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Organic-Inorganic Hybrid Green Materials for Soil Improvement

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Synonyms

Bio-cementation; Hybrid biomaterial; Polymer; Soil improvement; Urease

Definition

Organic inorganic hybrid green material produced from polymer modified urease-based bio-cementation is a sustainable grouting material for soil improvement, posing numerous benefits over conventional grouts.

Background

Soil is one of the most important civil engineering materials, and almost all of the civil engineering structures rest on the soil. Therefore, the loads/stresses coming from the structures are directly transferred to the soil, and the soil should have enough capability to bear all the stresses without any failures. In another words, the soil should have enough bearing capacity and lower settlement to ensure the safety of the structure. However, the soils with good engineering characteristics cannot be found everywhere in the earth, thus weak soils are also in need to be used for the constructions due to the lack of the suitable lands owing to the rapid population growth and industrialization. Therefore, it is mandatory to improve the physical properties of the soil to the required level before the constructions. The process is called as ground improvement, and most of the currently available ground improvement methods are not sustainable and eco-friendly. Among the available ground improvement methods, vibro-compaction, and dynamic compaction are extensively used to densify the gravelly soils, and associated undesirable vibration can cause several other problems (Sarker and Abedin 2015). Grouting is another type of widely used treatment method, which is mainly applied on fine-grained soils. Among the available grouting techniques, cement and chemical grouts are commonly used to upgrade the soil properties. However, due to the associated several drawbacks, use of these grouting techniques is not always sustainable. Mainly, cement and chemical grouts are not environmentally friendly. Even though the cement is recognized as one of the best engineering materials due to its higher

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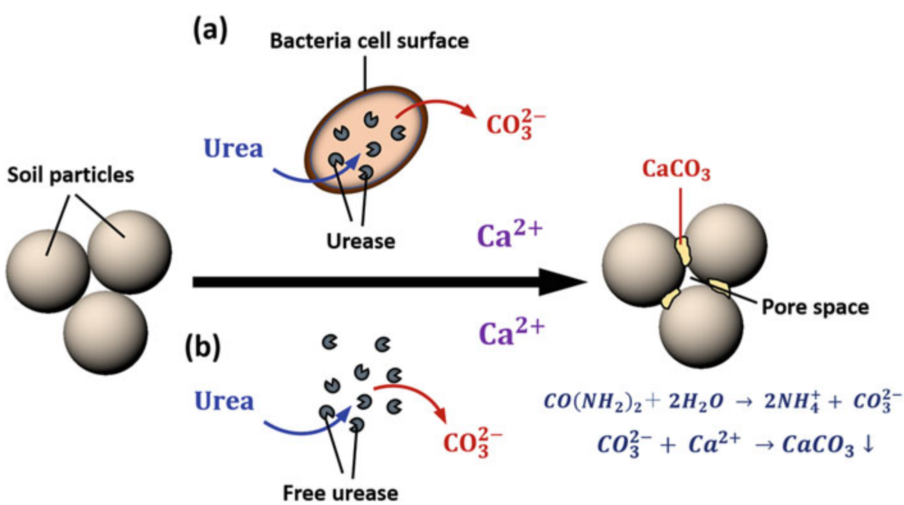
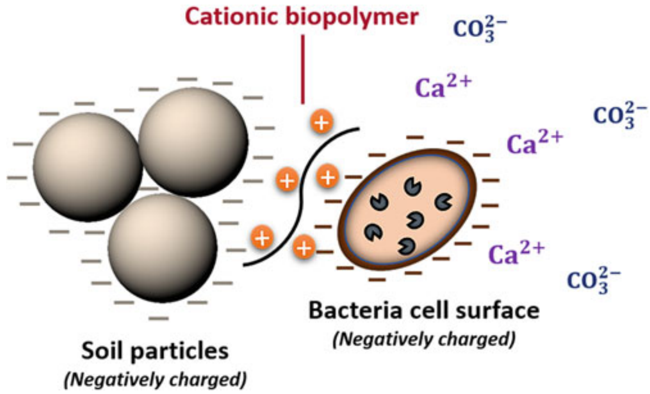
strength, durability, and workability (Chang et al. 2016; Daraei et al. 2018; Kumar et al. 2020), it largely contributes to the greenhouse gasses emission, particularly carbon dioxide (CO_2) is released to the environment during the cement production, and which can cause for the global warming and several other environmental impacts. Each one ton of cement emits nearly 0.95 tons of CO_2 during the cement manufacturing, and worth to note that the CO_2 emission due to the geotechnical applications is 2% from the total CO_2 from cement (Chang et al. 2016). Similarly, most of the chemicals used for the grouts are very toxic for the human and animals. Specially, grouts containing acrylamides and polyurethane hugely damage the environment (DeJong et al. 2010; Ivanov and Chu 2008).

Due to the above-mentioned issues, biological approaches to generate bio-grouting materials have gained much attention recently as an eco-friendly and sustainable technique to treat the weak soils. Among them, polymer modified urease-based bio-cementation has been recognized as a more sustainable and eco-friendly method to produce organic-inorganic biomaterials. Urease-based bio-cementation is a biogeochemical process, which produces calcium carbonate ($CaCO_3$) artificially. Process is catalyzed by the enzyme urease. Urease enzyme has a capability to hydrolyze urea and produce ammonia and bicarbonate ions. Bicarbonate ions instantaneously give $CaCO_3$ in the presence of Ca^{2+} ions under the alkaline condition (Nawarathna et al. 2019; Fujita et al. 2017; Mortensen et al. 2011). Urease is a multi-subunit nickel-containing enzyme (Holm and Sander 1997), mainly found in some bacteria species called ureolytic bacteria and the process is then called as microbial-induced carbonate precipitation (MICP) (Wei et al. 2015; Gowthaman et al. 2019; Nawarathna et al. 2019; Barabesi et al. 2006). At the same time, free enzymes can also be extracted from the plant species, and the process is called as enzyme-induced carbonate precipitation (EICP) (Sirko and Brodzik 2000; Hamdan et al. 2016; Zhao et al. 2016). The mechanisms of the urease-based bio-cementation are graphically illustrated in Fig. 1.

Biogenic $CaCO_3$ produced by urease-based bio-cementation has an extreme ability to improve the properties of the weak soils and several investigations have proved that it can successfully work on the ground improvement, slope stability, erosion prevention, bioremediation, crack healing, etc. (Van Paassen 2009; Gomez et al. 2014;



Organic-Inorganic Hybrid Green Materials for Soil Improvement, Fig. 1 Urease-based bio-cementation processes. (**a**) Urease producing bacteria hydrolyses the urea to form $CaCO_3$ in the presence of calcium ions – MICP. (**b**) Extracted free enzymes hydrolyzes the urea to form $CaCO_3$ within the soil matrix- EICP



Organic-Inorganic Hybrid Green Materials for Soil Improvement, Fig. 2 Urease-based bio-cementation with cationic biopolymer. Improve the bio-cementation process by efficient formation of $CaCO_3$

Chou et al. 2011; Montoya and DeJong 2015; Gowthaman et al. 2019; Wang et al. 2017).

Even though the biomaterial produced from urease-based bio-cementation has enough capability to improve the weak soils, the associated brittleness reduces the efficiency of the process (Rahman et al. 2020). By incorporation of the organic biopolymer materials to the urease-based bio-cementation process (as illustrated in Fig. 2), the brittleness of the treated soils can be reduced, and the tensile strength of the sample can be increased. The process is designated as polymer modified urease-based bio-cementation, and that produces organic-inorganic biogenic green materials. In this entry, a short review on the use of polymer modified urease-based bio-cementation to produce organic-inorganic green materials for soil improvement is presented.

Polymer Modified Urease-Based Bio-Cementation

It has been proved that by adding polymer into the urease-based bio-cementation, organic-inorganic hybrid green material can be formed and can be applied successfully for the soil improvement. The concept is inspired from the nature and closely related to the organic matrix-mediated biomineralization. One of the best examples available in nature for the organic matrix-mediated biomineralization is exoskeleton of the crustacean, wherein inorganic $CaCO_3$ has been deposited on the organic chitin and give higher strength for the cuticle (Raabe et al. 2005). Therefore, by incorporating organic materials into the urease-based bio-cementation, organic-$CaCO_3$ hybrid green material with excellent physical properties can be produced.

However, several factors should be considered when selecting organic materials for the urease-based bio-cementation. Basically, organic materials should not interfere with the urease enzyme activity and should not disturb the nucleation and growth of the $CaCO_3$. According to literature, several polymers have performed well with urease-based bio-cementation. Different studies have been carried out earlier to investigate the efficiency of this hybrid green material on soil improvement using MICP and EICP.

Green Materials from Polymer Modified MICP and Applications on Soil Improvement

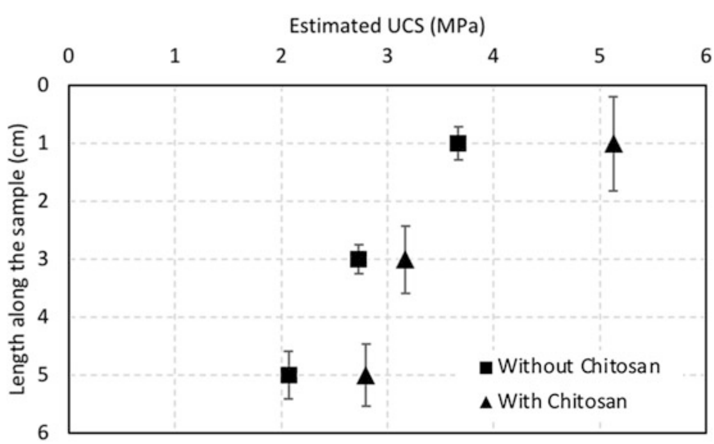
Polymer can have several impacts on the bacterial growth and enzyme activities. Therefore, selection of a suitable polymer is very important for polymer modified MICP. Due to these difficulties, limited number of researches have been carried out earlier to investigate the effect of polymer modified MICP for green material formation and soil improvements.

Nawarathna et al. (2018) could successfully produce organic-$CaCO_3$ hybrid green material by introducing synthetic polymer, poly-L-lysine into the MICP. Poly-L-lysine is a cationic polypeptide, which is charged positively in neutral pH (Dzakula et al. 2009) and mainly used as a cell adhesion reagent in biomedical field (Mazia et al.

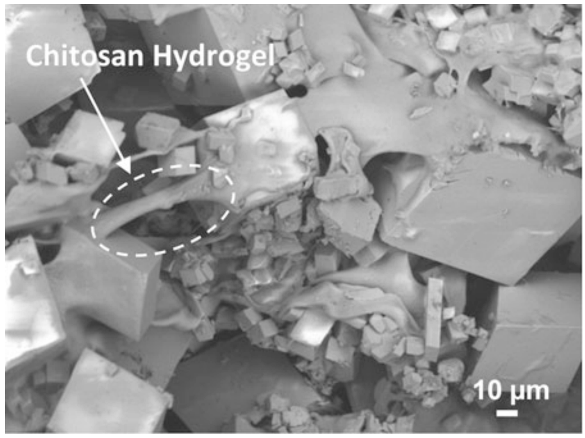
1975). Nawarathna et al. (2018) have produced biogenic $CaCO_3$ with and without poly-L-lysine by using ureolytic bacteria, *Pararhodobactor* sp. in the presence of $CaCl_2$. $CaCO_3$ formation efficiency has increased with poly-L-lysine, and this organic-$CaCO_3$ hybrid green material has peanut-like twin sphere-shaped morphology compared with the inorganic $CaCO_3$. This is due to the conformation change of the poly-L-lysine chain from random coil conformation to α-helix conformation under the alkaline conditions. Further, by adding poly-L-lysing to the MICP, well-cemented sand specimen has been obtained than without poly-L-lysine and unconfined compressive strength (UCS) has increased by 31% than the control sample (only with MICP).

Wu and Zeng (2017) have introduced sodium alginate polymer to the MICP and produced biogenic green material. Sodium alginate is a natural polysaccharide, which is mainly extracted from the brown sea weeds (Butler et al. 2006). Due to the gelling property of the sodium alginate, bacteria cells were immobilized and formed $CaCO_3$ on the sodium alginate gel. Polymorphism of the $CaCO_3$ crystals with sodium alginate has changed drastically with the time. Initially, it formed spherical vaterite crystals, and after sometime it completely transitioned into dumbbell shape calcite crystals. Authors have explained that due to the presence of the sodium alginate stabilizer, initially it formed vaterite crystals, and with time, it converted to calcite due to thermodynamic instability of the vaterite. Vaterite is a metastable form of $CaCO_3$ and easily transformed to the stable form of aragonite or calcite with time (Kralj et al. 1997; Lopez et al. 2001). Further, morphology of the $CaCO_3$ changed drastically with the sodium alginate concentration. Lower concentration of sodium alginate, hexagonal-shaped crystals have formed and capsule-shaped crystals have formed at higher polymer concentrations due to the higher negative charge on the gel surface (Wu and Zeng 2017).

Another natural cationic polysaccharide chitosan also has been used for MICP to create hybrid green materials (Nawarathna et al. 2019). Chitosan is derived from the chitin, which is mostly available in exoskeleton of the crustacean, marine diatoms, and in some algae (Nisticò 2017; Yang 2011). It is a cationic polysaccharide, which is charged positively in the diluted acidic environments (pH < 6.5) (Liu et al. 2015; Wang and Heuzey 2016). Nawarathna et al. (2019) have found that by adding chitosan into the MICP process, hybrid green material can be produced, and it upgrades the $CaCO_3$ formation efficiency by offering additional nucleation sites. Calcium ions can attach to the chitosan hydrogel due to acid–base reaction and can produce more nucleation sites. Laboratory-scale sand solidification experiments have been conducted by using injection method with and without addition of the chitosan with the MICP process. Chitosan-$CaCO_3$ green material assisted to improve the UCS of the sandy soil through effective filling of the pore spaces. As shown in Fig. 3, the strength



Organic-Inorganic Hybrid Green Materials for Soil Improvement, Fig. 3 Variation of the UCS value of the MICP treated samples with and without chitosan. Strength of the samples have decreased from top to bottom of the pecimens



Organic-Inorganic Hybrid Green Materials for Soil Improvement, Fig. 4 SEM image of the $CaCO_3$ precipitate with chitosan

of the top of the specimen has increased by 40% by adding chitosan into the MICP than without chitosan. The strength has reduced from top to bottom due to the accumulation of the bacteria, cementation solution, and chitosan at top of the specimen. On the other hand, chitosan hydrogel itself assisted to form a better bridge between the soil particles as obtained from the scanning electron microscope (SEM) images given in Fig. 4.

Polyvinyl alcohol (PVA) has been used as an organic additive for the MICP to form hybrid green materials (Wang and Tao 2018). PVA is a synthetic polymer with excellent adhesive and emulsifying property. Wang and Tao (2018) have found that PVA has an ability to upgrade the $CaCO_3$ formation and vaterite crystals are dominant in the precipitate. The higher the absorption capacity of the PVA, the more favorable the condition to form vaterite crystals than calcite (Kim et al. 2004). During sand solidification, sand treated with PVA modified MICP has been obtained higher strength compared with the sand treated with only MICP (Wang and Tao 2018).

Sun et al. (2022) have been applied the polyacrylamide (PAM)-treated MICP for surface erosion prevention of loess-slopes. MICP-PAM-treated slopes showed better resistance for the erosion than the slopes only treated by MICP. They have concluded that optimum PAM concentration to achieve excellent erosion resistance and higher surface strength is 1.5 g/L. Addition of PAM assisted to maintain stronger resistance to tension and shear forces.

Green Materials from Polymer Modified EICP and Applications on Soil Improvement

Compared with the MICP, EICP has several positive points. When it comes to the field applications of MICP, culturing and growth of bacteria are tedious procedures and need more time, special equipment, and careful observations. Therefore, use of the extracted urease enzyme from plants would be a more sustainable approach than using whole cell urease. Further, Similar to the MICP, by adding polymer to the EICP, hybrid green material can be produced. Some research works can find in literature the use of this hybrid green material for soil improvement.

Xanthan gum is an anionic polysaccharide, which is produced by the fermentation of sucrose and glucose (Kosanovic et al. 2017). Xanthan gum does not produce its hydrogel under normal gelation procedures, and it shows subsequent annealing and cooling-induced gelation (Iijima et al. 2007). Xanthan gum has been used as an additive to enhance the performance of the EICP process, mainly due to its gelling ability and favorable nature for the $CaCO_3$ crystallization. Hamdan et al. (2016) have investigated the effect of the xanthan gum on the $CaCO_3$ crystallization and soil stabilization using jack bean urease. They concluded that xanthan gum hydrogel did not adhere with the enzyme activities and $CaCO_3$ precipitate. Further, xanthan gum modified EICP extended the reaction time and increased the precipitation efficiency. And also, xanthan gum hydrogel is capable of creating strong interaction with the water molecules, and it holding water molecules tightly and preventing significant evaporation.

Pasillas and research group have studied the performance of the viscosity-enhanced EICP solution for high permeable (Ottawa 20/30) and low permeable (F-85 sand) sand using xanthan gum (Pasillas et al. 2018). Experiments were conducted for unenhanced EICP solution and the xanthan gum-enhanced EICP solution under soaked and drained conditions. Sand column treated with xanthan gum modified EICP shows the highest water retaining ability. Therefore, xanthan gum-$CaCO_3$ hybrid material performed well in soil improvement than only using biogenic $CaCO_3$.

Zhao et al. (2016) have introduced poly (acrylic acid) to EICP process due to its non-toxicity and the favorable nature of the $CaCO_3$ crystallization in order to produce hybrid green materials. Poly (acrylic acid) is an anionic synthetic polymer, which is widely used in disposable diapers due to its extensive water absorption ability. It is also used in ion exchange resins, adhesives, and detergents. Zhao and his team have used Jack bean urease to treat the sand specimens, and experiments were conducted with and without poly (acrylic acid) under different experimental conditions. Strongly cemented sand specimen have been obtained with the incorporation of the poly (acrylic acid) than the conventional EICP. Sand specimen treated with poly (acrylic acid) modified EICP could be sustained even under pressure as high as 4.8 MPa, while for sand treated with EICP the pressure is 64 KPa. Porous hydrogel network created by poly (acrylic acid) acts as a binder to bind soil grains effectively by forming polymer calcite composite. And also, excellent water retaining capacity of poly (acrylic acid) makes it more efficient for $CaCO_3$ precipitation. The most interesting behavior of poly (acrylic acid) is its ability to absorb NH_4^+ ions and it can remove the NH_4^+ ions from the reaction system very effectively. Therefore, adding poly (acrylic acid) into the EICP make it more eco-friendly, which provide a hint to apply it into the MICP process since generation of the NH_3 is one of the long-existing disadvantage in the EICP/ MICP process.

Chitosan also has been used to produce organic-$CaCO_3$ green materials by incorporating to EICP (Nawarathna et al. 2018). As explained previously during the $CaCO_3$ precipitation, chitosan hydrogel acts as a nucleation site for $CaCO_3$ crystals to nucleate and growth. In detail, calcium ions can absorb into the chitosan hydrogel by hard acid hard base reaction. These embedded calcium ions provide nucleation sites for $CaCO_3$ crystals to nucleate. Therefore, adding chitosan upgrades the performance of the EICP process by providing additional nucleation sites, and on the other hand, chitosan hydrogel would be acting as a binder during soil stabilization.

Polyvinyl alcohol (PVA) fiber was used to improve the efficiency of the EICP process previously (Yuan et al. 2021). The length and content of the PVA fiber greatly affect the strength of the treated soil. Compared with the fiber content, fiber length has significant influence on the UCS and calcium carbonate content. Yuan and his team concluded that the optimum length and content of the fiber to achieve a better strength are 9 mm and 0.4% by sand weight. By adding PVA fiber to EICP, UCS of the treated sand could be increased by 84% and $CaCO_3$ content by 36%. Due to the deposition of the $CaCO_3$ on the fiber surface, fiber-$CaCO_3$-sand network has formed. Due to the overlapping of the fiber, 3-D mesh is formed, and it prevents the displacement and deformation of the sand while increasing the strength.

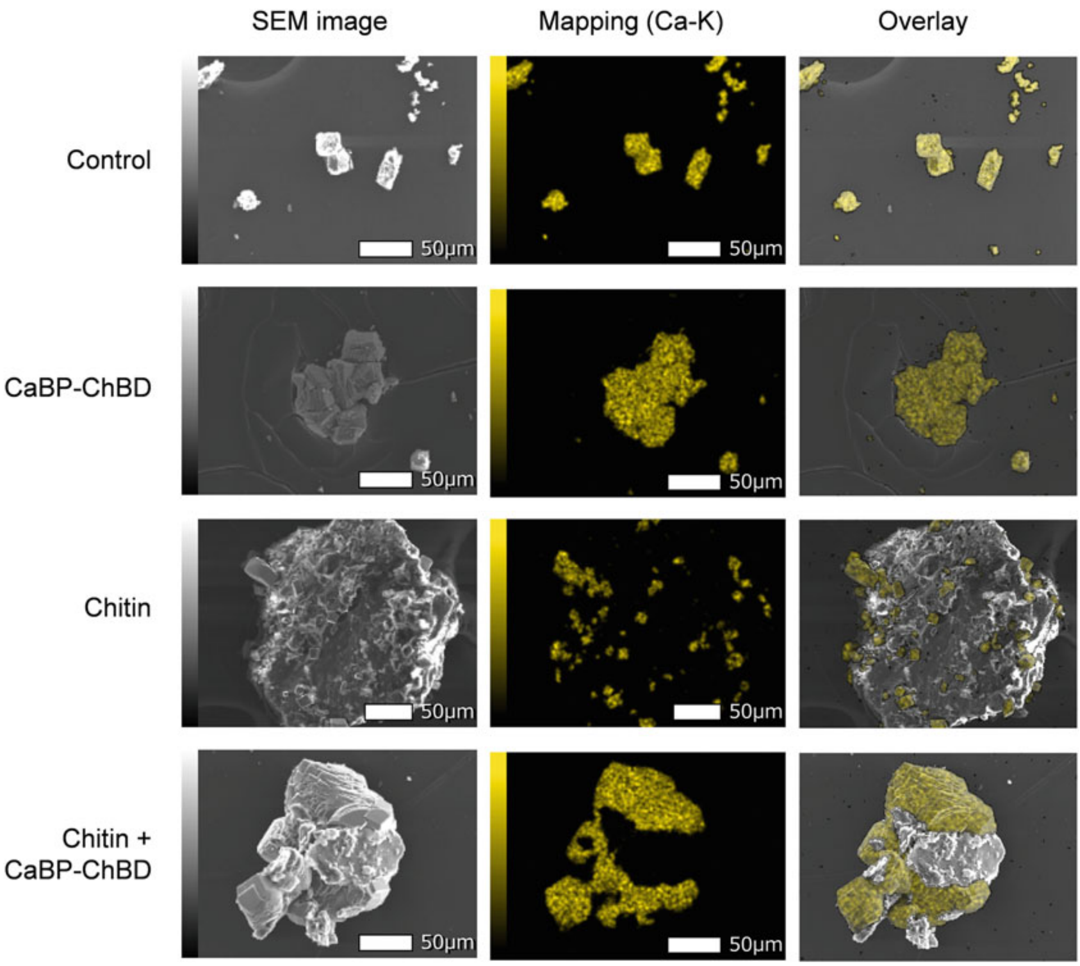
Similarly, Wu et al. (2021) have investigated the effect of the PVA modified EICP for aeolian sand solidification and analyzed the strength, wind, and water erosion resistance. They found that 3% (by weight) of the PVA is the optimum concentration, which gives higher strength. The surface soil treated with PVA + EICP has a greater resistance to wind erosion and water erosion. During the model tests, accumulated soil weight percentage after 120 min of exposure to water were 0.03% for slopes treated with EICP+-PVA and 71.13% for the slopes treated only with the EICP. By adding PVA, organic-$CaCO_3$ has been formed, and it generated a film with a network structure, which helped to make a good bridge between the soil particles.

Almajed et al. (2020) have introduced sodium alginate to the EICP for the prevention of wind erosion of the desert sand in Saudi Arabia. They could obtain a better prevention of the erosion by introducing sodium alginate with EICP than with the EICP alone. Thick crust has been formed at the surface of the treated soil, and the UCS of the crust increased with the increase of the sodium alginate concentration due to the formation of the cross-linking network by exchange of the sodium ions with the divalent calcium ions. Similarly, applicability of the polymer modified EICP for rainfall erosion prevention has been studied by Sun et al. (2021) by using polyvinyl acetate (PVAc) and polyethylene glycol (PEG) with EICP to a dust soil sample. Sample treated with EICP-PVAc or EICP-PEG could improve the shear resistance and rainfall erosion resistance. However, surface strength has reduced with repeated rainfall. Adding both PVAc and PEG with EICP exhibited good shear resistance and higher surface strength for repeated rainfall. Combination of the 50 g/L of PVAc and 30 g/L of PEG has been found as the best combination to obtain better dust control and repeated rainfall-erosion durability.

Very recently, Nawarathna et al. (2021) have developed an artificial fusion protein to facilitate the formation of organic-inorganic hybrid green material by efficient precipitation of $CaCO_3$ on the chitin matrix using EICP process. Fusion protein (CaBP-ChBD) has been fabricated by introducing a short sequence calcium-binding peptide (CaBP) to the chitin-binding domain (ChBD). $CaCO_3$ precipitation experiments have been conducted by hydrolysis of urea using jack bean urease, and the amount of the $CaCO_3$ precipitate has been increased drastically in the presence of fusion protein compared with the control sample. The authors mentioned that the presence of a higher number of basic amino acid residues in both of CaBP and ChBD would be the prime reason for the excellent performance of CaBP-ChBD in $CaCO_3$ formation. Further, $CaCO_3$ was efficiently formed on the chitin in the presence of the CaBP-ChBD as given by energy-dispersive X-ray spectroscopy (EDX, Fig. 5). Most interestingly, this hybrid green material could perform well during sand solidification. By introducing both CaBP-ChBD and chitin to the urease-based bio-cementation, well-cemented sand specimen with higher UCS value and fracture resistance could be obtained.

Concluding Remarks

Bio-grout has been recognized as a more sustainable and eco-friendly ground improvement method over the conventional grouting methods. Most of the conventional grouting methods are not environmentally friendly and emit large amounts of greenhouse gases, indirectly contributing to the global warming and several other environmental issues. Among the bio-grouting techniques, polymer modified urease-based bio-cementation has gained much attention recently as a more sustainable approach to treat the weak soils. By incorporating polymer into the urease-based bio-cementation, organic-inorganic hybrid green material with better physical properties is formed and can be applied effectively to improve the properties of the weak soils. Further, it has the capability to reduce the brittleness of the samples treated with urease-based bio-cementation. Several synthetic and natural polymers have been used successfully with both the MICP and EICP, indicating the capacity of polymer modified urease-based bio-cementation in producing organic-$CaCO_3$ hybrid green material, which can readily be applied on weak soils.



Organic-Inorganic Hybrid Green Materials for Soil Improvement, Fig. 5 EDX analysis of the $CaCO_3$ precipitate with and without organic materials

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