Green biocement utilizing enzyme-induced carbonate precipitation with waste coffee grounds

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Abstract

This study explores a novel method of biostabilization using crude enzyme-induced carbonate precipitation assisted by kitchen waste coffee grounds. As one of the most widely consumed beverages globally, coffee generates substantial amounts of solid waste annually in the form of coffee grounds. Improper disposal of these grounds can lead to environmental pollution. Our findings indicate that coffee ground extracts can maintain the reaction system at a pH below neutral during the solidification process, facilitating the conversion of ammonia to ammonium ions and thereby reducing ammonia emissions. When a 0.2 g/mL concentration of coffee ground extract was added, the pH of the reaction system in the consolidation test was 6.89 after a single injection, with the unconfined compressive strength (UCS) reaching 385 kPa. Additionally, further research and validation are needed to explore the potential for improved uniformity and effectiveness through multiple treatments. The application of coffee grounds aligns with the principles of a circular economy, enhancing cost efficiency by reducing waste management costs. This study offers a new perspective on the green application of biocement.

Keywords: *Enzyme-Induced Calcium Carbonate Precipitation (EICP); Ammonia emission; Waste recycling; Soil improvement.*

1. Introduction

In recent years, biocement has garnered significant attention as a novel geotechnical engineering technology developed at the intersection of biogeochemical processes[1–3]. One pivotal reason for this attention is its superior sustainability compared to traditional geotechnical engineering techniques, as it mitigates substantial energy consumption and pollution, particularly the emissions associated with the calcination process in traditional cement production[4–6]. However, despite the potential of biocement to improve geotechnical properties and thereby achieve desired engineering applications, such as preventing coastal erosion[7–9], preserving heritage[10–12], enhancing recovery rates[13–15], reinforcing foundations[16–18], suppressing wind erosion[19–21], and stabilizing slopes[22–24], its practical scalability remains constrained. Particularly, in reactions involving urea as a substrate, biocement releases significant amounts of ammonia as illustrated in Equations 1—3, posing challenges to on-site construction and leading to various environmental issues including haze pollution, nitrogen deposition, soil acidification, eutrophication, and loss of biodiversity[25–27].

$$CO(NH_2)_2 + 2H_2O \xrightarrow{\text{enzyme}} H_2CO_3 + 2NH_3 \qquad \textbf{(1)}$$

$$H_2CO_3 \leftrightarrow HCO_3^- + H^+ \leftrightarrow CO_3^{2-} + 2H^+ \qquad \textbf{(2)}$$

$$Ca^{2+} + CO_3^{2-} \longrightarrow CaCO_3 \qquad \textbf{(3)}$$

$$NH_3 + H^+ \longrightarrow NH_4^+ \qquad \textbf{(4)}$$

Feasible methods to alleviate this issue include pH regulation to convert ammonia into ionic states, as illustrated in Equation 4. Currently, in the field of agriculture, where the largest ammonia emissions occur, large-scale acidification of slurry with sulfuric acid has proven effective in reducing ammonia emissions[28–32]. Presently, in biocementation processes, adjusting pH with hydrochloric acid in Calcium Phosphate biocement can suppress ammonia emissions[33–37], while the use of hydrochloric acid in Microbially Induced Calcium Carbonate Precipitation (MICP) reduces ammonia emissions by more than 90%[38], and Enzyme-Induced Calcium Carbonate Precipitation (EICP) with polyacrylic acid can entirely eliminate ammonia emissions[39]. However, these methods necessitate additional acid addition, thereby compromising sustainability to some extent and increasing costs.

This study explores a novel approach to biocementation by utilizing kitchen waste coffee grounds, leveraging the acidity of coffee ground extracts to regulate pH and control ammonia emissions, thereby eliminating the need for externally added acids. Coffee is one of the most widely consumed beverages in human society, annually generating substantial amounts of directly associated solid waste, known as coffee grounds. Improper management of these coffee grounds may lead to environmental pollution[40–42]. As illustrated in the flowchart in

Figure 1, this study investigates the potential advantages of using coffee grounds to assist in crude enzyme-induced biocementation. Notably, pH monitoring during the consolidation process indicates that the reaction system can be maintained below neutral pH, facilitating the conversion of ammonia into ammonium ions. This minimizes ammonia emissions, providing a sustainable solution to the challenges associated with large-scale applications.

2.Materials and methods

2.1 Preparation of coffee ground extract, crude enzyme, and consolidation solution

The coffee grounds waste used in this study, as shown in Figure 2(a), was sourced from the same batch of kitchen waste from a local café to minimize variations associated with different coffee grounds. Following preliminary experiments, the preparation steps for the crude coffee ground extract were determined as follows: the coffee grounds were dried, ground, and sieved through a 150-micron filter to ensure sufficiently small particle sizes, thereby optimizing extraction efficiency. Two grams of the powder were weighed and soaked in 10 mL of distilled water for 24 hours. After soaking, the mixture was centrifuged, and the supernatant was collected and defined as a 0.2 g/mL crude coffee ground extract. This crude extract was used in subsequent experiments and diluted as needed. Figure 2(b) shows the Scanning Electron Microscope (SEM) images of the coffee ground powder obtained using a Hitachi TM3000 microscope, and Figure 2(c) shows the Energy-dispersive X-ray spectroscopy (EDS) analysis of the coffee ground powder conducted using the JEOL JSM-IT200, indicating that its main components are organic substances, consistent with the findings of other researchers[43–45]. Figure 2(d) displays the appearance of the crude coffee ground extract diluted according to the gradient in Table 1, showing that the extract is brown, with the color intensifying as the dilution factor decreases. Figure 2(e) shows the pH of the crude coffee ground extract and its dilutions, measured with a Horiba LAQUA F-71 benchtop pH meter, indicating that the pH gradually decreases as the dilution factor decreases.

The soybeans used in the experiments were sourced from the same batch purchased from a local market to ensure consistency. The method for extracting the crude enzyme from soybeans was similar to that used for the crude coffee ground extract: the soybeans were dried, ground, and passed through a 150-micron sieve. 5 grams of the resulting powder were weighed and soaked in 100 mL of distilled water for 24 hours. After soaking, the mixture was centrifuged, and the supernatant was collected as the crude soybean enzyme at a concentration of 50 g/L, with an enzymatic activity of approximately 11.2 U at 25 °C.

The consolidation solution is prepared as described in Table 1, by mixing 5 mL of 1M calcium chloride and urea equimolar solution with an equal volume of the crude enzyme solution. Subsequently, 1 mL of distilled water is added as the control group, or 1 mL of crude coffee ground extract or its diluted solution is added as the test group for the consolidation solution.

Table 1. *Synergistic test and consolidation test formulations.*

Case name		formula
Synergistic test	Consolidation test	
SA	CA	1mL distilled water+ 5mL crude enzyme + 5 mL of 1M calcium chloride and urea equimolar solution
SB	CB	1mL coffee ground extract(0.2 times dilution) + 5mL crude enzyme + 5 mL of 1M calcium chloride and urea equimolar solution
SC	CC	1mL coffee ground extract(0.4 times dilution) + 5mL crude enzyme + 5 mL of 1M calcium chloride and urea equimolar solution
SD	CD	1mL coffee ground extract(0.6 times dilution) + 5mL crude enzyme + 5 mL of 1M calcium chloride and urea equimolar solution
SE	CE	1mL coffee ground extract(0.8 times dilution) + 5mL crude enzyme + 5 mL of 1M calcium chloride and urea equimolar solution
SF	CF	1mL coffee ground extract(No dilution) + 5mL crude enzyme + 5 mL of 1M calcium chloride and urea equimolar solution

2.2 Synergistic test

In order to comprehensively understand the impact of coffee ground crude extract on soybean crude enzyme-induced carbonate precipitation, a collaborative synergistic experiment was initially conducted to simulate the liquid-phase conditions during the coagulation testing.

Following the formulation depicted in Table 1, 1 milliliter of distilled water, coffee ground crude extract, or its diluted solution was added to a mixture comprising 5 milliliters of 1M calcium chloride and urea, and 5 milliliters of soybean crude enzyme. Specifically, distilled water was added to SA as a control group, while coffee ground crude extract and its diluted solution were added to experiments SB to SF, respectively. All utilized test tubes were sealed to prevent the escape of ammonia gas, thereby ensuring that the ammonia/ammonium concentration did not fall below the actual values.

The test tubes were then shaken at a rate of 160 times per minute for 24 hours at 25° C. Subsequently, the pH, ammonia/ammonium concentration, and calcium ion concentration in the solutions within the test tubes after 24 hours were measured. This was performed to comprehensively evaluate the effect of coffee ground crude extract on the activity of urease and the precipitation of biocement. The sediment at the bottom of the test tubes was dried in a 60° C oven for 48 hours and observed using SEM to analyze the morphology of the precipitate. Furthermore, crystalline phase analysis was conducted using the Rigaku MiniFlex benchtop X-ray diffractometer.

The determination of ammonia/ammonium concentration was performed using the indophenol spectrophotometric method based on the Berthelot reaction[46–48]. The absorbance values of the indophenol dye produced from the hydrolysis of urea to ammonia/ammonium were measured at 630 nanometers using a Jasco V-730 UV-Visible spectrophotometer. A calibration curve was established by correlating the absorbance values with different concentrations of ammonium chloride to ascertain the concentration of ammonia/ammonium. Calcium ions were measured using the Horiba LAQUAtwin Ca-11 compact Calcium ion meter.

2.3 Small-scale consolidation test

To minimize procedural complexity and facilitate future large-scale applications, a single-phase injection method was employed in this experiment due to its reduced number of injections and simpler steps[49]. Mikawa No. 4 silica sand was chosen for this study. It is a standard silica sand characterized by a loose structure, low cohesion, and poor stability. Its main component is silicon dioxide. The average grain diameter is 0.870 mm, the uniformity coefficient is 1.27, the curvature coefficient is 0.97, the particle density is 2.66 g/cm^3, the maximum dry density is 1.476 g/cm^3, the minimum dry density is 1.256 g/cm^3, and its relatively uniform properties help to maximize the reduction of potential complexities during the biocementation process.

Thirty grams of sand were placed into a cylindrical mold and lightly tapped to ensure a tightly and uniformly packed sample. Three replicates of the treatment were performed following the procedures outlined in Table 1. One pore volume of the consolidation solution was injected from the top to infiltrate the sand solely by gravity. After 24 hours, the effluent was collected and its calcium ion concentration, ammonia/ammonium concentration, and pH were measured. Upon completion of the treatment process, the samples were thoroughly rinsed with five pore volumes of distilled water to remove any residual unreacted chemicals. The samples were then placed in a 60 °C oven for 72 hours to ensure complete drying. The molds were carefully cut and the samples were extracted, as illustrated in Figure 5(a).

The Calcium Carbonate Content (CCC) is obtained by weighing a certain amount of sample, acid washing and drying, and the weight difference is the amount of calcium carbonate. Unconfined compressive strength (UCS) was measured to evaluate the effectiveness of the biocement consolidation treatment. The UCS of the samples was measured using an automatic Instron 2511-308 load cell with an axial strain rate of 0.036 mm/min until failure conditions were reached. After measuring the UCS of the treated samples, the morphology of the samples was examined at the microscopic scale using SEM. Subsequently, the samples were ground into powder and subjected to the X-ray diffraction (XRD) analysis to identify the crystalline phases present in the consolidated samples.

3. Results and Discussion:

3.1 Synergistic test analysis

Figure 3(a) shows the appearance of the synergistic experimental samples after 24 h. It is noteworthy that when crude coffee extract was added, the liquid and precipitate exhibited a green color instead of brown. This green hue deepened with increasing coffee extract concentration. This interesting phenomenon is consistent with the "green" theme mentioned in the title. A plausible explanation for this observation is that coffee contains abundant organic matter, such as chlorogenic acids, which undergo Maillard reactions and caramelization during roasting, giving coffee its complex flavor and brown color. The crude enzyme interacts with oxidized chlorogenic acids (CGA) to form green complexes. In addition, the intermediates of enzymatic urea hydrolysis and changes in pH may modify certain organic acids in the coffee grounds to produce green precipitates[50–52]. The specific mechanism behind this process deserves further investigation.

During the experiment, the pH of the discharged waste liquid was monitored, as shown in Figure 3b. Figure 3c depicts the ammonia/ammonium concentration, reflecting the extent of urea hydrolysis by the crude enzyme in the liquid phase. Compared to the control group SA without crude coffee extract, the pH values in all experimental groups decreased, with the extent of reduction increasing with higher concentrations of the coffee extract. This decrease is attributed to the abundant organic acids in the crude coffee extract, which lower the system's pH as their concentration increases, deviating from the optimal pH range for enzyme activity. Additionally, various complex organic substances might inhibit the activity of the soybean enzyme to some extent, reducing the ammonia/ammonium produced from urea hydrolysis, thus leading to a lower pH compared to the control group.

Figure 3d shows the calcium ion consumption, demonstrating that although the coffee extract influences EICP, the decreasing trend is not significant. While the crude coffee extract somewhat reduces substrate availability, the lowered pH favors the conversion of ammonia to its ionic form, thereby mitigating uncontrolled ammonia emissions. This finding lays the theoretical groundwork for achieving green construction in future large-scale applications, reinforcing the “green” theme highlighted in the title.

Figure 4 illustrates the sedimentation analysis conducted via SEM and XRD. It is evident that with the addition of coffee ground extract, although XRD analysis reveals that most of it precipitated as calcite, the precipitate size of calcium carbonate appears to increase. In the presence of SA, a significant number of small calcium carbonate particles were observed, whereas SF primarily consisted of larger calcium carbonate particles. This phenomenon can be partially attributed to the thermodynamic and kinetic factors governing crystal growth, as informed by the Gibbs-Thomson equation. With increasing concentrations of coffee ground extracts, the lower initial pH may have partially inhibited enzymatic activity, resulting in reduced supersaturation levels. Such conditions tend to favor the formation of smaller crystal particles due to constrained crystal growth. Additionally, with the deceleration in precipitation rate, calcium carbonate is afforded more time to grow from fewer nucleation sites, resulting in the formation of larger particles. In contrast, in the absence of coffee ground extract, the biochemical reaction proceeds at a faster rate, leading to the formation of more nucleation sites. However, the growth time of each nucleation site is shorter, ultimately yielding a higher number of smaller particles. Given the significant impact of calcium carbonate size on sample treatment efficacy, further research is warranted to achieve control over precipitation size.

3.2 Consolidation effect analysis

Figure 5(a) presents photographs of the samples, all of which demonstrate consolidation and possess certain mechanical strength. Visually, as the concentration of coffee ground extract added increases, the color of the consolidated samples appears to darken. This could be attributed to the reduced reaction extent in the consolidation experiment, as the reaction substrate does not experience the same oscillation and full contact as in a pure liquid-phase environment, thus the color of the samples remains dominated by the coffee hue.

The pH diagram presented in Figure 5(b) reveals a gradual decrease in the final reaction pH at the drainage end with increasing amounts of added coffee grounds. Similar to the analysis of the synergistic mechanism, this observation can be attributed to two main factors. Firstly, residual coffee ground extract plays a role, and secondly, the initially low pH environment inhibits enzyme activity, resulting in a reduction of the reaction products, thus effectively lowering the final pH. However, the reduction in pH is more significant compared to the synergistic experiment, indicating that the sand-liquid mixed phase to some extent impedes the extent of the reaction.

The ammonia/ammonium concentration diagram shown in Figure 5(c), reflects the extent of urea hydrolysis in the system. Figures 5(d) and the calcium ion concentration diagram demonstrate the consumption of calcium ions, calcium ion consumption is often highly correlated with the calcium carbonate content in the final sample. Similar to the synergistic test in a pure liquid phase environment, both Figure 3 and Figure 4 exhibit a decreasing trend with increasing amounts of added coffee grounds. However, the decrease in these figures is greater than that observed in the synergistic test conducted in a pure liquid phase environment, possibly due to the reduced extent of the reaction in the consolidation experiment, where the reaction substrate does not experience the same oscillation and full contact as in a pure liquid-phase environment.

Figures 5(e) and 5(f) assess the key parameters of the consolidation treatment effect, namely CCC and UCS, while Figure 6 presents the microscopic and phase results. It can be observed that although the addition of coffee ground extract slightly reduces the CCC treatment effect, there is typically no linear relationship between CCC and UCS. When a small amount of coffee ground extract is added, the reduction in effectiveness is not significant and appears to be acceptable. SEM analysis also indicates that bridging occurs at the contact points of sand particles in all groups. However, in the consolidation experiment, due to the reduced extent of the reaction, the size of calcium carbonate particles is significantly smaller compared to the synergistic experiment. The XRD analysis also indicates that in all cases, the calcium carbonate phase is not the stable calcite phase, which warrants further exploration in the future.

3.3 Large-scale application analysis

In future engineering applications, multiple treatments are typically required to meet strength requirements. Some studies suggest that adding urease inhibitors or reducing enzyme activity through methods such as low temperatures and pH adjustment can mitigate clogging issues that often arise with repeated treatments, thereby enhancing treatment uniformity and overall effectiveness[53–55]. This study primarily explores the preliminary feasibility of using coffee grounds, and thus, only a single treatment was conducted, avoiding the common clogging issues associated with multiple treatments. However, the potential of coffee grounds in repeated treatments, especially regarding their effectiveness in enhancing multiple treatments and uniformity, requires further investigation and validation.

One of the critical considerations for large-scale applications is the potential ecological disturbances and impacts on the surrounding environment as shown in Figure 7. The ammonia gas generated by biocement presents a significant challenge in large-scale applications, as it inevitably affects the local ecosystem and poses risks to on-site construction personnel. In this biocement, the addition of crude coffee extract lowers the pH of the reaction system, converting ammonia into ammonium ions, which are less prone to volatilization. Moreover, the plant-derived enzymes used in EICP eliminate potential biosafety issues associated with microbial cultures[56–58]. These factors collectively contribute to minimizing ecological disturbances at the site.

Additionally, the interaction between biocement and vegetation growth should be a focal point of large-scale trials. The root systems of growing plants can synergize with biocement to improve slope stability[59]. It is important to note that although coffee grounds have been explored for use as sustainable fertilizers, this typically requires pre-treatment processes such as composting[60–62]. Untreated coffee grounds contain certain chemicals that, if not decomposed by microorganisms, could inhibit plant growth. Future environmental toxicology analysis is necessary for the waste liquid collected after solidification in this study.

The hardened crust formed on the treated soil surface reduces the rate of water evaporation, which benefits plant growth[63]. Additionally, unreacted residual urea and ammonium ions can serve as fertilizers for plants. However, the residual salts in the soil can increase soil osmotic pressure, potentially limiting plant growth[64]. Compared to laboratory-scale consolidation experiments, field experiment environments are more complex, and the impact of biocemented soil on plant growth requires further investigation.

From a cost perspective, this study utilizes waste coffee grounds rather than additional acids, aligning with the principles of a circular economy and reinforcing the theme of green biocement. Coffee is one of the most widely consumed beverages in human society, generating a significant amount of coffee grounds annually. If not properly managed, these waste materials can cause environmental harm. Thus, the application of coffee grounds has been widely promoted as part of bioeconomic strategies and net-zero emissions initiatives. Recent studies have demonstrated the effectiveness of coffee grounds in stabilizing soil structure, enhancing soil nutrients and fertility, lowering the soil carbon-to-nitrogen ratio, improving soil water retention capacity, reducing bulk density, and controlling weeds[65–67]. This research offers a novel approach to utilizing coffee grounds, providing a sustainable solution for their disposal. However, relying solely on household coffee grounds is insufficient to meet the demands of large-scale engineering projects. Therefore, future efforts could focus on establishing more comprehensive collection systems, collaborating with coffee producers, coffee shop chains, and coffee processing companies to facilitate large-scale recovery of waste coffee grounds generated during production and consumption. Moreover, this study employs crude enzymes, offering a cost advantage[68–70]. Future efforts could explore the extraction of crude enzymes from agricultural waste[71], further enhancing cost efficiency.

4. Conclusion

This study explores a novel method for biocementation using EICP assisted by waste coffee grounds from kitchens. The experimental results indicate that coffee grounds extract can effectively maintain the pH of the reaction system below neutral, thereby facilitating the conversion of ammonia to ammonium ions and reducing ammonia emissions, without significantly diminishing the treatment efficacy. This finding is of significant importance for the large-scale application of biocement, as it can reduce environmental pollution and ecological impact at construction sites. Future research should focus on the uniformity of treatment in large-scale applications, ecological impact assessment, and the synergistic effects with vegetation growth to further optimize and promote this green technology.

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6. Funding declaration

This work was supported by JST SPRING, Grant Number JPMJSP2119, and was partly supported by JSPS KAKENHI Grant Number JP22H01581.

7. Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

8. Data availability

All the experimental data that support the findings of this study are available from the corresponding author upon reasonable request through email.

FIGURES

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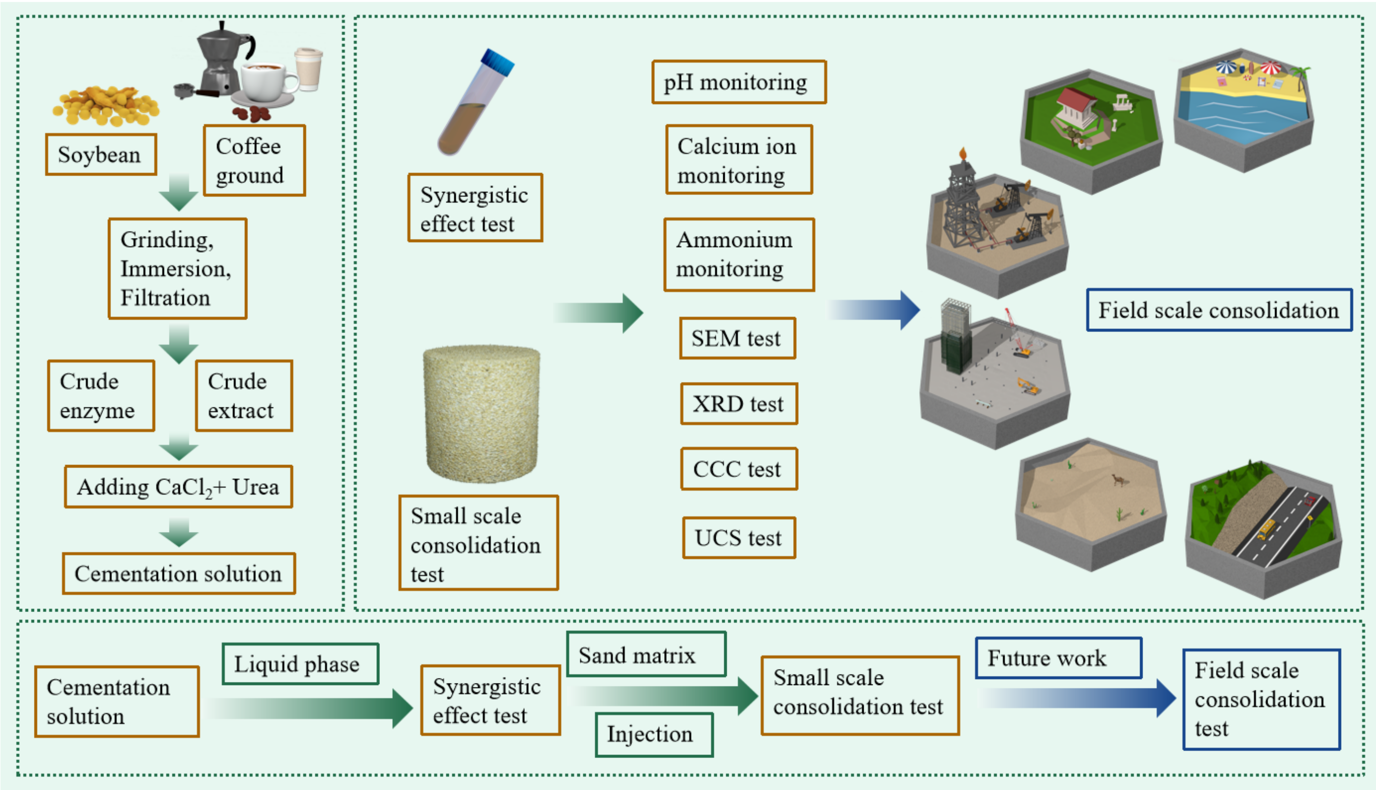

Figure 1. Flow chart of the experiment

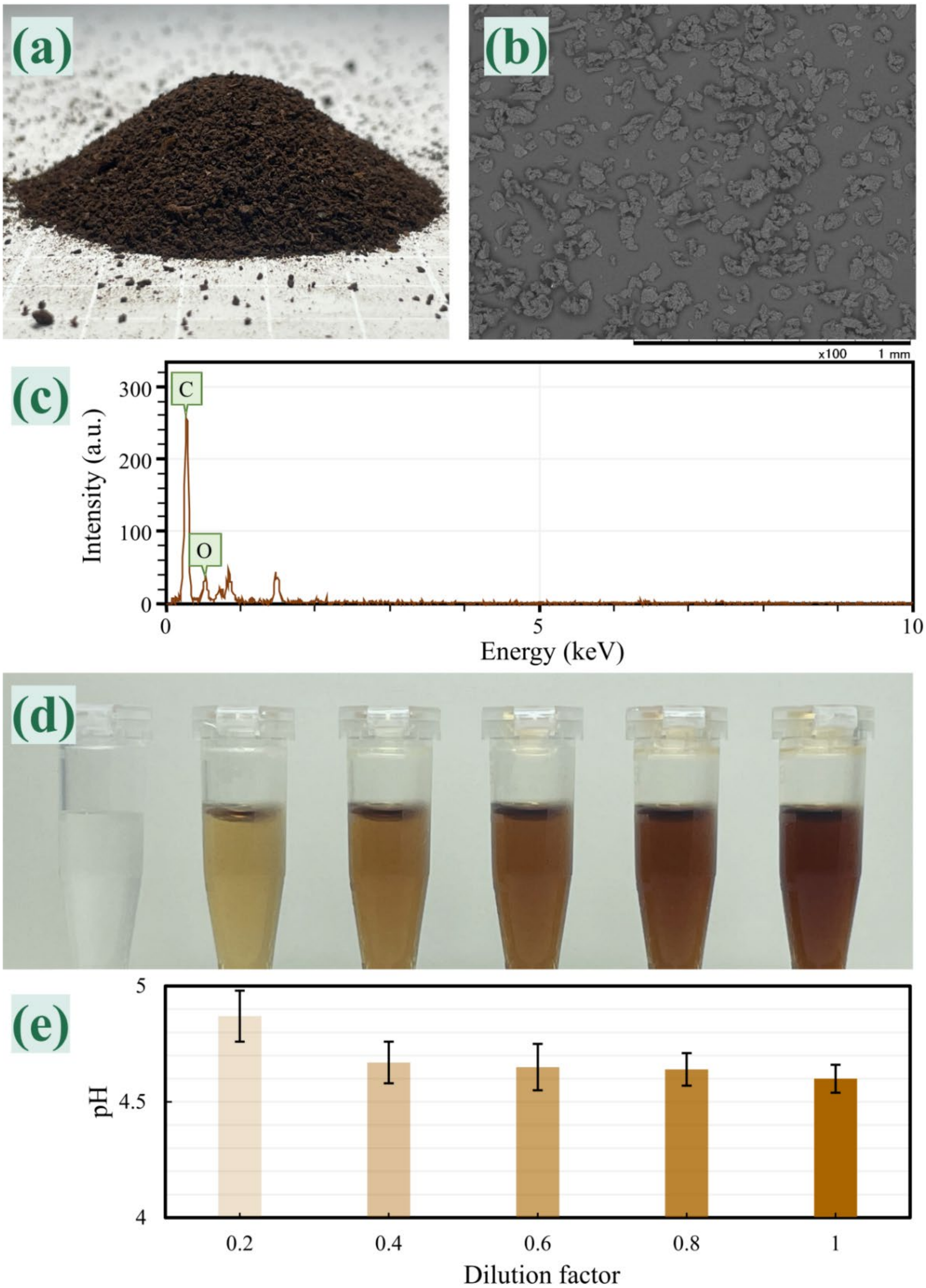

Figure 2. (a) is the appearance of the collected coffee grounds; (b) is the SEM image of the coffee grounds after grinding; (c) is the EDS image of the coffee grounds; (d) is the appearance of the coffee grounds extract and diluent; (e) is the pH of the coffee grounds extract and dilution.

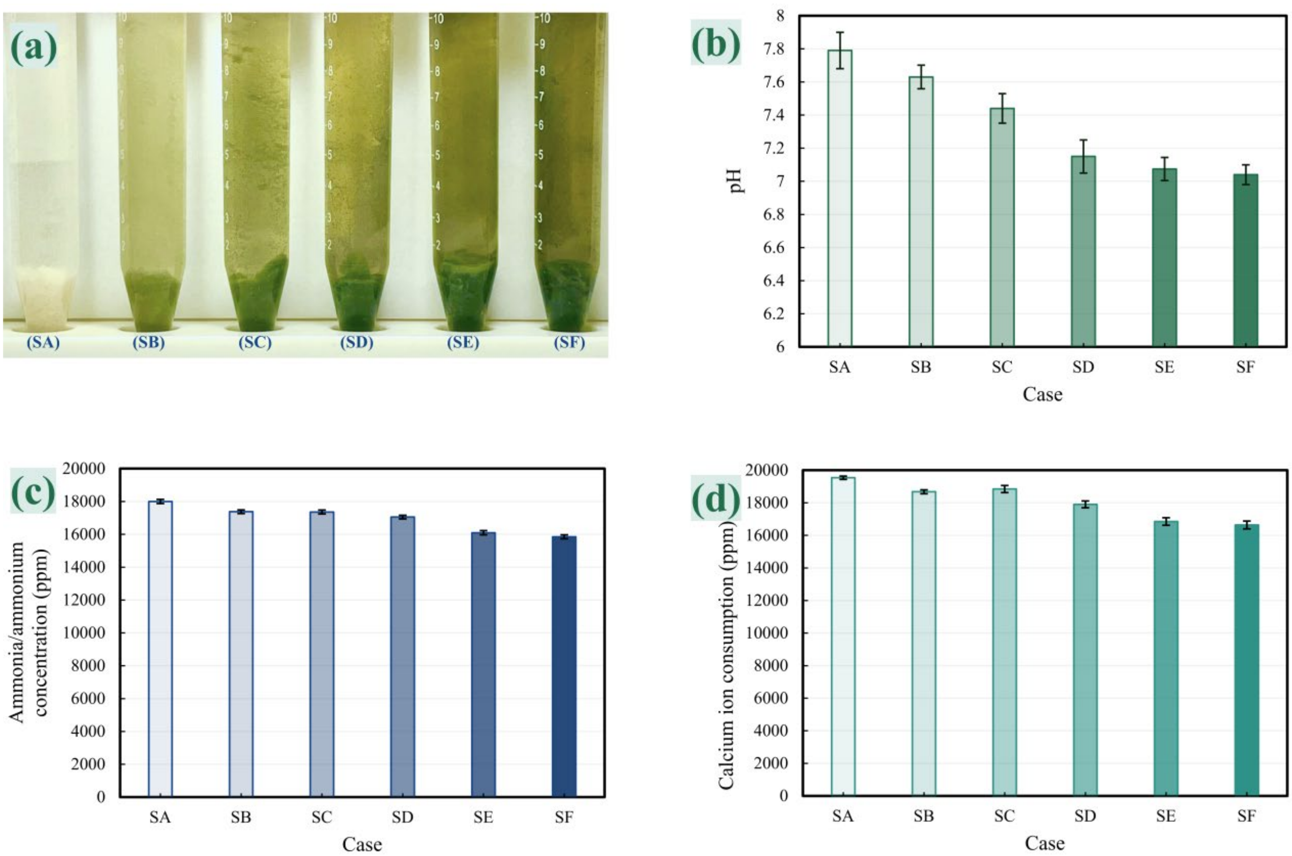

Figure 3. (a) is the appearance of the synergistic test; (b) is the pH value of the reaction system; (c) is the ammonia/ammonium concentration in the reaction system; and (d) is the calcium ion consumption.

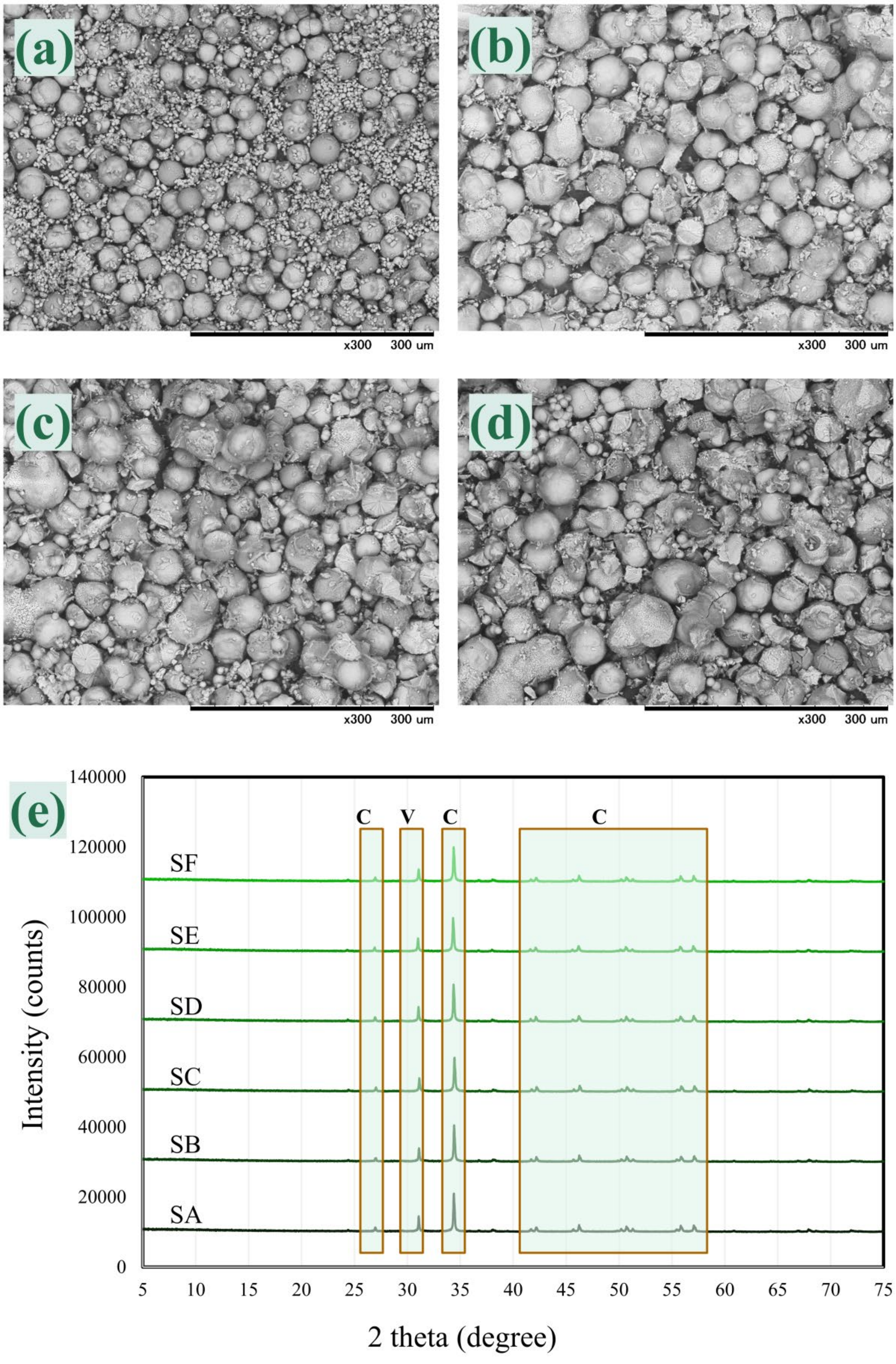

Figure 4. (a), (b), (c), (d) show the SEM images of SA, SB, SD, and SF precipitation respectively; (e) shows the XRD analysis of the synergistic test precipitation, C stands for calcite and V stands for vaterite.

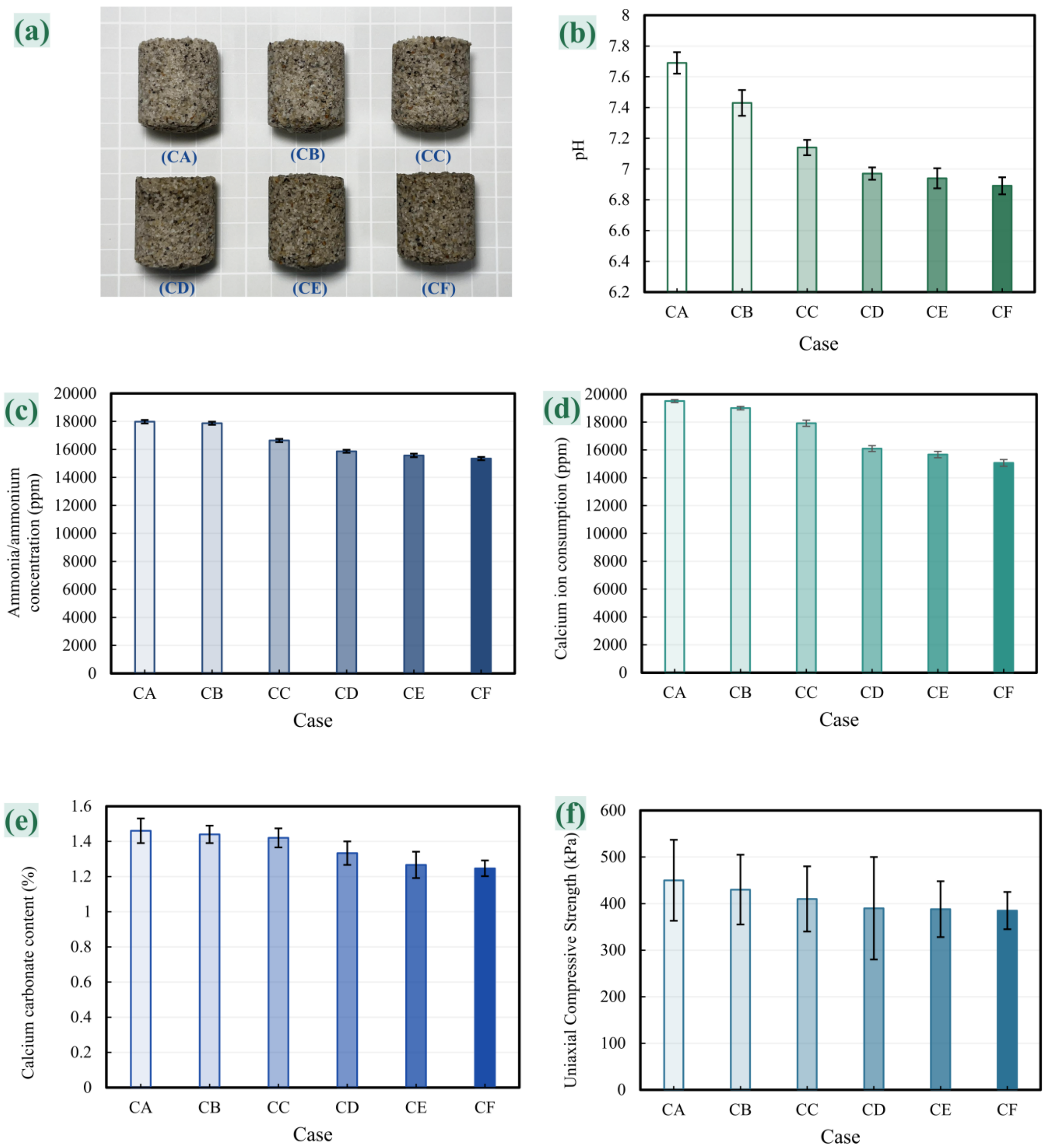

Figure 5. (a) is the appearance of the consolidated sample; (b) is the pH of the waste liquid after the consolidation test; (c) is the ammonia/ammonium concentration of the waste liquid after the consolidation test; (d) is the amount of calcium ions consumed in the consolidation test; (e) is the calcium carbonate content of the consolidated sample; (f) is the UCS strength of the consolidated sample.

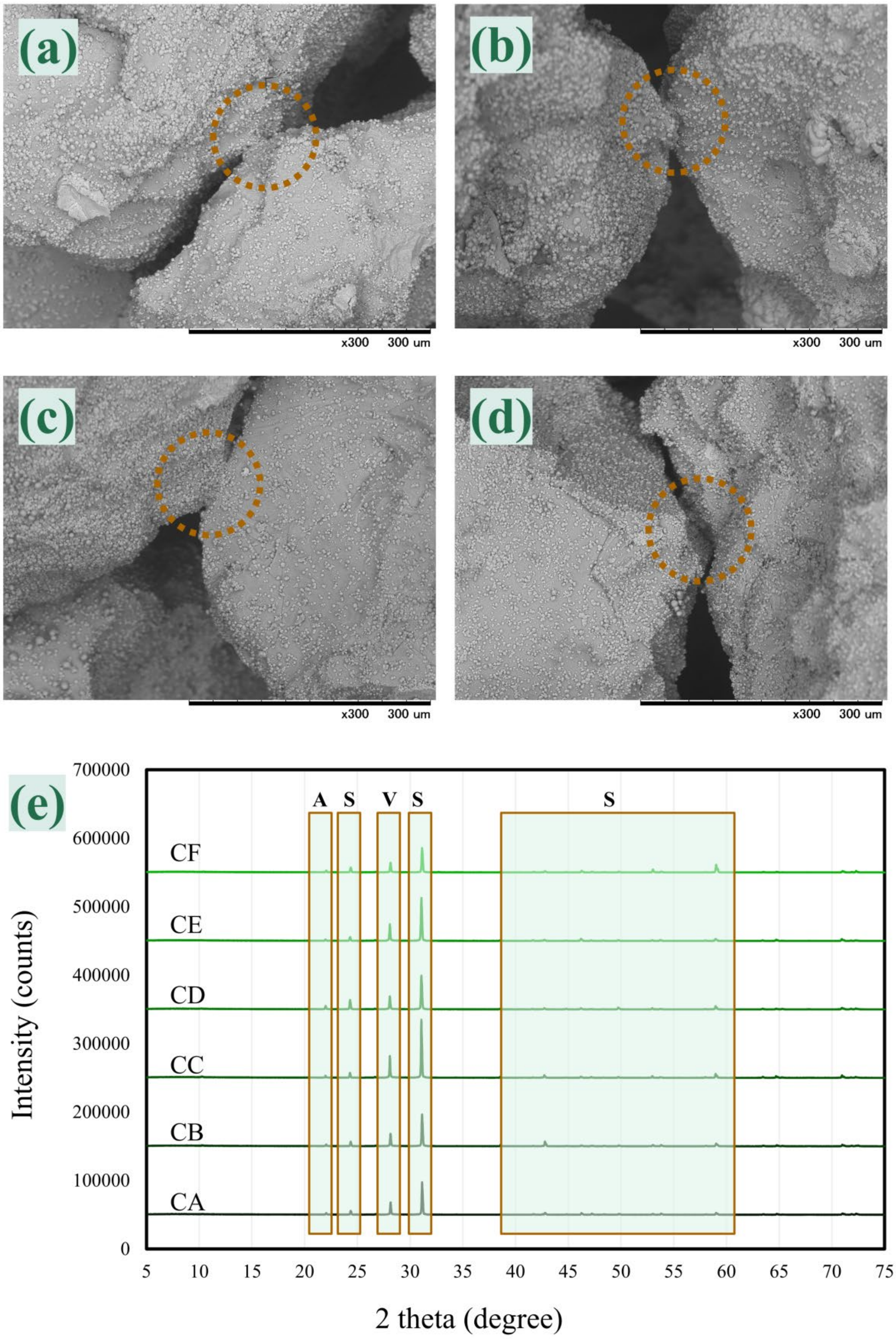

Figure 6. (a), (b), (c) and (d) are SEM images of CA, CB, CD and CF precipitates, respectively, the brown circles are bridges formed by calcium carbonate; (e) is the XRD analysis result of the consolidation test precipitate, S stands for silica, A stands for aragonite, and V stands for vaterite.

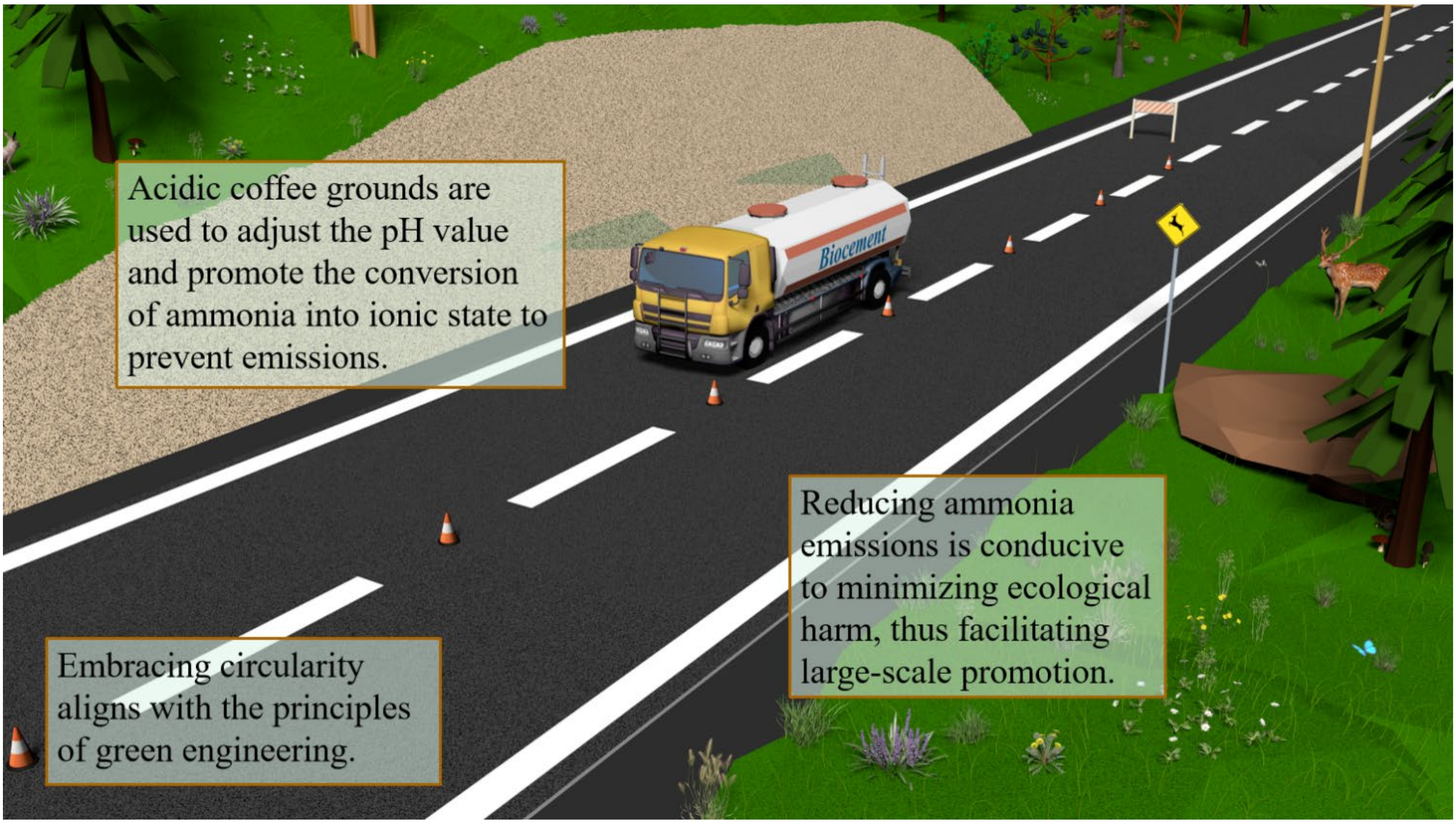

Figure 7. shows a potential application scenario.