

To overcome this problem, Chunxiang et al. (2009) suggested that both cementation solution and bacterial solution be introduced simultaneously since the bacterial cell wall is composed of numerous negative charges (Dardau et al., 2021) and thus, if positively charged ions are introduced first without urea, Ca^{2+} ions spontaneously adhere to the bacterial surface (even in the presence of urea), severely influencing and delaying bacterial activity. Thus, enzyme degradation is hindered no matter when urea is added, as bacterial surfaces would be coated with Ca^{2+}, which impairs urea passage. In summary, the efficiency of the MICP process can be enhanced by using a low concentration of $CaCl_2$ and injecting both $CaCl_2$ and urea solutions at once in the soil sample. On the other hand, Chuo et al. (2020)

recommend higher concentrations of bio-cementation solution or longer treatment cycles to be used in MICP experiments to improve soil liquefaction susceptibility. The same pattern was also seen in the study by Duo et al. (2018), where the permeability coefficient of sand samples treated with 0.5, 1.0, 1.5, 2.0, and 2.5 M cementation solution dropped by 42.6, 71.3, 97.5, 98.6, and 99.1 %, respectively, as compared to traditional aeolian sand through eight treatment cycles.

The Impact of $CaCl_2$ on the Unconfined Compressive Strength

The effectiveness of MICP treatment to bind soil particles together is also evaluated using the UCS test. The highest UCS test value (1659 kPa) was attained when 0.5 M $CaCl_2$ was used in the cementation solution (Al Qabany & Soga, 2013). As with permeability, all experiments from selected articles were conducted in different configurations with various types of sand. However, similar patterns of increasing strength were discovered to be obtained. The combined data from four studies, Choi et al., 2017, Cheng et al., 2014; Liang et al., 2020 and Al Qabany & Soga, 2013, suggest the direct impact of incremental $CaCl_2$ concentrations on the UCS values (Table 2). In a study by Duo et al. (2018), the same trend was reported where the soil samples were treated with 0.5, 1.0, 1.5, 2.0, and 2.5 M solidification solutions for the UCS test with observed values of 1.71, 4.93, 6.64, and 3.69 MPa, respectively.

Although the UCS value reported in selected studies and previous studies varies depending on the type of test soil and $CaCl_2$ concentration, the MICP-treated sand shows an improvement in shear strength compared to the control. Soon et al. (2017) reported a 25–100 % improvement in a blend of coarse and fine grains sand, whereas Lu et al. (2010) published studies on fine sands that improved by 25–120 %. It was proposed that in any type or condition of sand, as long as pores exist between coarse grains, it will create a favourable environment for forming calcite bonds at particle-particle contacts, thereby improving the soil's shear strength (Ng et al., 2013). Most of the carbonate ions produced by urea hydrolysis combine with calcium ions to form $CaCO_3$ polymorphs in the intergranular spaces of soil columns (Okwadha & Li, 2010), leading to an improvement of the shear strength properties. Calcite particles (one of the polymorphs) that cover soil grains and deteriorated calcite crystal fines formed by shearing cause an increase in overall particle roughness, significantly impacting the UCS values (Feng & Montoya, 2016).

They also report that relatively high cementation solution concentrations result in increased calcite precipitation, manifested as large crystal formation on soil particles and localised deposition. Shahrokhi-Shahraki et al. (2015) state that more brittle responses are caused by stiffer, higher solution concentrations where the maximum UCS test value of 240 kPa was achieved in this report with a 1 M $CaCl_2$ concentration in soil capillary pores. The shear strength of samples treated with 0.25 M cementation reagent improved by

26–57 %, while the UCS of specimens treated with 0.5 M reagent improved by 25–69 %. Good correlations with UCS improvement were observed between 1.0 and 2.5 % calcite content (R^2 = 0.87), with the maximum enhancement in the UCS achieved at about 2.5 % calcite content (Soon et al., 2014).

Solutions with higher concentrations of calcium and urea are hypotheses to promote the binding between soil particles due to higher levels of calcite precipitation (Cui et al., 2021). The strength of bio-cemented calcareous soil also appeared to rise as the cementation level increased (Al Qabany et al., 2012). The MICP treatment may significantly improve the rigidity of calcareous soil (Cui et al., 2021) due to calcium carbonate cementation. However, Soon et al. (2014) and Al Qabany and Soga (2013) report that any increment of $CaCl_2$ beyond 0.5 M will reduce the UCS value to 40 % and 50 %, respectively. Studies by Al Qabany and Soga (2013) and Soon et al. (2014) showed a decrement in UCS test values when a 1 M calcium chloride solution was used in their experiments. When a high-concentration treatment (1 M) was employed, they observed a less homogenous calcite deposition pattern with bigger crystal sizes.

This proposed pattern was supported by Velpuri et al. (2016), as the calcite precipitation rate is affected by calcium concentration. Calcite precipitation occurs with increasing calcium ion concentration and remains relatively unchanged under specified urea and bacterial conditions. Nevertheless, with increasing calcium ion concentration levels, injection blockage became more severe, which made it harder to achieve uniformly cemented soil samples. Sheikh and Atmapoojya (2022) also indicated that crystal growth grows in size as the concentration of the cementation solution increases, yet inhibiting a homogeneous precipitation process. Al Qabany and Soga (2013) reported that the increased sample strength observed while using a low-concentration solution is linked to a better distribution or bigger proportion of deposition at particle interactions, resulting in more uniform cementation in the samples. Whiffin et al. (2007) created cemented samples with a 1 M solution, and when compared to the 0.25 M data (Al-Qababy et al., 2012), the strength values of the 1 M samples are lower. It is worth noting that Whiffin et al. (2007) work on the cemented core samples, as compared to Al-Qabany et al. (2012) studies on non-cemented samples. It is because the homogenous distribution of precipitated calcium carbonate all a uniform layer surrounding the soil particles, which leads to a higher UCS value (DeJong et al., 2010; Gebru et al., 2021).

It is also reported that inconsistently sized $CaCO_3$ distribution patterns with bigger crystal sizes are formed when the cementation solutions with higher $CaCl_2$ concentrations are used (Soon et al., 2014). It could support the promising results (better soil strength properties) obtained with MICP processes that used alternative calcium sources at low calcium concentrations compared to the commercial $CaCl_2$ (1 M). Figure 2 illustrates different sizes of $CaCO_3$ deposition in soil structures at various chemical concentrations.

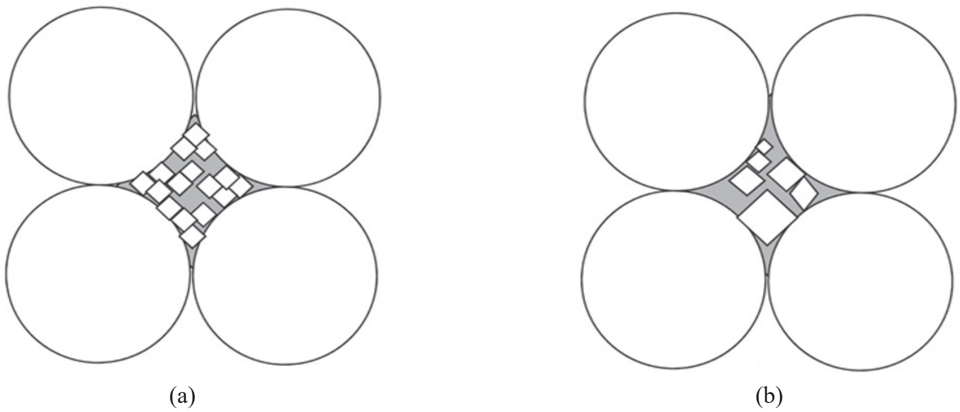

Figure 2. Pore blockage as an outcome of MICP utilising (a) a lower chemical concentration and (b) a higher chemical concentration is depicted in a conceptual illustration (Al Qabany & Soga, 2013)

The unfavourable effect of the 1 M cementation solutions on soil properties was also reported by Soon et al. (2014), where no discernible difference in shear strength or hydraulic conductivity was recorded post-treatment. Additionally, measurements of ammonium concentration and pH values indicated no observable urease activity. This claim is also supported by Whiffin (2004) during the first 8 hours of the cementation process in the experiment, when a two-fold rise in calcium concentration reduced urease activity by 10 %, with no impact beyond that. Al Qabany and Soga (2013) also noticed that the precipitation patterns indicated that greater amounts of cementation solution not only lead to thicker calcite structures but may also result in a rapid reduction of bacterial activity as urea becomes less accessible to the enclosed bacterial cells to catalyse hydrolysis. Therefore, it was proposed to use a minimal cementation solution that can be applied with uniform $CaCO_3$ deposition and many nucleation spots to gradually raise the consolidation solution to minimise the number of injections. Although this strategy may sound theoretically relevant, a further experimental study is required to demonstrate its efficacy.

In contrast, a controversial strategy has been proposed for applying $CaCl_2$ and urea to enhance the MICP process based on supersaturation. Bosak and Newman (2005) proposed that smaller deposits of $CaCO_3$ can be achieved through rapid nucleation by using higher cementation solutions and bacterial concentrations. Al-Thawadi et al. (2012) tested this concept and reported that ureolytic bacteria help precipitate calcite crystals as nanocrystalline clusters (specifically smaller particles) when exposed to high urea and calcium ions. These particles will enhance the solidification of soil structures by constructing bridge points between soil grains. Smaller $CaCO_3$ particles are thus formed due to the rise in supersaturation index (SI) in the soil system, leading to smaller calcite crystal formations (De Muynck et al., 2010; Mujah et al., 2019).

CONCLUSION

The $CaCl_2$ concentrations observed are directly proportional to soil strength properties. Greater concentrations of $CaCl_2$ lead to a rapid loss in permeability and elevate the UCS values. However, an optimum range of $CaCl_2$ concentrations in cementation solutions needs to be observed, as higher $CaCl_2$ concentration above a threshold will lead to localised clogging and inhibition of bacterial activity. Consequently, the heterogeneity of the calcite distribution in the soil will be disturbed with a reduction in the soil strength.

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