

Article

Research on Activation of Solid Waste Through Microbial Desilification

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Abstract

To investigate the biosilicification capabilities of *Bacillus mucilaginosus* and *Bacillus polymyxa*, silicon concentrations in supernatants from quartz and calcium silicate cultures were monitored over a 12-day period using inductively coupled plasma optical emission spectrometry (ICP-OES). Fourier-transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), and scanning electron microscopy (SEM) were employed to evaluate changes in the absorption intensity of Si–O–Si characteristic peaks, crystalline phase transformations in the reaction products, and the microstructural morphology of quartz and calcium silicate before and after microbial leaching. The results show that after leaching with *B. mucilaginosus*, the dissolved silicon concentration in the quartz supernatant reached a maximum of 73.868 mg/L on day 8. In contrast, following treatment with *B. polymyxa*, the silicon concentration in the calcium silicate supernatant peaked earlier, at 149.153 mg/L on day 4. After microbial leaching, both substrates exhibited marked changes in the intensity of the infrared absorption peaks at 1071 cm^{-1} and 1083 cm^{-1} , suggesting the formation of Si–O–R type organosilicon complexes. Iron tailings (containing inert silica) and fly ash (containing active silica) were selected for experimental validation. Following treatment with *B. mucilaginosus* for desilication over an 8-day period, the activity index of iron tailings increased from 77.83% to 90.51%, while that of fly ash rose from 66.32% to 85.01%. ICP-OES analysis confirmed that under the action of *B. mucilaginosus*, the trends in silicon concentration and activity index in the supernatant of silica-containing solid wastes, such as iron tailings and fly ash, were consistent with those observed in quartz, thereby demonstrating effective biological desilication. These findings provide novel insights into the development of environmentally sound disposal methods for a wider range of solid waste types.

Keywords: microorganisms; quartz; calcium silicate; solid waste; desilication; activation



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1. Introduction

The rapid development of the social economy has been accompanied by a rising consumption of mineral resources and the accumulation of solid waste. Iron ore tailings represent one of China's largest categories of solid waste and continue to accumulate at a significant rate. Consequently, the comprehensive utilization of iron ore tailings has become an important research focus in China [1]. It is therefore essential to explore innovative approaches for improving the resource efficiency of iron ore tailings [2].

In agricultural production, silicate bacteria were initially employed to decompose silicate minerals such as feldspar and mica, transforming insoluble elements like silicon

and phosphorus into soluble forms that could be absorbed and utilized by plants [3,4]. In recent years, bioleaching has gained increasing attention as a promising approach for tailings treatment [5–7]. Compared to conventional physical and chemical activation methods, bioleaching offers advantages in terms of cost-effectiveness and environmental friendliness [8–10]. Studies have demonstrated that certain silicate bacteria exhibit efficient silicon-leaching capabilities. For example, *P. mucilaginosus* and *B. circulans* are known for their strong abilities in nitrogen fixation, phosphorus solubilization, and potassium release, making them effective in leaching silicate minerals [11,12]. Consequently, research on the application of silicate bacteria for industrial waste recycling and mineral desilication has begun to attract scholarly interest [13].

In recent years, *B. mucilaginosus* and *B. circulans* have shown significant potential in decomposing lithium-bearing silicate minerals. Moreover, these bacteria have been successfully applied in the bioleaching of electrolytic manganese slag, vanadium-bearing shale, and steel slag [14,15]. Unlike chemical leaching, the bioleaching process mediated by silicate bacteria involves both direct microbe–mineral interactions and biochemical reactions. Previous studies indicate that *P. mucilaginosus* secretes substantial amounts of polysaccharides, which can complex with minerals such as lithiopyroxene and lepidolite. However, excessive polysaccharide production may lead to bacterial aggregation, reducing the contact area between bacterial cells and mineral particles and consequently lowering leaching efficiency [16]. In addition, enzymes such as α -amylase, cellulase, and β -amylase produced by *Bacillus* species are capable of catalyzing polysaccharide hydrolysis [17,18]. To investigate the desilication effect and underlying mechanisms of silicate bacteria on bauxite, as well as bacterial adsorption on mineral surfaces, Sun Desi et al. isolated and screened silicate bacteria directly from bauxite samples [19–22].

To achieve silica activation, Li Jia et al. introduced mixed cultures of *B. mucilaginosus* and *B. circulans* into electrolytic manganese slag. Their results indicated that the direct microbe–mineral contact method was more effective for activating silica, with the effective silicon content in the leachate reaching 163.27 mg/L [23–25]. In a separate study, Ching Teng et al. conducted reverse flotation experiments on magnesite with and without pretreatment using silicate bacteria. The tests showed that bacterial pretreatment reduced the SiO_2 content in the concentrate from 4.61% to 2.56% [26]. Compared to monocultures, Yan et al. highlighted that mixed microbial cultures offer distinct advantages in mineral leaching, including greater metabolic diversity, an expanded pool of functional genes, and enhanced environmental adaptability [27]. They further noted that constructive interactions among microorganisms in mixed cultures—such as synergistic effects—can stimulate competition for survival and nutrients, leading to the secretion of stress-related metabolites and ultimately improving the efficiency of bioleaching systems [28].

The mechanisms governing interactions at the bacterium–mineral interface are not yet fully understood. A deeper comprehension of these interactions is critical for addressing numerous environmental and technological challenges. Therefore, elucidating the complex interfacial phenomena between bacterial metabolites and mineral surfaces remains an urgent research priority. This study aims to investigate the microbial activation of inert silica present in solid waste residues into active silica, and to examine its subsequent leaching behavior. Given that silicon primarily occurs in the forms of silica and silicates, the initial phase of this work focuses on the biological desilication of quartz and calcium silicate. Changes in dissolved silicon concentration in the supernatant were monitored, and a comparative analysis was conducted to evaluate the silicon leaching efficacy of *B. mucilaginosus* and *B. polymyxa* on these two substrates. Subsequently, *B. mucilaginosus* was applied to activate iron tailings to explore the underlying silicon leaching mechanisms.

Finally, the feasibility and practical reliability of this approach were assessed through macromechanical property tests in a cementitious system.

2. Materials and Methods

2.1. Experimental Raw Materials

Quartz (Zhongshan Xilong Scientific Chemicals Co., Ltd. Zhongshan, China) and calcium silicate (Zhongshan Xilong Scientific Chemicals Co., Ltd. Zhongshan, China) were purchased as analytical-grade mineral samples, both of which are essentially insoluble in water. P•O 42.5 grade cement (Shan Shui brand) (Fuzhu Building Materials, Weifang, China) was used in the experiments. Iron tailings and fly ash were finely ground to a particle size of 100 mesh. The mineralogical and elemental compositions are shown in Figure 1 and Table 1, respectively. Table 2 summarizes the physical performance parameters of the cement.

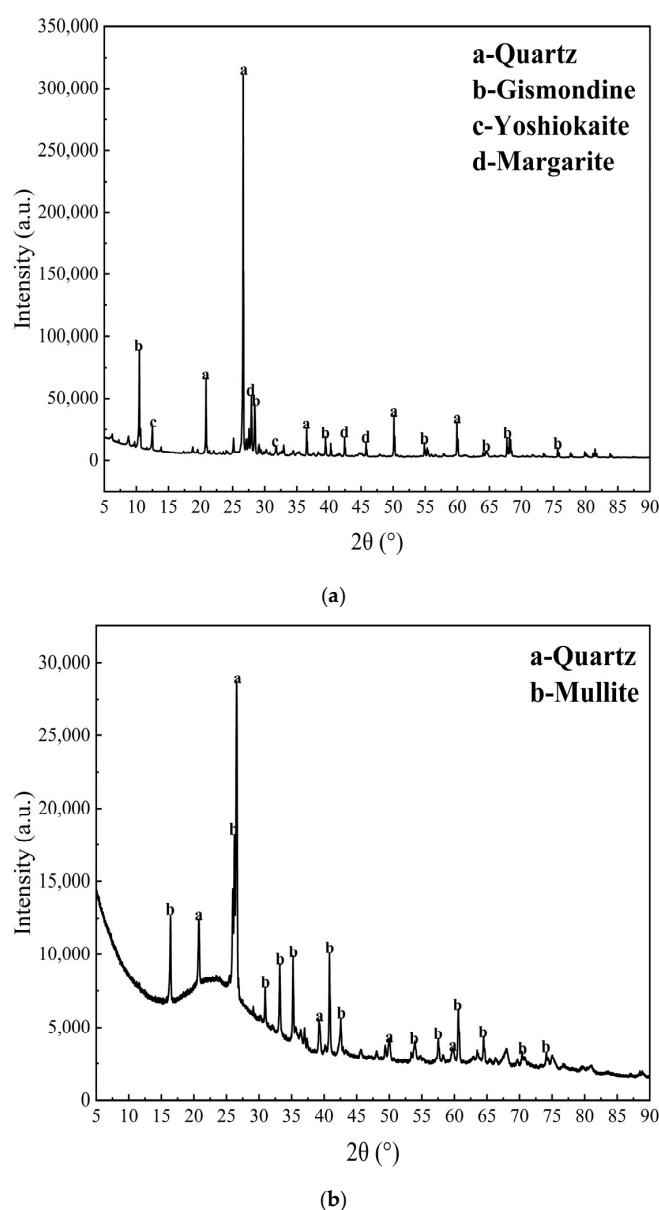


Figure 1. (a)-XRD pattern analysis of iron ore tailings, (b)-XRD pattern analysis of fly ash.

Table 1. Main chemical composition of iron ore tailings.

Chemical Composition	SiO ₂	Fe ₂ O ₃	Al ₂ O ₃	MgO	CaO	Else
Iron ore tailings	71.68%	9.60%	6.92%	4.40%	4.02%	3.38%
fly ash	50.49%	5.73%	32.21%	1.40%	3.23%	6.94%

Table 2. Cement physical property indexes.

Materials	Stability	Condensation Time/min		Flexural Strength/MPa		Compressive Strength/MPa	
		Condensation	Congeval	3 d	28 d	3 d	28 d
Cement	Eligible	95	210	6.3	8.2	18.9	49.2

2.2. Selection and Cultivation of Microorganisms

The Shanghai Microbiology Conservation Center provided the lyophilized powders of DSM 36 *B. polymyxa* and AS1.232 *B. mucilaginosus*, which were used.

The culture medium of *B. mucilaginosus* contained (g/L): MgSO₄•7H₂O 0.5; CaSO₄•H₂O 0.1; Na₂HPO₄ 2; FeCl₃ 0.005; glucose 5.0; glass powder 1.0.

B. polymyxa medium contains (g/L): beef paste 3.0; peptone 5.0; NaCl 5.0.

2.3. Microbial Desilication Experiment

To conduct the leaching experiment, 120 g of quartz and 120 g of calcium silicate were each placed into six 250 mL conical flasks. Then, 100 mL of culture medium containing a bacterial suspension (inoculum size: 10%) was added to each flask. An equivalent volume of distilled water was added to the blank control flasks. All flasks were incubated in a constant temperature shaking water bath at 30 °C with continuous agitation at 190 rpm. On days 4, 8, and 12, one flask containing quartz and one containing calcium silicate were removed from the shaker (two control flasks were allowed to settle for 4 days before supernatant collection). To terminate biological activity, each conical flask was immersed in boiling water (100 °C) for 30 min. Afterwards, 3 mL of the liquid was transferred with a pipette into a 50 mL centrifuge tube, diluted with 12 mL of distilled water, and centrifuged for 30 min. The supernatant was then collected for silica concentration analysis. The same experimental procedure was applied to the iron tailings samples.

2.4. Test Methods

Using an Agilent 5110 (OES) (Agilent Technologies, Stevens Creek Blvd, Santa Clara, CA, USA) inductively coupled plasma emission spectrometer, USA, the amount of silicon present in the supernatant was determined.

The infrared spectra of dried quartz and calcium silicate powders after bacterial leaching were obtained using a Thermo Fisher Scientific Nicolet iS20 FTIR (Thermo Fisher Scientific, Waltham, MA, USA) spectrometer (USA). This analysis aimed to compare the characteristic absorption bands in the infrared spectra before and after leaching. By examining changes in the intensity and wavenumber positions of characteristic peaks, the types of substances formed during the process were identified, thereby facilitating an analysis of the underlying mechanism of bacterial desilication.

Using a Rigaku Smart Lab 9 kw X-ray diffractometer (Rigaku Corporation, Tokyo, Japan), the mineral phase changes of calcium silicate and quartz prior to and following bacterial leaching were examined.

The JSM-7800F (Japan Electronics Co., Ltd., Tokyo, Japan) electron field emission scanning electron microscope was used to observe and analyze the surface morphology of calcium silicate and quartz both before and after bacterial leaching.

3. Results and Discussion

3.1. Analysis of Silicon Concentration and pH in the Supernatant from Quartz and Calcium Silicate Leached by *B. mucilaginosus* and *B. polymyxa*

Tables 3 and 4 Display the Silicon Concentrations of *B. mucilaginosus* and *B. polymyxa* Leached Quartz and Calcium Silicate Supernatants, Statistical Analysis of Silicon Concentrations in Quartz and Calcium Silicate Supernatants Is Presented in Figures 2 and 3.

Table 3. Silicon concentration of *B. mucilaginosus* leach supernatant (mg/L).

	Quartz	Quartz's pH	Calcium Silicate	Calcium Silicate's pH
Blank group	15.904	7.05	20.939	7.03
4 d	23.731	7.16	40.434	7.27
8 d	73.868	7.24	36.163	7.22
12 d	17.819	7.13	19.329	7.15

Table 4. Silicon concentration of *B. polymyxa* leaching supernatant (mg/L).

	Quartz	Quartz's pH	Calcium Silicate	Calcium Silicate's pH
Blank group	15.904	7.05	20.939	7.03
4 d	20.598	7.11	149.153	7.32
8 d	24.537	7.23	29.833	7.14
12 d	25.608	7.26	58.976	7.21

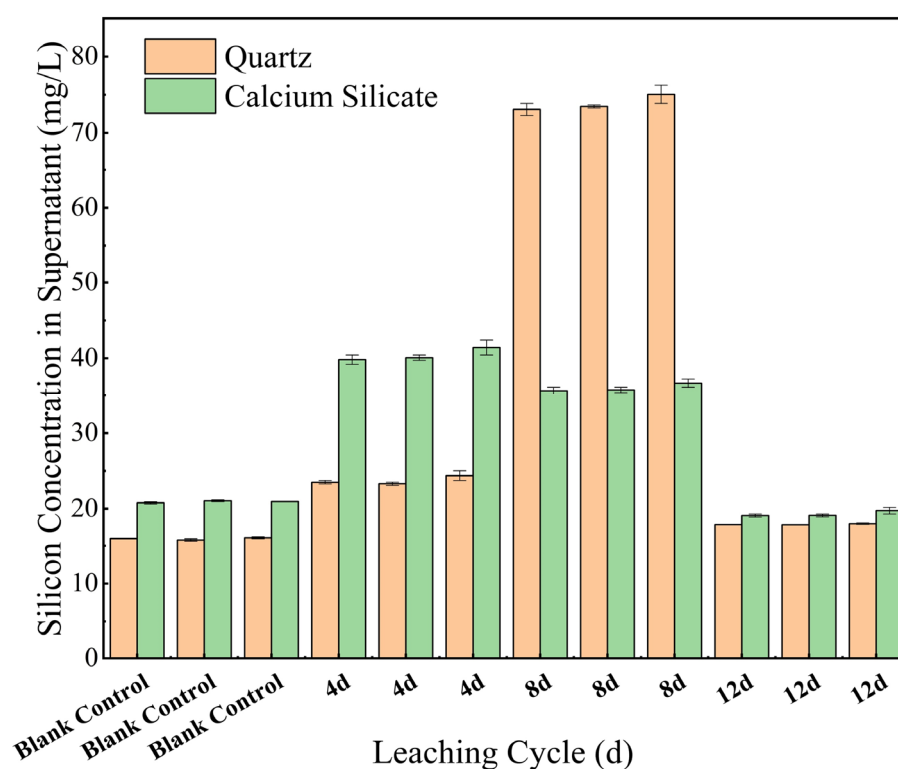


Figure 2. Statistical analysis of silica concentration in quartz supernatant.

The quartz and calcium silicate used in this study are both inert materials. Therefore, changes in silicon concentration within the bacterial leachate reflect the intensity of bacterium-mineral surface interactions, which is directly related to the number of bacteria

adhered to the mineral surface. While previous studies often employed column-based bioleaching setups [29], the present experiment utilizes a shaker leaching method to maximize microbial attachment to the mineral surface.

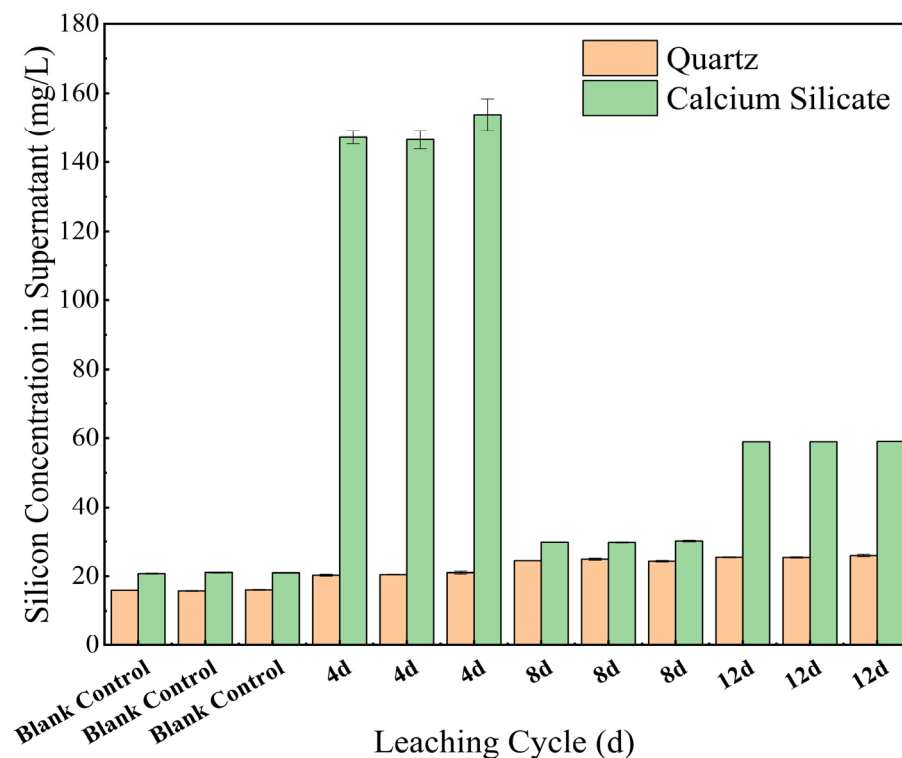


Figure 3. Statistical analysis of silica concentration in calcium silicate supernatant.

Tables 3 and 4 show the changes in silica concentration and pH in the supernatant after leaching quartz and calcium silicate with two *Bacillus* strains. Figures 2 and 3 provide the corresponding statistical analyses of silica concentration in the supernatants from quartz and calcium silicate, respectively. Fluctuations in silica concentration during leaching may be attributed to two main factors. First, silica ions released from the mineral surfaces may adsorb onto biomass or combine with other components in the ore to form secondary minerals, thus disturbing the dissolution equilibrium in the liquid phase and altering the measured silica concentration [30]. Second, organic acids generated during bacterial leaching can react with alkaline substances in the solution, changing the pH and consequently affecting leaching efficiency, which in turn leads to variations in dissolved silicon concentration.

As shown in Table 3, the silicon concentration in the quartz supernatant peaked at 73.868 mg/L after 8 days of leaching with *B. mucilaginosus*, showing an initial rise followed by a decline. In contrast, the calcium silicate supernatant reached its highest silicon concentration of 40.434 mg/L on day 4, after which the concentration decreased steadily. The polysaccharide metabolites produced by *B. mucilaginosus* are known to bind with silicon ions from quartz and calcium silicate, forming soluble silicon complexes in the leachate [31].

As the leaching process continues, nutrient depletion in the culture medium leads to bacterial apoptosis and a decline in cell numbers, thereby reducing mineral dissolution activity. Additionally, bacterial growth during bioleaching requires silicon as a nutrient for cellular development (as evidenced by experiments supplementing *B. mucilaginosus* medium with glass powder). Consequently, part of the silicon dissolved from quartz or

calcium silicate is assimilated by the bacteria, lowering the dissolved silicon concentration in the liquid phase.

Structurally, calcium silicate is less stable than quartz, and its crystalline defects are more vulnerable to bacterial attack. Bacteria readily colonize its surface, and their metabolic byproducts raise the pH of the leachate to alkaline levels, which in turn promotes silicon release into the supernatant. As leaching proceeds, the dissolved silicon concentration eventually approaches equilibrium. However, the dense bacterial colonization of calcium silicate surfaces hinders further silicon dissolution into solution.

In contrast, quartz undergoes gradual structural erosion under sustained bacterial activity, allowing substantial silicon to pass into the liquid phase. This explains why the supernatant from quartz leaching ultimately contains a much higher silicon concentration than that from calcium silicate.

The decline in dissolved silicon during later leaching stages for both minerals can be attributed to nutrient exhaustion in the medium. The resulting reduction in bacterial population and drop in leachate pH together lower the efficiency of silicon extraction, leading to decreased silicon concentrations in the supernatant.

B. polymyxa exhibited significantly higher leaching efficacy on calcium silicate than on quartz. Throughout all leaching stages, the dissolved silicon concentration in the calcium silicate supernatant consistently exceeded that in the quartz system. Studies indicate that the optimized culture medium for *B. polymyxa* contains calcium, suggesting that elevated calcium levels in the solution may stimulate bacterial growth and reproduction [32]. In the initial leaching phase, the inherent calcium in calcium silicate supported robust bacterial proliferation while simultaneously raising the pH of the leachate. Extensive bacterial adsorption onto the mineral surface contributed to a sharp increase in soluble silicon concentration. As leaching continued, bacterial colonization on the calcium silicate surface progressively increased. However, excessive bacterial accumulation, coupled with a subsequent decline in pH, reduced leaching efficiency and ultimately led to a decrease in dissolved silicon concentration in the liquid phase.

Based on the adsorption kinetics of bacterium–mineral interactions, the leaching efficiencies of *B. mucilaginosus* and *B. polymyxa* on quartz and calcium silicate are governed by the bacterial concentration in the liquid phase and the pH. However, an excessively high bacterial concentration reduces leaching efficiency—a phenomenon also noted during quartz leaching [33].

3.2. FTIR Analysis of Calcium Silicate and Quartz

3.2.1. FTIR Analysis of Quartz and Calcium Silicate After *B. mucilaginosus* Leaching

Infrared spectroscopy was performed on the dried powders of quartz and calcium silicate subsequent to their leaching by *B. mucilaginosus*. Analysis focused on the variation in absorption intensity of the characteristic Si–O–Si peak, the results of which are displayed in Figure 4.

Figure 4A–C compare the infrared absorption peak intensities of quartz and calcium silicate across the same wavenumber ranges. For a given leaching duration, the characteristic absorption peak intensities of quartz are consistently higher than those of calcium silicate.

The intensity of the characteristic Si–O–Si absorption peak is 1.06 for unleached quartz and 0.72 for unleached calcium silicate. Compared with their original infrared spectra, the characteristic Si–O–Si peaks in both leached samples show a shift in position, with the peak intensity of quartz remaining consistently higher than that of calcium silicate. The Si–O–R absorption band near 1090 cm^{-1} exhibits broadening, suggesting that bacterial extracellular polysaccharides react with dissolved silicon ions to form organosilicon complexes. The appearance of a doublet in the infrared spectrum indicates that the R group is likely a

C₂H₅ (ethyl) moiety [34], whereas a singlet corresponds to a CH₃ (methyl) group. Higher infrared absorption peak intensities reflect a greater abundance of the corresponding functional group, which aligns with the variations in silicon concentration observed in the supernatants of quartz and calcium silicate across different leaching stages.

A comparison of the infrared spectra of quartz and calcium silicate under identical leaching durations shows that quartz consistently exhibits higher intensities of the Si–O–Si characteristic absorption peaks than calcium silicate. Furthermore, the width of the Si–O–Si peak in quartz is narrower than that of calcium silicate only after the 8-day leaching period, a trend that corresponds well with the changes in silicon concentration observed in the supernatants of both materials. Integrating the silica concentration data with the infrared spectroscopic results indicates that *B. mucilaginosus* exerts a pronounced leaching effect on both quartz and calcium silicate, though its efficacy is greater on quartz. The infrared spectra of calcium silicate also display characteristic functional group signals, including a peak at 1560 cm^{−1} assigned to the C=O stretching vibration (Amide I) and a peak at 1617 cm^{−1} attributed to the bending vibration of the —C=O—NH— group (Amide II).

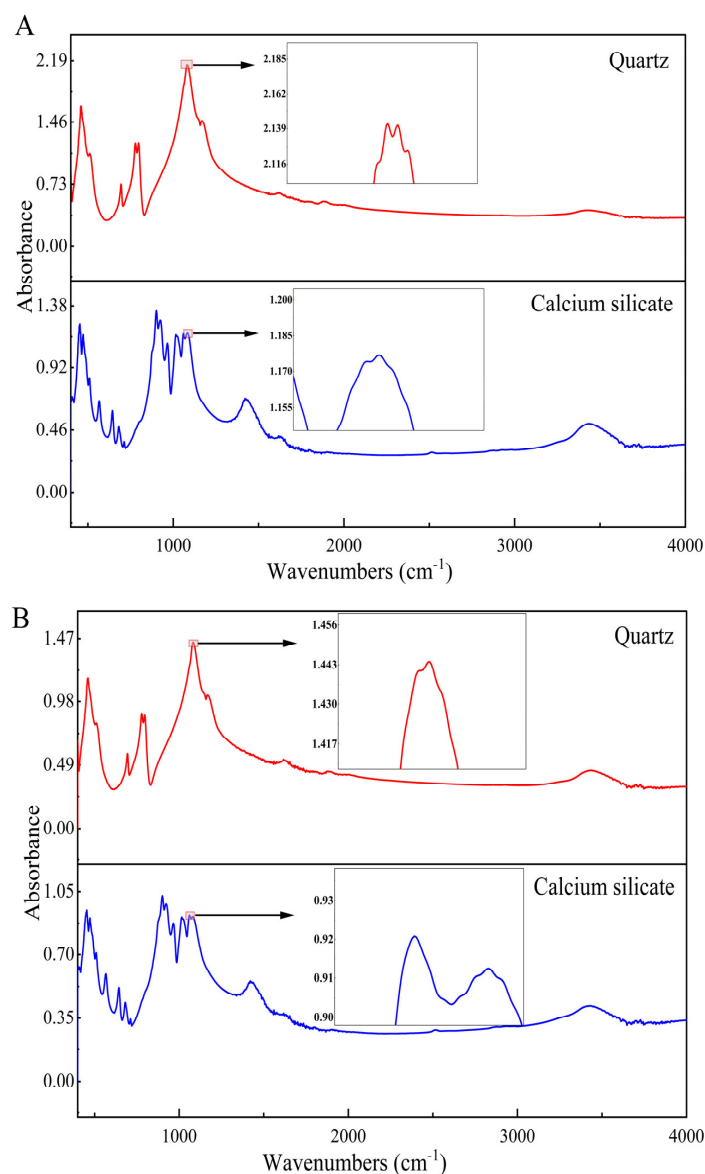


Figure 4. Cont.

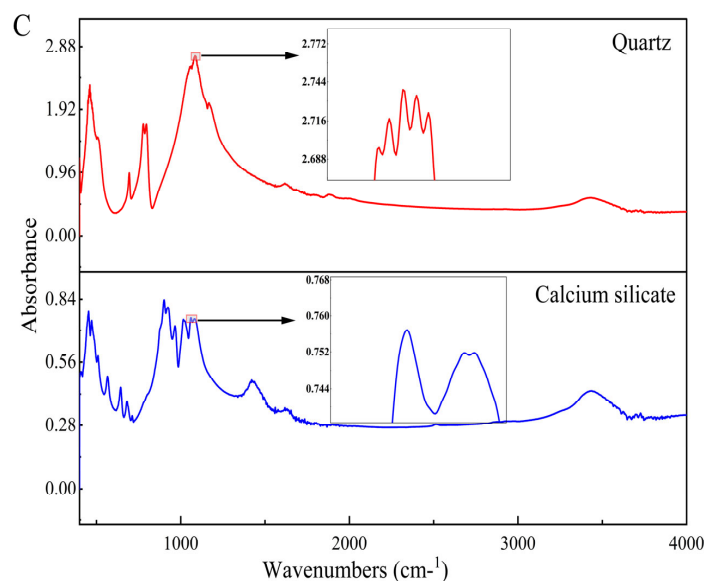


Figure 4. RTIR absorption spectra of quartz (red line) and calcium silicate (blue line) after *B. mucilaginosus* spores leaching. The inset shows the intensity of the characteristic Si-O-Si absorption peak. (A)—after 4 days; (B)—after 8 days; (C)—after 12 days of *B. mucilaginosus* leaching.

3.2.2. FTIR Analysis of Quartz and Calcium Silicate After *B. polymyxa* Leaching

Infrared spectroscopy was performed on dried powders of quartz and calcium silicate following leaching by *B. polymyxa*. Analysis focused on the absorption intensities of the characteristic Si-O-Si peaks, with results displayed in Figure 5.

Figure 5A–C present comparative infrared spectra of quartz and calcium silicate treated with *B. polymyxa* across different leaching durations. Throughout the leaching process, the absorption peak width of calcium silicate remained consistently broader than that of quartz. This variation in peak width corresponded to the changes in silicon concentration observed in the respective supernatants. New absorption peaks appeared at 1167 cm^{-1} and 514 cm^{-1} , which were tentatively assigned to fatty amine and fatty ketone functional groups, respectively, based on spectral database matching. The broadening of the characteristic infrared absorption peaks in both minerals is attributed to increased complexation of extracellular polysaccharides with silicon released from quartz and calcium silicate.

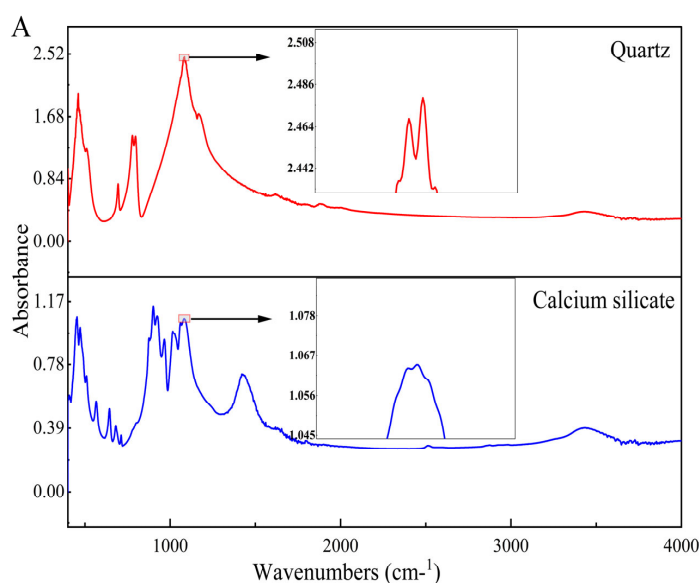


Figure 5. Cont.

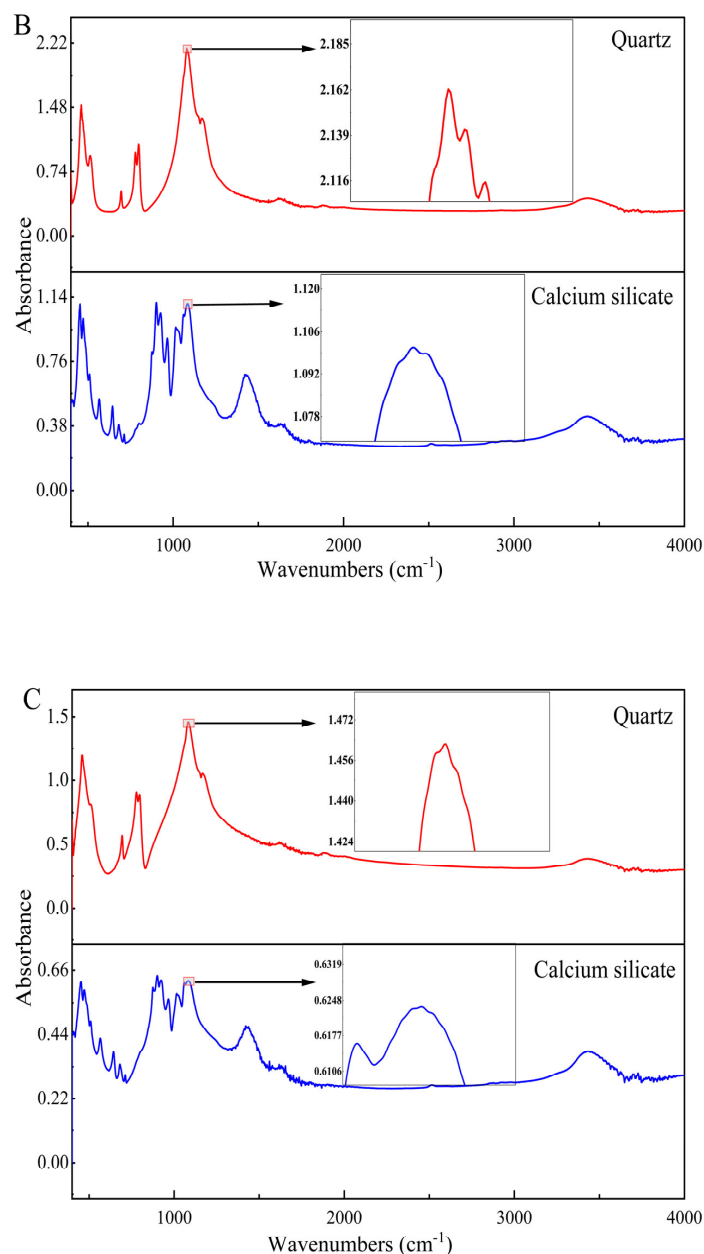


Figure 5. RTIR absorption spectra of quartz (red line) and calcium silicate (blue line) after leaching with *B. polymyxa*. The inset shows the intensity of the characteristic Si-O-Si absorption peak. (A–C) represent samples after 4, 8, and 12 days of leaching, respectively.

After 12 days of leaching, calcium silicate displays several new absorption peaks in the infrared spectrum: the peak at 1560 cm^{-1} is assigned to the C=O stretching vibration (Amide I); the peak at 1617 cm^{-1} originates from the bending vibration of the —C=O—NH— group (Amide II); the peaks at 1654 cm^{-1} and 1685 cm^{-1} correspond to amide-group and aromatic-acid absorptions, respectively. The peak at 1700 cm^{-1} is attributed to a saturated C=O stretch, while the peak at 1719 cm^{-1} is tentatively assigned to a saturated ketone. The peak at 1734 cm^{-1} may represent a saturated aliphatic aldehyde, and the peak at 1752 cm^{-1} is tentatively ascribed to a saturated ester. Absorption features above 3500 cm^{-1} likely arise from —NH groups [34].

3.3. XRD Analysis of Calcium Silicate and Quartz

Figure 6 presents infrared spectrum analysis before and after microbial treatment. Quartz and calcium silicate samples corresponding to the peak supernatant silicon concen-

tration were selected, dried, and subjected to crystal phase analysis. Figure 6A presents the XRD pattern of unleached quartz, while Figure 6B,C show the patterns of quartz after leaching with *B. mucilaginosus* and *B. polymyxa*, respectively. The intensity of the characteristic silica diffraction peaks was lower following leaching with *B. mucilaginosus* than with *B. polymyxa*. This can be attributed to the stronger leaching effect of *B. mucilaginosus* on quartz, which promotes the dissolution of organosilicon complexes into the liquid phase and thereby reduces the residual silica content in the solid.

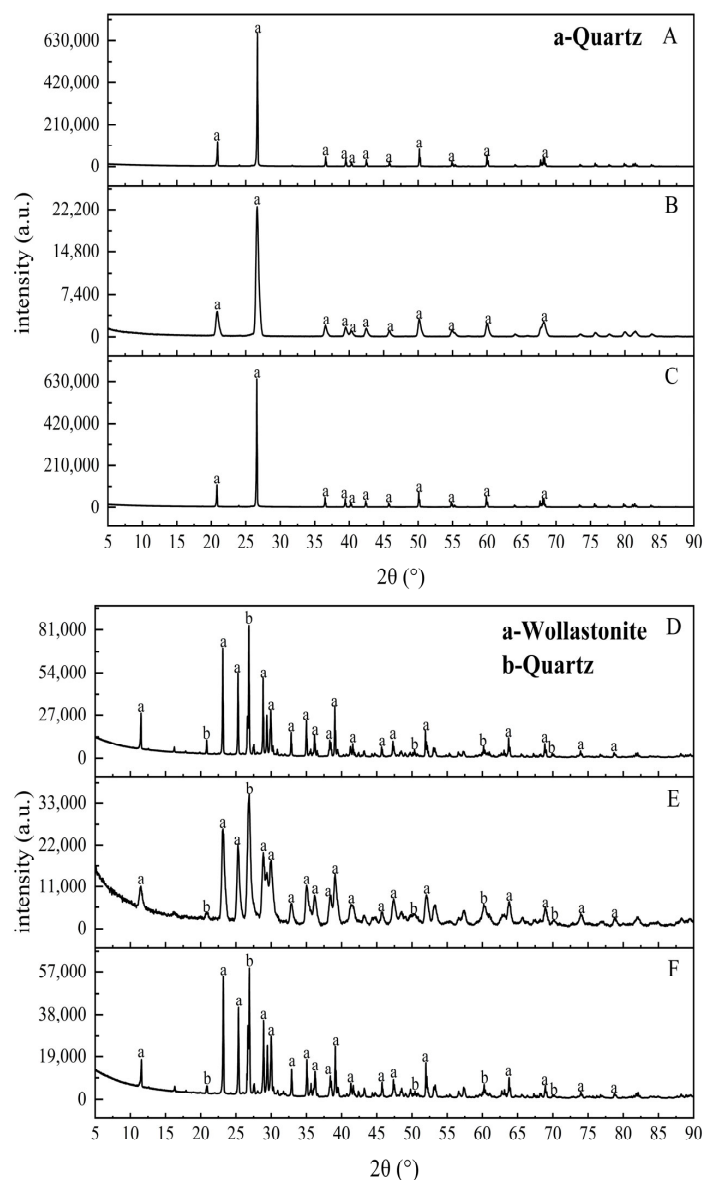


Figure 6. XRD patterns of quartz and calcium silicate before and after leaching with *B. mucilaginosus* and *B. polymyxa*. (A)—Quartz XRD pattern prior to leaching; (B)—Quartz XRD pattern after 8 days of *B. mucilaginosus* leaching; (C)—Quartz XRD pattern after 4 days of leaching by *B. polymyxa*, (D)—Unleached calcium silicate XRD pattern, (E)—Calcium silicate XRD pattern after 4 days of leaching by *B. polymyxa*, (F)—Calcium silicate XRD pattern after 12 days of leaching by *B. mucilaginosus*.

Figure 6D displays the XRD pattern of unleached calcium silicate, and Figure 6E,F present the patterns after leaching with *B. polymyxa* and *B. mucilaginosus*, respectively. Clearly, after treatment with *B. polymyxa*, the intensity of the characteristic silica diffraction peaks decreases markedly and is lower than that observed after leaching with

B. mucilaginosus. This further confirms that *B. polymyxa* exhibits a greater leaching efficacy on calcium silicate than on quartz.

3.4. SEM Analysis of Quartz and Calcium Silicate

3.4.1. SEM of Untreated Quartz and Calcium Silicate

Prior to microbial leaching, the surface morphologies of quartz and calcium silicate were examined, as shown in Figure 7a,b. A distinct microstructural contrast is evident: quartz possesses a granular form, in contrast to the rod-like morphology of calcium silicate. Additionally, fine particles are distributed across the surfaces of both materials.

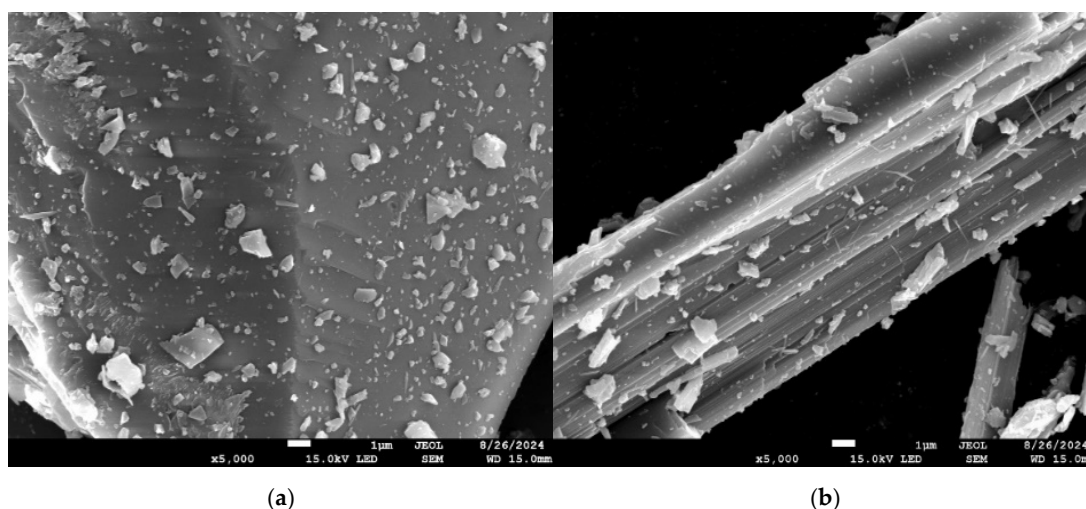


Figure 7. SEM images of unsoaked quartz and calcium silicate, (a)—Quartz SEM image, (b)—Calcium silicate SEM image.

3.4.2. SEM of Calcium Silicate and Quartz Following *B. mucilaginosus* Leaching

Figure 8a–f show the surface topography of quartz and calcium silicate after leaching with *B. mucilaginosus*. Compared to the unleached samples, both materials display pronounced surface damage and layered exfoliation, with erosion features becoming more severe as leaching time increases. This progression is attributed to the substantial amounts of organic acids and extracellular polysaccharides produced by *B. mucilaginosus*, which continuously corrode the mineral surfaces. Image analysis indicates that quartz particles primarily undergo layered delamination, whereas calcium silicate particles exhibit not only delamination but also collapse at crystal fractures and the formation of new, fine columnar crystals [31]. *B. mucilaginosus* thus has a pronounced destructive effect on both quartz and calcium silicate, with quartz experiencing more severe erosion. These observations are consistent with the corresponding trends in silica concentration and the changes indicated by infrared spectroscopy.

3.4.3. SEM of Calcium Silicate and Quartz Following *B. polymyxa* Leaching

Figure 9a–f present the surface topographies of quartz and calcium silicate after leaching with *B. polymyxa*. In comparison with the unleached samples (Figure 5), both materials show the formation of fine crystalline particles on their surfaces, the abundance of which increases with leaching time. A contrast with Figure 8 further reveals that the mode of action of *B. polymyxa* on these minerals differs distinctly from that of *B. mucilaginosus*. When SEM images of quartz and calcium silicate from the same leaching period are compared, the surface of calcium silicate consistently exhibits a higher density of adherent crystalline particles than that of quartz. Combined with the corresponding changes in

silicon concentration and infrared spectral data, these observations indicate that *B. polymyxa* exerts a stronger leaching effect on calcium silicate than on quartz.

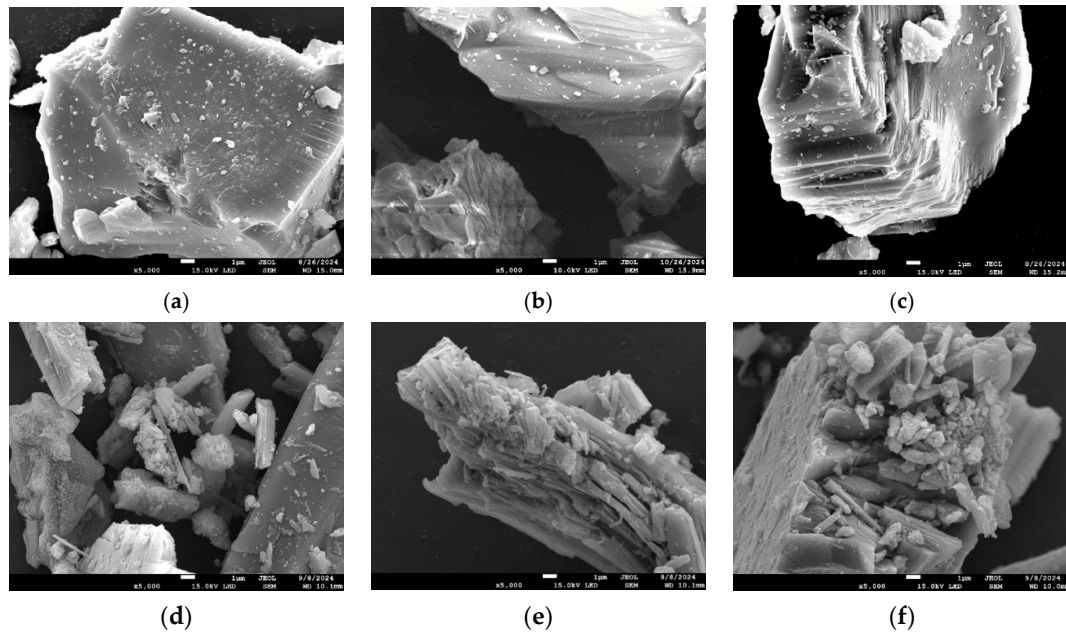


Figure 8. SEM images of quartz and calcium silicate after leaching with *B. mucilaginosus*, (a)—Quartz SEM image after 4 days of leaching, (b)—Calcium silicate SEM image after 4 days of leaching, (c)—Quartz SEM image after 8 days of leaching, (d)—Calcium silicate SEM image after 8 days of leaching, (e)—Quartz SEM image after 12 days of leaching, (f)—Calcium silicate SEM image after 12 days of leaching.

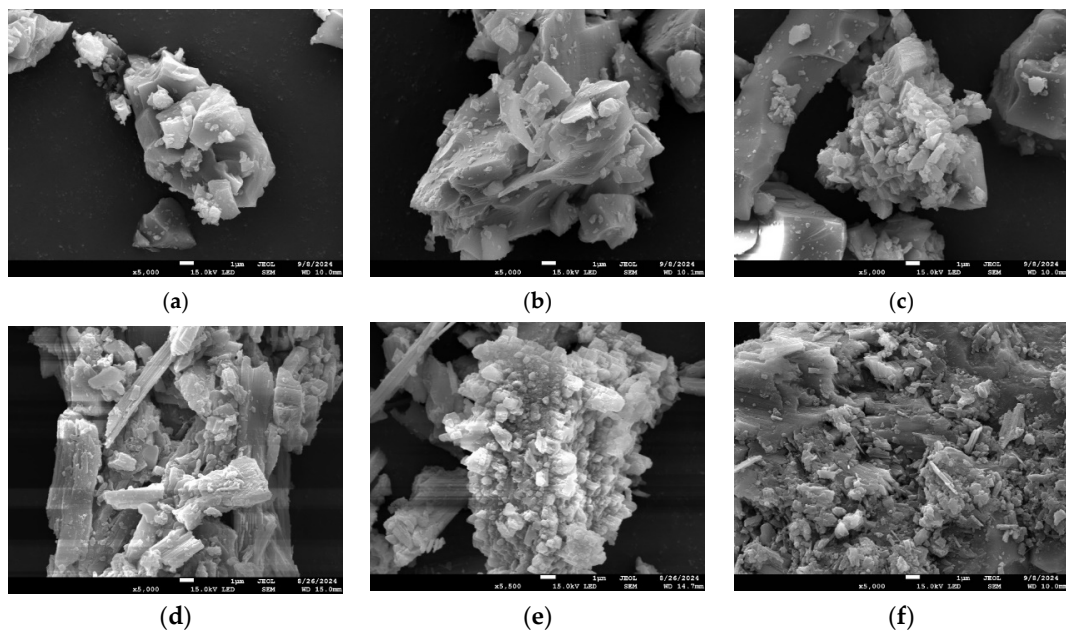


Figure 9. SEM images of quartz and calcium silicate after leaching with *B. polymyxa*: (a)—Quartz SEM image after 4 days of leaching (b)—Calcium silicate SEM image after 4 days of leaching (c)—Quartz SEM image after 8 days of leaching (d)—Calcium silicate SEM image after 8 days of leaching (e)—Quartz SEM image after 12 days of leaching (f)—Calcium silicate SEM image after 12 days of leaching.

3.5. The Cement-Solid Waste System's Macromechanical Characteristics

3.5.1. Testing of Silicon Concentration in Solid Waste Leached by *B. mucilaginosus*

Building on the demonstrated silicon-leaching efficacy of *B. mucilaginosus* in prior experiments, this section further examines its leaching performance on solid wastes rich in quartz and silicates, specifically iron tailings and fly ash. The dissolved silicon concentration in the supernatant during the later leaching stage was measured, and the results are summarized in Table 5.

Table 5. The amount of silicon (mg/L) in the supernatant of iron ore tailings that *B. mucilaginosus* leach.

Bacterial Species	Leaching Cycle	Iron Ore Tailings	pH of Iron Ore Tailings	Fly Ash	pH of Fly Ash
<i>B. mucilaginosus</i>	8 d	41.524	7.26	40.707	7.29
	12 d	16.586	7.17	20.345	7.21

Table 5 summarizes the variations in dissolved silicon concentration and pH in the supernatant during the leaching of iron tailings and fly ash. In both materials, the silicon concentration in the liquid phase decreased progressively with leaching time. Compared to pure quartz and calcium silicate, these solid wastes possess more complex elemental compositions. During leaching, elements such as aluminum, iron, and magnesium—alongside silicon—modulate the pH of the solution, which directly affects bacterial proliferation. While a moderate rise in pH can stimulate bacterial growth, excessively high pH levels inhibit microbial activity, thereby reducing silicon leaching efficiency and ultimately leading to a decline in dissolved silicon concentration.

A comparison with the leaching experiments on quartz and calcium silicate shows that the trends in silicon concentration for iron tailings and fly ash resemble those of quartz, both exhibiting a significant decrease in the later stages. This pattern is consistent with the late-phase effect of *B. mucilaginosus* on quartz and calcium silicate, where a moderate increase in pH can enhance silicon leaching efficiency to some extent. Between days 8 and 12 of leaching, the silicon concentration in the iron tailings supernatant declined more sharply than that in the fly ash supernatant. By day 12, the silicon level in the iron tailings leachate was also lower than that in fly ash. This difference can be attributed to the release of intracellular silicon from bacterial cells into the liquid phase during the later leaching period, which contributed to a pronounced reduction in dissolved silicon content, particularly in the iron tailings system.

The variations in silica concentration detected in the leachates of both solid wastes align with the results derived from the corresponding XRD analyses.

3.5.2. XRD Analysis of Iron Tailings and Fly Ash Before and After *B. mucilaginosus* Leaching

XRD analysis of iron tailings before and after bioleaching with *B. mucilaginosus* (Figure 10a, A–C) revealed a leaching-time-dependent increase in the diffraction intensity of quartz peaks. This intensification is interpreted as resulting from the interaction between residual silicon oxides in the solid and organosilicon compounds (identified by FTIR in the leachate) during the drying process, reflecting the transfer and re-association of silicon. The stability of zeolite and mica diffraction peaks throughout the leaching process highlights the targeted efficacy of *B. mucilaginosus* towards quartz in the complex iron tailings matrix.

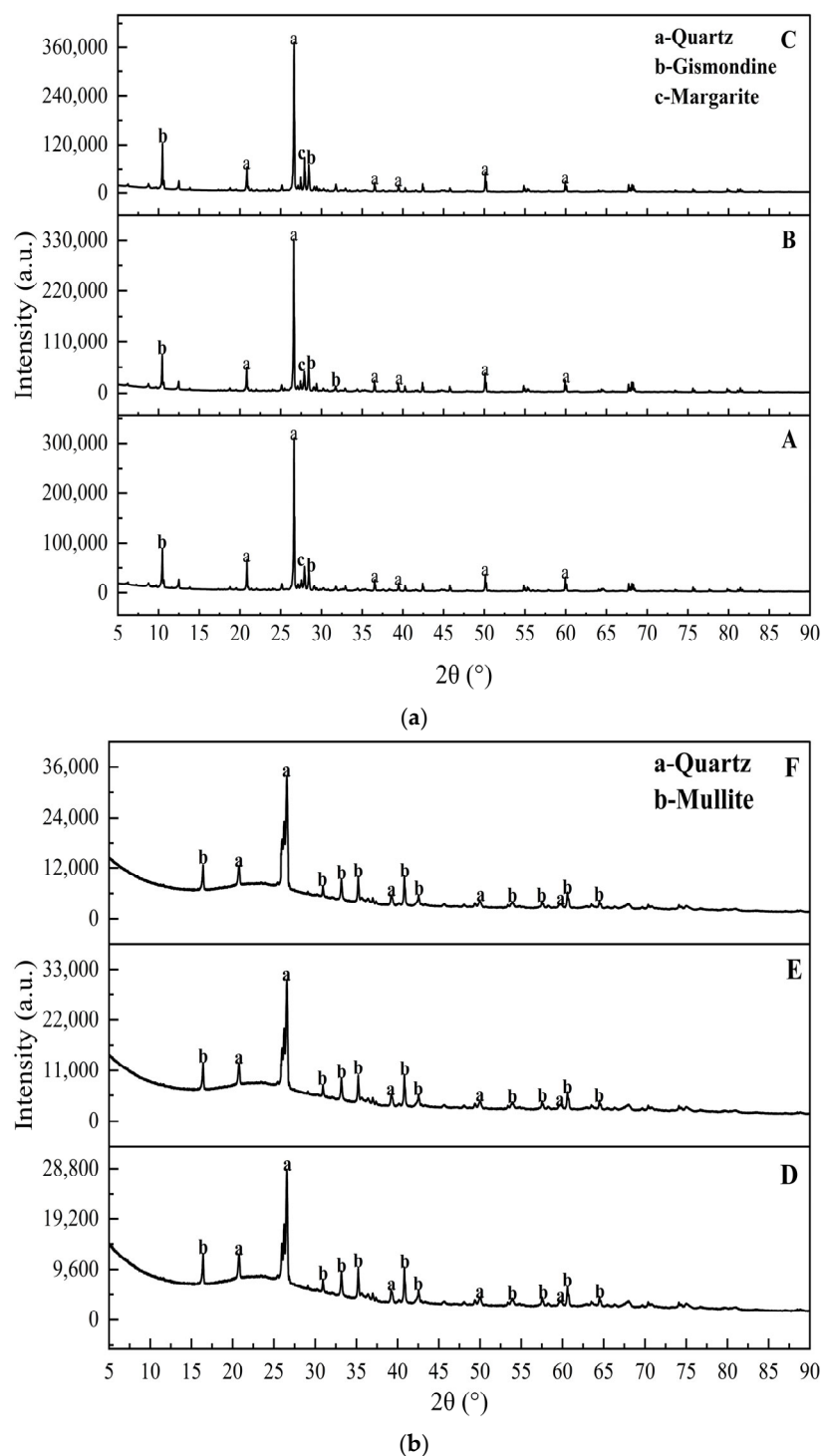


Figure 10. XRD patterns of solid waste before and after *B. mucilaginosus* spores leaching: (a)—XRD patterns of iron tailings before and after leaching, where A represents the XRD pattern of unleached iron tailings, B represents the XRD pattern of iron tailings after 8 days of leaching, and C represents the XRD pattern of iron tailings after 12 days of leaching; (b)—XRD patterns of fly ash before and after leaching, where D represents the XRD pattern of unleached fly ash, E represents the XRD pattern of fly ash after 8 days of leaching, and F represents the XRD pattern of fly ash after 12 days of leaching.

The XRD patterns of fly ash before and after leaching (Figure 10b, D–F) reveal a trend in diffraction peak intensity analogous to that observed in iron tailings. Following bioleaching with *B. mucilaginosus*, the diffraction intensity of the quartz phase increased progressively with time, which is ascribed to its interaction with organosilicon compounds

accumulating in the leachate. Conversely, the mullite phase showed no appreciable change in diffraction intensity. These observations confirm that *B. mucilaginosus* also effectively mobilizes silica from fly ash.

3.5.3. An Analysis of the Activity Index of Solid Waste Before and After Leaching with *B. mucilaginosus*

Iron tailings and fly ash powders leached with *B. mucilaginosus* were dried, ground, and used to replace 30% of the cement in specimen preparation. After hardening and demoulding, the specimens were cured under standard conditions. The 28-day compressive strength was measured, and the corresponding activity indices for each leaching cycle were calculated, as shown in Figure 11.

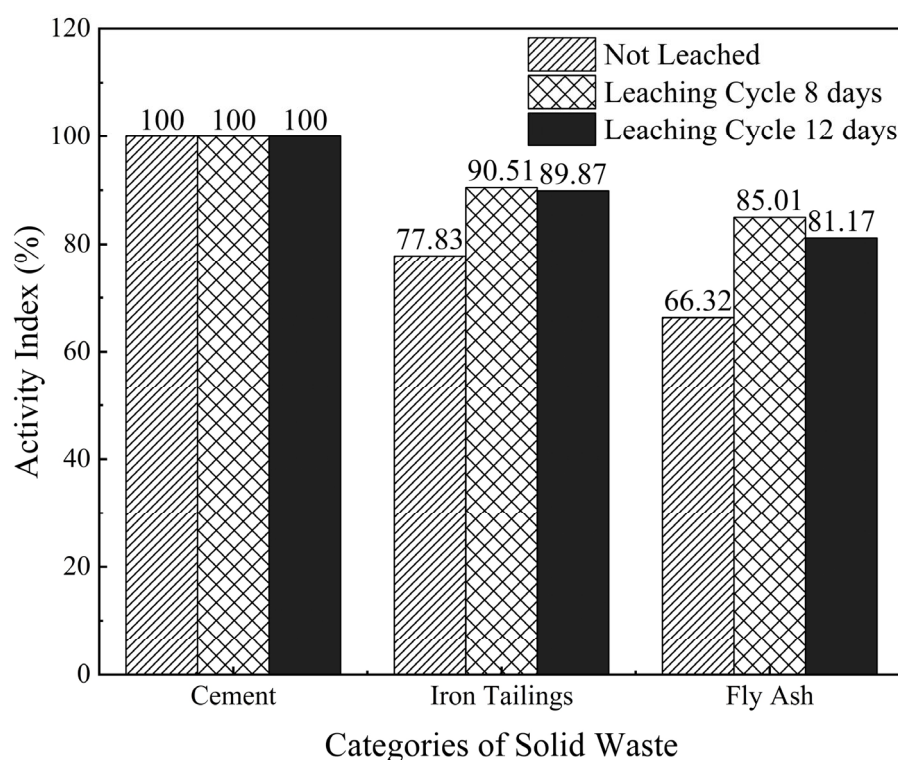


Figure 11. Activity Index of Solid Waste over a 28-Day Ageing Period The diagonal-lined boxes denote the activity index of unexposed solid waste specimens. The grid-lined boxes denote the activity index of specimens exposed for 8 days. The solid-lined boxes denote the activity index of specimens exposed for 12 days.

The activity indices of both iron tailings and fly ash increased after leaching compared to their unleached counterparts, a trend that correlates with the variation in silicon concentration observed in the leachate supernatant over time. The highest activity indices were achieved at the 8-day leaching point—90.51% for iron tailings and 85.01% for fly ash. As shown in Figure 11, the activity indices of the two leached materials initially increased and then decreased during the 28-day period. This pattern is attributed to the relatively high quartz content in both wastes. By integrating the trends in supernatant silicon concentration, XRD patterns, and the 28-day activity index, it can be concluded that the silica leaching behavior of iron tailings and fly ash resembles that of pure quartz.

4. Conclusions

This study elucidates the interaction mechanisms between two *Bacillus* strains—*B. mucilaginosus* and *B. polymyxa*—and two types of siliceous minerals: quartz and calcium silicate. Comparison of the silica leaching efficiency of these strains over different leaching

durations revealed that *B. mucilagenosus* exhibited higher leaching efficiency on quartz, while *B. polymyxa* demonstrated superior performance on calcium silicate.

Furthermore, the activation effect of *B. mucilagenosus* on iron tailings and fly ash was investigated. The experimental results indicate that the interaction between bacteria and minerals occurs via two primary pathways: (1) direct attachment of bacterial cells to silica-containing mineral surfaces, and (2) formation of organosilicon complexes between bacterial metabolites and silicon within the mineral matrix, facilitating dissolution into the leachate. Fourier-transform infrared (FTIR) spectroscopy analysis showed that extracellular polysaccharides, as bacterial metabolic products, bind to silicon in siliceous minerals through Si–O–R bonds, thereby achieving biological desilication.

Inductively coupled plasma (ICP) data confirmed that *B. mucilagenosus* reached its peak silicon leaching efficiency on quartz at 8 days, whereas *B. polymyxa* achieved its maximum efficiency on calcium silicate at 4 days. The leaching efficacy of *B. mucilagenosus* on iron tailings (containing inert silica) and fly ash (containing active silica) was comparable to that on quartz. After leaching, the activity indices of both solid wastes followed trends consistent with the silica concentration variations in their corresponding supernatants.

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