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Stabilization of Lead-Contaminated Mine Soil Using Natural Waste Materials

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Abstract: Stabilization was applied as a remediation option for immobilizing lead (Pb) in contaminated mine soil. Four types of waste materials were used as stabilizing agents, namely waste oyster shells (WOS), calcined oyster shells (COS), natural starfish (NSF), and spent-coffee-grounds-derived biochar (SCGB). The Pb-contaminated mine soil was treated with the stabilizing agents ranging from 0 to 10 wt% and a curing period of 28 days. The toxicity characteristic leaching procedure (TCLP) was employed for evaluating the effectiveness of the remedial process. The Pb immobilization mechanism in the treated mine soil was investigated using scanning electron microscopy–energy dispersive X-ray spectroscopy (SEM-EDX) analyses. The obtained results showed that the TCLP Pb levels decreased as the stabilizing agent dosage increased. The WOS and COS stabilizing agents immobilized Pb effectively in the contaminated mine soil. Application dosages of 4 wt% and 2 wt% for WOS and COS, respectively, were sufficient for compliance with the TCLP regulatory level of 5 mg/L. For the NSF and SCGB treatments, 4 wt% and 10 wt%, respectively, were necessary to meet the TCLP regulatory limit. The effectiveness of Pb immobilization was found to increase in the following order: SCGB < NSF < WOS < COS. The chemical fraction analyses showed that the lower F2 (weak acid soluble) fraction and higher F3 (reducible) and F4 (oxidizable) fractions were most likely associated with the high level of Pb immobilization. Moreover, the SEM-EDX analysis results showed that the most effective Pb immobilization could be strongly associated with the pozzolanic reaction products.

Keywords: stabilization; immobilization; lead; oyster shell; starfish; biochar

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1. Introduction

Lead (Pb) contamination is highly problematic in mine soil. In addition to high toxicity and carcinogenicity, Pb is recognized to trigger various adverse health effects to humans (i.e., mental, neurologic, hematologic, reproductive, developmental, and renal) [1,2]. There are about 2600 abandoned mines located in Korea, and approximately 50% of them are known to be linked to serious Pb-release problems [3]. The mine tailings generated from the ore dressing are usually stored in a mine tailing dam. However, loss of mine tailings often occurs as a result of heavy rainfalls and landslides. In turn, this may lead to contamination of agricultural lands located near mine sites. Therefore, remedial action is warranted to prevent Pb releases from mine soil.

Among various remedial techniques, stabilization/solidification (S/S) is regarded as a viable option for immobilizing Pb effectively in contaminated mine soil. In the past, various S/S agents (e.g., fly ash, quicklime, Portland cement, cement kiln dust, etc.) have been utilized for immobilizing heavy metals in contaminated soils [4–13]. Recently, to improve the sustainability of S/S remedial processes, the application of natural and/or consumer-use waste materials as stabilizing agents has received some attention [14–16]. However, the use of sustainable waste materials as stabilizing agents for mine soil is very limited, and comparative effectiveness studies have not been reported in the literature.

In this study, two natural waste materials (waste oyster shells, WOS; natural starfish, NSF), a modified natural waste material (calcined oyster shells, COS), and a modified consumer-use waste (spent coffee grounds derived biochar, SCGB) were used as stabilizing agents to treat Pb-contaminated mine soil. At the southern coast of the Republic of Korea, it is estimated that only 10% (25,000 tons) of the waste oyster shells generated annually are recycled as fertilizer [17]. Foul odors and related environmental degradation are issues associated with uncontrolled oyster shell dumping (approximately 40%) in coastal areas, often necessitating the implementation of remedial actions. Calcite, the dominant mineral phase of WOS, is converted into quicklime (CaO) via calcination at high temperatures (900 °C for 2 h). Quicklime is a well-known S/S agent for heavy metal immobilization, widely applied to contaminated soil and hazardous waste [8]. Calcined oyster shells have been used effectively for stabilizing Pb in the soil of firing ranges [18].

Starfish are an invasive species known to devastate natural marine ecosystems. Since the population of triton shellfish, a natural predator of starfish, is very limited, oyster fishermen often capture a great number of starfish to control outbreaks of this invasive species. However, the recycling of captured starfish is limited, and the beneficial reuse applications have not been widely explored. Natural and calcined starfish treatments have been applied effectively for Pb immobilization in contaminated rice paddy soil [16]. In addition, a 10 wt% treatment of natural starfish on Pb/Zn/As-contaminated agricultural soil caused a notable reduction (almost 100%) in the TCLP Pb concentration [19].

Biochar is a multifunctional material derived from a variety of natural and waste substrates (including spent coffee grounds) often used for carbon sequestration, metal remediation, and soil conditioning [20–22]. Effective use of soybean stover-derived biochar for Pb immobilization has been reported [18]. Moreover, the biochars derived from soybean stover and pine needles at 300 °C and 700 °C showed a significant reduction of Pb mobility by 95% from contaminated agricultural soil [23].

The objective of this study is to evaluate the feasibility of the beneficial reuse of natural and consumer-use waste materials for the immobilization of Pb in thecontaminated mine soil. The Pb immobilization effectiveness in mine soil is assessed by the TCLP test. The TCLP regulatory limit for Pb is set at 5 mg/L. The investigation of the Pb immobilization mechanism in the contaminated mine soil is based on scanning electron microscopy-energy dispersive X-ray spectroscopy (SEM-EDX) analyses. X-ray powder diffraction (XRPD) is used for evaluating the mineral phases present in the mine soil and stabilizing agents. Application of the selected waste materials (WOS, COS, NSF, and SCGB) for stabilizing Pb-contaminated mine soil is rather limited. Therefore, the results obtained in this study could be used for the selection of optimal stabilizing agents for mine soil remedial action.

2. Materials and Methods

2.1. Contaminated Mine Soil Collection

A sample of about 10 kg of Pb-contaminated mine soil from an active mine site in Bonghwa County, Gyeongsangbuk-do, Korea was collected using a shovel, following procedures described in the Standard Methods of Soil Sampling and Analysis [24]. This mine has been active and operating for the extraction of gold, lead, and zinc since the early 1900s. Contaminated mine soil was sampled from near the mine tailing dam. The collected mine soil was transported to the laboratory and spread uniformly on a vinyl sheet. Subsequently, the samples were air-dried for one week and sieved through a U.S. standard #10 mesh sieve (2 mm) in order to obtain a homogeneous material. After sieving, the contaminated mine soil was thoroughly mixed and used for the stabilization treatment. The contaminated mine soil had a total Pb concentration of approximately 2800 mg/kg and a pH of 6.7. The characteristics of the mine soil including mineralogical information are presented in Table 1. The chemical characterization of the Pb-contaminated mine soil, performed by X-ray fluorescence (XRF, ZSX100e, Rigaku, Japan), is given in Table 2.

Table 1. Characterization of the Pb-contaminated mine soil.

Soil Properties	Contaminated Mine Soil	Korean Warning Standards [1]
pH (1:5)	6.7	
Organic matter content (%) [2]	5.94	
CEC ($cmol_c$/kg)	7.59	
EC (dS/m)	1.86	
Composition (%) [3]		
Sand	47.2	
Silt	26.7	
Clay	26.1	
Texture [4]	Sandy clay loam	
Total Pb (mg/kg)	2800	200
Major mineral compositions [5]		
	Quartz, Muscovite Pyrite, Calcite Gypsum, Dolomite Arsenopyrite	

[1] Korean warning standards for soils in residential areas. [2] Organic matter content (%) determined by measured loss-on-ignition (LOI) [25,26]. [3] Soil classification determined by particle size analysis (PSA); sand, 20–2000 µm; silt, 2–20 µm; clay, <2 µm. [4] Soil texture based on the United States Department of Agriculture (USDA) classification. [5] Mineral composition obtained by the Jade software [27].

Table 2. Major chemical composition of contaminated mine soil, waste oyster shells (WOS), calcined oyster shells (COS), natural starfish (NSF), and spent-coffee-grounds-derived biochar (SCGB).

Major Chemical Composition (%)	Contaminated Mine Soil	WOS	COS	NSF	Major Chemical Composition (wt%)	SCGB
SiO_2	34.8	3.51	2.59	0.31	C	46.1
Al_2O_3	8.62	1.36	0.96	0.14	Mg	3.83
Na_2O	0.03	0.86	0.73	1.37	P	3.68
MgO	3.72	0.71	0.86	7.24	K	33.1
K_2O	1.64	0.27	0.13	0.17	Ca	11.9
CaO	12.3	88.07	87.69	86	Si	0.15
Fe_2O_3	22.3	0.53	0.40	0.06	Fe	0.53
SO_3	2.19	0.69	0.65	2.68		
MnO	5.76	0.04	0.04	0.02		
pH (1:5)	6.7	10.5	12.4	7.28		5.93

2.2. Stabilizing Agents

The raw WOS, collected at a location in Tong-young, Korea, were homogenized by pulverization to a size that cleared a #20 sieve (0.853 mm). Calcination at 900 °C for 2 h in an electric furnace (J-FM3, JISICO, Korea) converted calcite ($CaCO_3$), the main mineral phase of WOS, into quicklime (CaO), and the treated material was labeled as calcined oyster shells (COS). The starfish (*Asterina pectinifera*) used in this study was obtained from Yeosu City, Jeollanam-do Province, Korea. The collected natural starfish (NSF) was soaked in tap water for one day to remove the salty layer and other impurities. Subsequently, the NSF was subjected to multiple rinses using deionized (DI) water, was air-dried in the sun for 7 days, and was ground with a crushing blender. The resulting homogeneous powder cleared the U.S. standard #10 sieve (2 mm). The spent coffee grounds were collected from a coffee shop and put through the U.S. standard #10 sieve (2 mm). The spent-coffee-grounds-derived biochar (SCGB) was obtained by carbonization in a closed container placed in a Nabertherm N11/H furnace operating at a heating rate of 7 °C/min to reach a temperature of 200 °C without N_2 purging. The peak temperature was maintained for 2 h to complete the carbonation reaction. The chemical characterizations of the various stabilizing agents (WOS, COS, NSF, and SCGB) measured using XRF are shown in Table 2.

2.3. Stabilizing Experiments

The Pb-contaminated mine soil was treated with the stabilizing agents (WOS, NSF, SCGB) at 1~10 wt% and COS at 1~5 wt%. The control treatments for each stabilizing agent, prepared using 0 wt% mixing ratios, were useful for establishing benchmark levels. A liquid to solid (L:S) mass ratio of 20 to 1 was applied in all the treated samples to ensure full hydration. Following the stabilization treatment, all samples were subjected to a curing period of 28 days in a sealed plastic vessel maintained under ambient conditions (20 °C, 25% humidity). The treatment matrix for the Pb-contaminated mine soil using S/S agents (WOS, COS, NSF, SCGB) is presented in Table 3.

Table 3. Treatability matrix for Pb-contaminated mine soil.

Sample ID	Contaminated Mine Soil (wt%)	WOS/NSF/SCGB (wt%)	COS (wt%)	L:S Ratio
Control	100	0	0	20:1
2 wt% WOS/NSF/SCGB	100	2	0	20:1
4 wt% WOS/NSF/SCGB	100	4	0	20:1
6 wt% WOS/NSF/SCGB	100	6	0	20:1
8 wt% WOS/NSF/SCGB	100	8	0	20:1
10 wt% WOS/NSF/SCGB	100	10	0	20:1
1 wt% COS	100	0	1	20:1
2 wt% COS	100	0	2	20:1
3 wt% COS	100	0	3	20:1
4 wt% COS	100	0	4	20:1
5 wt% COS	100	0	5	20:1

2.4. Analyses of Chemical Fractions

Determination of the chemical fractions of Pb-contaminated mine soil was made by the sequential extraction procedure (SEP), according to Tessier et al. [28] The SEP designates five apparent chemical fractions identified as exchangeable (F1), weak acid soluble (F2), reducible (F3), oxidizable (F4), and residual (F5). The Pb chemical fractions were determined for the most effective treatment of each stabilizing agent (10 wt% WOS/ 5 wt% COS/10 wt% NSF/10 wt% SCGB) along with the respective control sample.

2.5. X-ray Powder Diffraction (XRPD) Analysis

The mineralogical characterization of the mine soil and stabilizing agents was performed by X-ray powder diffraction (XRPD) analysis. Sample preparation for XRPD analysis entailed pulverization into a particulate residue clearing the U.S.-standard #200 sieve (0.075 mm). An XRPD diffractometer (X'Pert PRO MPD, PANalytical, Almelo, The Netherlands) was employed for collecting step-scanned diffraction patterns. The diffracted beam graphite monochromator with Cu radiation was used at 40 kV and 40 mA. The XRPD patterns were collected in the 2θ range of 5–65°, a step size of 0.02°, and a count time of 3 s/step. Mineral characterizations were performed by means of the Jade software v. 7.1 [27] and the PDF-2 reference database [29].

2.6. SEM-EDX Analyses

The treated samples demonstrating the highest decrease in leached Pb for each stabilizing agent were selected for the SEM-EDX testing. For the SEM-EDX analysis, sample aliquots positioned on a double-sided Pt-coated carbon tape were analyzed in a Hitachi S-4800 SEM instrument (Tokyo, Japan) equipped with Horiba EMAX EDX system (Tokyo, Japan). The lowest Pb leachability samples for each stabilizing agent, namely the 10 wt% WOS, 5 wt% COS, 10 wt% NSF, and 10 wt% SCGB, were analyzed to assess the Pb immobilization mechanism.

2.7. Physicochemical Analyses

The pH values of mine soil and the stabilizing agents were obtained based on the Korean Standard Test (KST) method [30] at a liquid-to-solid (L:S) mass ratio of 5:1. The stabilization effectiveness of the contaminated mine soil was assessed using the TCLP test [31]. For this purpose, 3 g of a mine tailing sample, mixed with 60 mL of the extraction solution, was shaken for 18 h in a tumbler. The TCLP Pb concentration was analyzed using an inductively coupled plasma-optical emission spectrometer (ICP-OES, Optima 8300DV, PerkinElmer, Waltham, CT, USA). The total Pb levels were determined using an aqua regia extraction agent consisting of 0.7 mL HNO_3 (65%, Merck, Kenilworth, NJ, USA) and 2.1 mL of HCl (37%, J.T. Baker, Phillipsburg, NJ, USA) [30]. All the determinations by ICP-OES were reported as the mean values (within a 10% error) of triplicate samples. The QA/QC protocol involved three quality-control standards for every 10 samples analyzed and a sample spiking with standard addition every 10 samples (recovery rate >95%).

3. Results and Discussion

3.1. Effectiveness of the Stabilization Treatment

The TCLP Pb levels and the associated pH values for the WOS, COS, NSF, and SCGB treatments are presented in Figures 1–4. In general, the TCLP Pb concentration decreased with increasing stabilizing agent dose.

In the WOS treatments, 4 wt% WOS was required to meet the Pb TCLP regulatory limit of 5 mg/L. The lowest Pb leachability (0.95 mg/L) was obtained with the 10 wt% WOS treatment, resulting in a decrease higher than 85% in the TCLP Pb level for the treated sample compared to that for the control (Figure 1). A similar result for Pb leachability reduction (>90%) has been reported for contaminated paddy soil upon 5 wt% WOS treatment using 0.1 N HCl extraction [32]. Moreover, a recent study using oyster shell powder (OSP) in artificially contaminated river sediment showed a significant Pb leachability decrease from 810.8 to 108.6 µg/L [33].

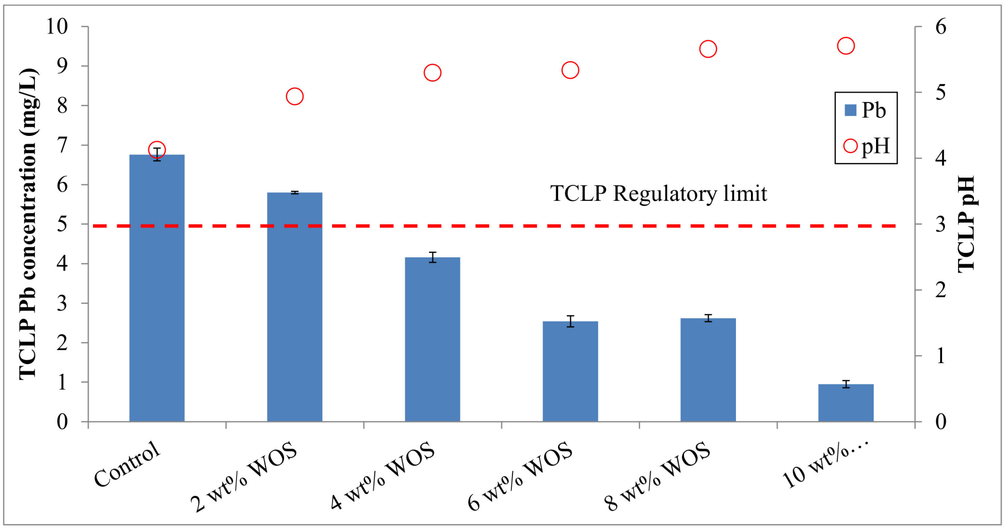


Figure 1. TCLP Pb concentrations for the control sample and waste oyster shells (WOS)-treated samples.

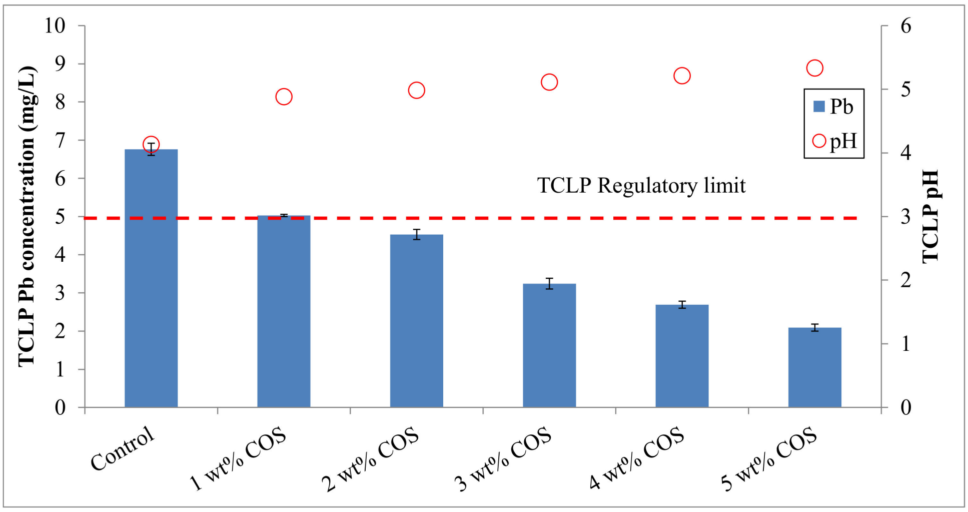


Figure 2. TCLP Pb concentrations for the control sample and calcined oyster shells (COS)-treated samples.

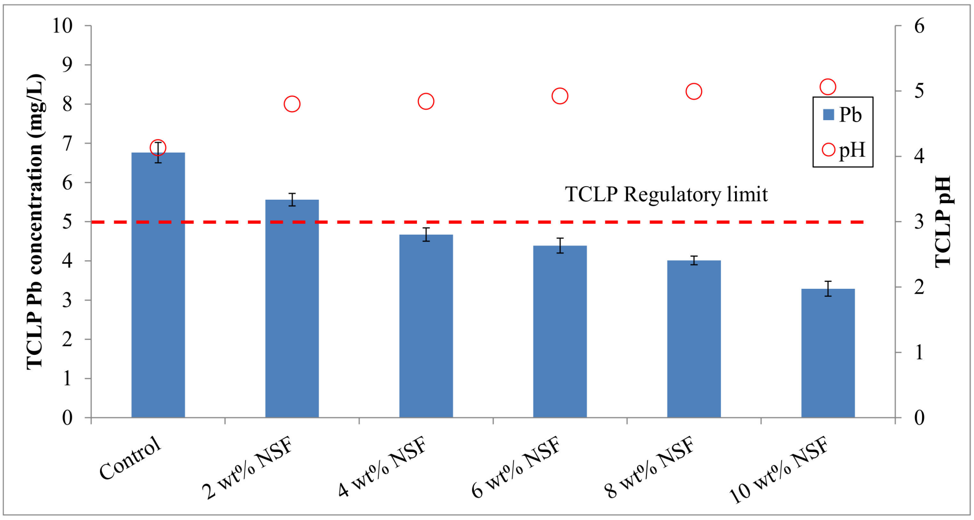


Figure 3. TCLP Pb concentrations for the control sample and natural starfish (NSF)-treated samples.

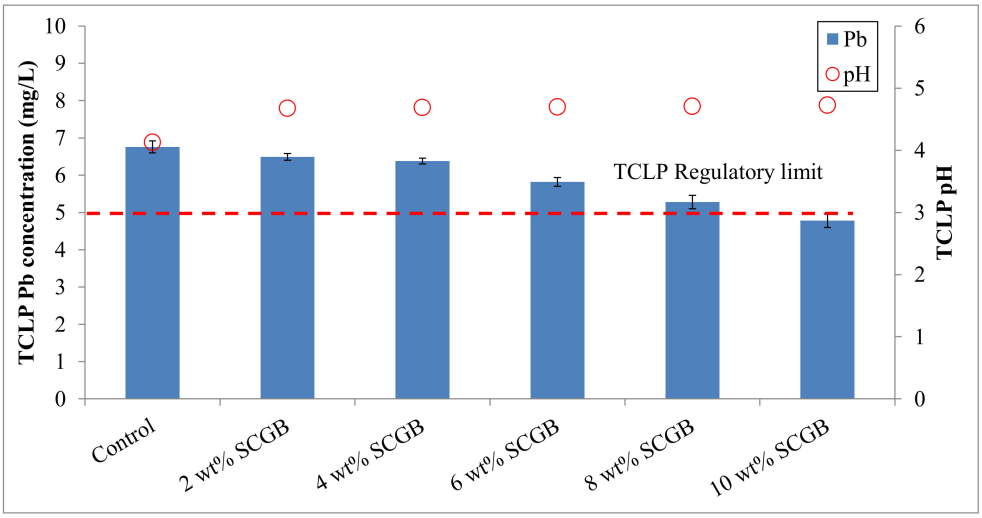


Figure 4. TCLP Pb concentrations for the control sample and spent-coffee-grounds-derived biochar (SCGB) treated sample.

The COS treatment outperformed the WOS treatment. More specifically, 2 wt% COS sufficed to meet the TCLP Pb regulatory limit (5 mg/L). The least TCLP Pb leachability level (2.1 mg/L), recorded for the 5 wt% COS treatment, represents a TCLP Pb leachability reduction higher than 68% compared to the control (Figure 2). In comparison to WOS treatment, COS treatment at half the WOS dosage was adequate for meeting the TCLP Pb regulatory limit. Similarly, the COS treatment outperformed the WOS treatment in the Cd- and Pb-contaminated paddy soil. Ok et al. [32] have reported that the Pb leachability reduction was more prominent for the COS rather than the WOS treatment. In terms of the mechanism responsible for Pb immobilization, highly effective Pb immobilization upon COS treatment may be linked to the mineral phases of cementitious reactions, including calcium silicate hydrates (CSHs) and calcium aluminum hydrates (CAHs). It has been reported that CSHs and CAHs were strongly associated with highly effective heavy metal immobilization upon lime treatments [8,15,34].

In the case of the NSF treatment, a 4 wt% NSF dosage was adequate for meeting the 5 mg/L TCLP regulatory limit. Comparison of the NSF to the WOS treatment reveals a larger reduction in TCLP Pb leachability for the latter than the former. This indicates that WOS was more beneficial than the NSF for immobilizing Pb in contaminated mine soil. The lowest Pb leachability (3.3 mg/L) was attained with the 10 wt% NSF treatment (Figure 3). Lim et al. [19] reported that more than 75% in TCLP extractable Pb was attained in contaminated agricultural soil withe 5 wt% NSF treatment. Moreover, the NSF treatment could be enhanced through the calcination process. It has also been reported that the calcined starfish (CSF) treatment was better at immobilizing Pb in contaminated soil than the NSF treatment [16,19]. Therefore, the use of the CSF could be considered as a potential treatment for Pb-contaminated mine soil. Overall, NSF can be used to effectively immobilize Pb in mine soil but was not as effective as WOS and COS.

For the SCGB treatment, 10 wt% was necessary to generate treatments that were capable of conforming with the applicable TCLP Pb regulatory limit (5 mg/L). Approximately 30% of the TCLP Pb leachability was reduced upon 10 wt% SCGB treatment (Figure 4). This indicates that the SCGB treatment showed the poorest Pb immobilization performance among all stabilizing agents used (WOS, COS, NSF). Moreover, larger dosages would be needed in order to provide similar performance for Pb immobilization with WOS, COS, and NSF. In previous research, using soybean stover-derived biochar, an approximately 50% reduction in TCLP Pb leachability was attained upon the 10 wt% biochar treatment, which is higher than that obtained in this study with the SCGB treatment. This may be due to the carbonization temperature being set at 200 °C in this study compared to 700 °C for the soybean stover-derived biochar [18]. The carbonization temperature is strongly associated with the biochar pH. The initial pH of the soybean stover-derived biochar was 10.5, while it was 5.93 for the SCGB used in this study. This indicates that pozzolanic reaction products (CSHs and CAHs) may not play a role in reducing Pb mobility. In a previous research study with soybean stover-derived biochar, both pozzolanic reaction products and Pb-P precipitates were responsible for the very effective Pb immobilization. However, this was not the case in this study, where only Pb-P compounds may be playing a role in immobilizing Pb in contaminated mine soil.

3.2. Sequential Extraction Results

Sequential extraction procedure (SEP) results for the samples treated with 10 wt% waste oyster shells (WOS), 5 wt% calcined oyster shells (COS), 10 wt% natural starfish (NSF), and 10 wt% spent-coffee-grounds-derived biochar (SCGB) are presented in Figure 5.

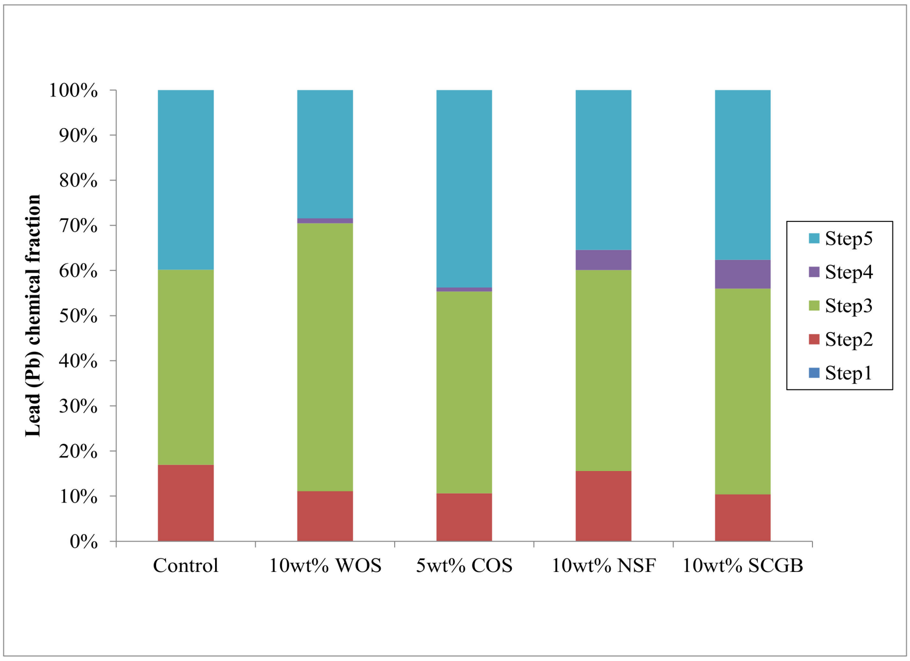


Figure 5. Lead (Pb) chemical fractions for the control sample, 10 wt% waste oyster shells (WOS), 5 wt% calcined oyster shells (COS), 10 wt% natural starfish (NSF), and 10 wt% spent coffee grounds derived biochar (SCGB) treated samples.

The selection of these samples was based on the lowest Pb leaching performance for each of the stabilizing agents (WOS, COS, NSF and SCGB). According to the results, the weak acid-soluble fraction (F2) decreased in all treated samples. Specifically, the values of the F2 fraction were 11.1%, 10.6%, 15.5%, and 10.4% for 10 wt% WOS, 5 wt% COS, 10 wt% NSF, and 10 wt% SCGB, respectively. In addition, the F2 fraction values of the treated samples were lower than that of the control sample which exhibited a value of 16.9%. The reducible (F3) and oxidizable (F4) fractions increased in the treated samples. Moreover, a significant increase in the residual fraction (F5) was recorded for the 5 wt% COS-treated sample. Overall, the results indicate that the high degree of Pb immobilization in the treated samples is probably associated with the reduction in the F2 fraction and an increase in the F3 and F4 fractions. The high degree of immobilization achieved with the 5 wt% COS was probably related to the increase of the F5 fraction. It has been reported that an increase in the F5 fraction was clearly observed upon the 5 wt% calcined starfish (CSF) treatment where CaO was the main mineral phase [16].

3.3. XRPD Analyses

The X-ray powder diffraction (XRPD) analysis results for the lead (Pb)-contaminated mine soil and the stabilizing agents are presented in Figures 6 and 7. Accordingly, quartz (PDF# 46-1045), muscovite (PDF# 07-0042), pyrite (PDF# 42-1340), calcite (PDF# 05-0586), gypsum (PDF# 33-0311), dolomite (PDF# 36-0426), and arsenopyrite (PDF# 42-1320) are the dominant mineral phases detected in the Pb-contaminated mine soil (Figure 6).

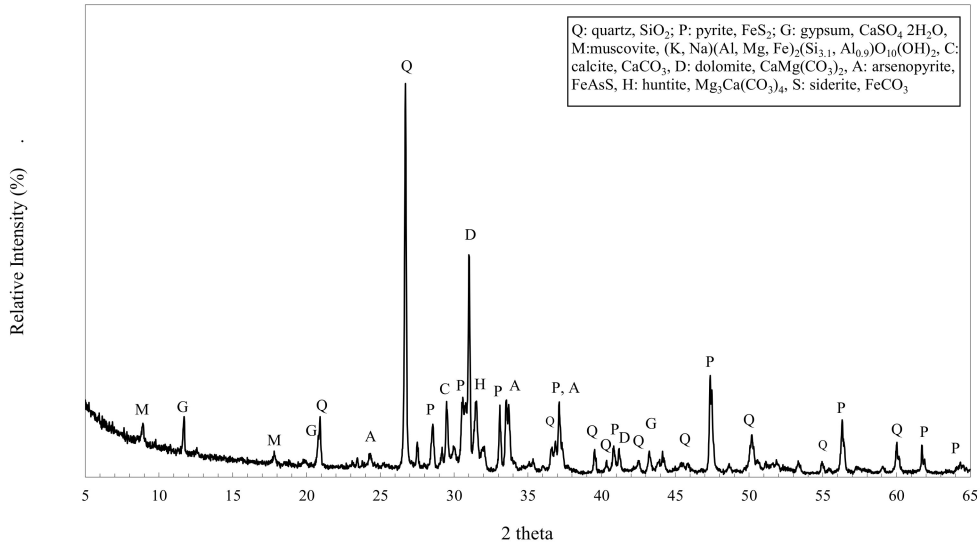


Figure 6. XRPD pattern for contaminated mine soil.

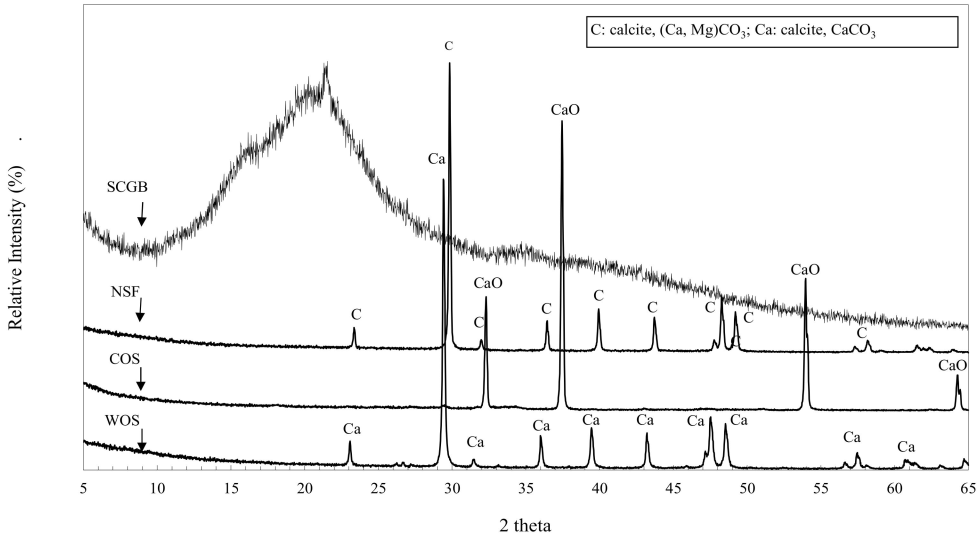


Figure 7. XRPD patterns for the waste oyster shells (WOS), calcined oyster shells (COS), natural starfish (NSF), and spent-coffee-grounds-derived biochar (SCGB).

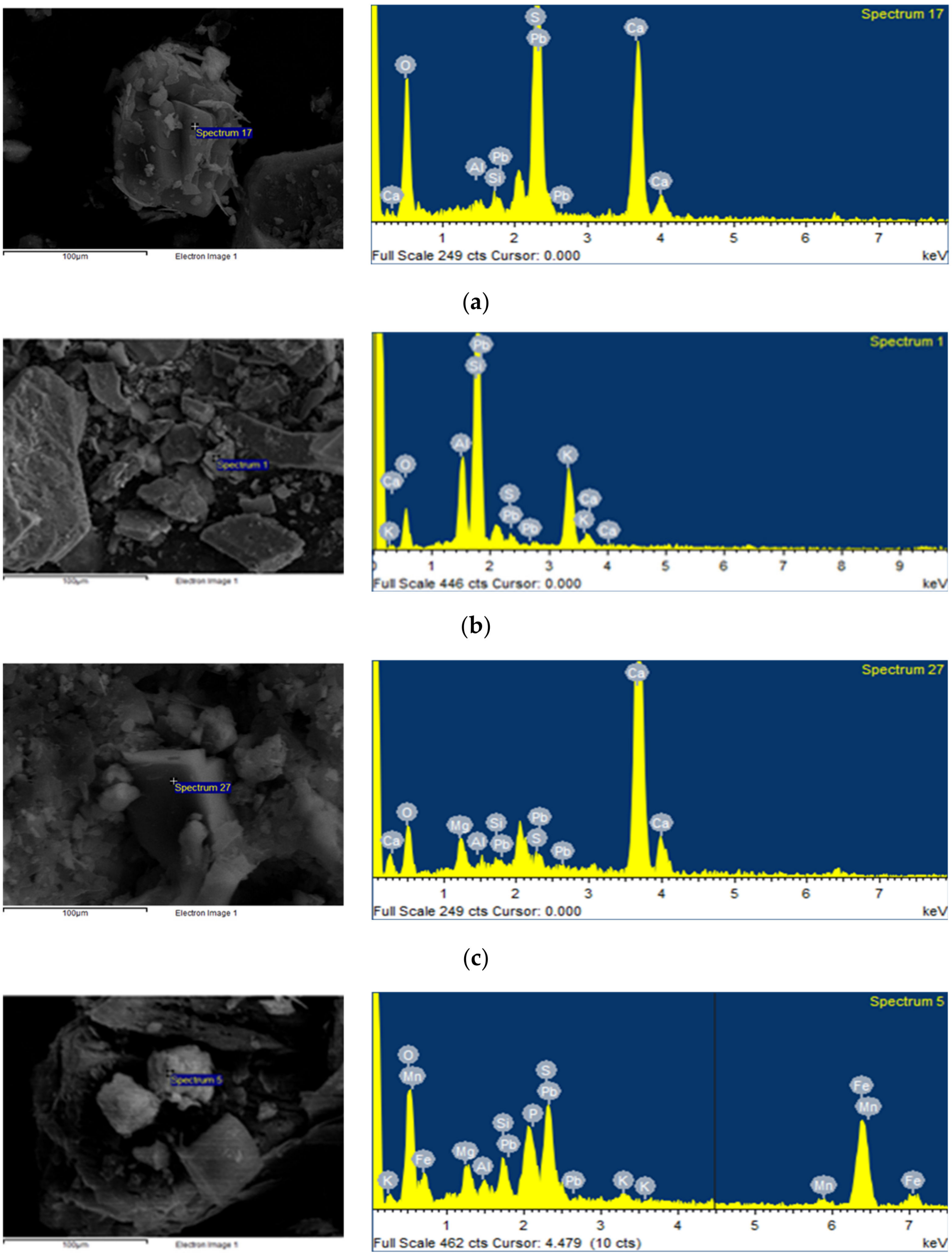
The XRPD results for the stabilizing agents indicate that a calcite (PDF# 47-1743, $CaCO_3$) was observed in the waste oyster shells (WOS) sample while a different type of calcite (PDF# 43-0697, (Ca, Mg)CO_3) was identified in the natural starfish (NSF) sample (Figure 7). This indicates that an Mg source, present in the NSF, does not exist in the WOS sample. Through the calcination process, which was conducted at 900 °C for 2 h, the existing calcite ($CaCO_3$) in the WOS was completely transformed into quicklime (CaO) (PDF# 48-1467) in the calcined oyster shell (COS) sample (Figure 7). This indicates that the total decomposition of $CaCO_3$ was achieved at 900 °C for 2 h. $CaCO_3$ can be transformed into CaO by a chemical decomposition reaction during the calcination process (Equation (1)).

$$CaCO_{3(solid)} + \text{Heat } (\geq 900\ ^\circ\text{C for 2 h}) \rightarrow CaO_{(solid)} + CO_{2(gas)} \quad (1)$$

Based on literature, the theoretical decomposition temperature for $CaCO_3$ is 848 °C when the reaction standard Gibb's free energy is equal to zero [35]. Moreover, no crystalline phase was identified in the SCGB (Figure 7).

3.4. SEM-EDX Analysess

The SEM-EDX results for the lowest Pb leachability samples (10 wt% WOS, 5 wt% COS, 10 wt% NSF, and 10 wt% SCGB) are presented in Figure 8.



(**a**)

(**b**)

(**c**)

(**d**)

Figure 8. SEM-EDX analyses for the Pb in the contaminated soil treated with 10 wt% waste oyster shells (WOS) (**a**); calcined oyster shells (COS) (**b**); natural starfish (NSF) (**c**); and spent coffee grounds derived biochar (SCGB) (**d**) treated samples.

In the samples treated with WOS (10 wt%), COS (5 wt%), and NSF (10 wt%), Pb, Ca, Al, Si, and O were identified by the SEM-EDX analysis. This indicate that the effective Pb immobilization is presumably linked to pozzolanic reaction products (calcium silicate hydrates, CSHs, and calcium aluminum hydrates, CAHs). This mechanism controlling Pb immobilization has also been reported in previous studies using COS [15,36]. Moreover, it has been reported that the increase in the pH upon application of $CaCO_3$ and CaO could enhance the adsorption of metal cations onto soil particles [37] and consequent formation of the insoluble precipitates (metal hydroxides/carbonates) under alkaline conditions [32,37,38]. Therefore, effective Pb immobilization may be achieved by a combination of the aforementioned reactions. Moreover, in the case of the SCGB-treated sample, P was also identified with Pb (Figure 8). This indicates that a Pb-P compound, such as pyromorphite, may also be the phase responsible for the effective Pb immobilization. This has also been reported in previous research [18]. Moreover, it has been reported that chloropyro-morphite [$Pb_5(PO_4)_3Cl$] and hydroxylpyro-morphite [$Pb_5(PO_4)_3OH$] compounds were responsible for the effective Pb immobilization upon soybean stover and pine-needles-derived biochar treatment [23].

4. Conclusions

In this study, natural waste materials (WOS, COS, NSF, SCGB) were utilized as sustainable stabilizing agents for the immobilization of lead (Pb) in contaminated mine soil. The stabilization treatment results showed that the reduction of TCLP Pb leachability was strongly associated with an increasing application dosage for all of the stabilizing agents. The application dosages required to meet the TCLP regulatory limit of 5 mg/L were: 4 wt% WOS, 2 wt% COS, 4 wt% NSF, and 10 wt% SCGB. The most effective natural waste material was found to be COS while the least effective stabilizing agent was SCGB. Based on the presented results, the selected four natural waste materials could be used as effective stabilizing agents to immobilize the Pb in contaminated mine soil. The optimum dosage applied to the Pb-contaminated mine soil for each stabilizing agent should be carefully evaluated based on soil properties, contaminant type, and concentration.

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Conflicts of Interest: The authors declare no conflict of interest.

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