

LEAD LEACHABILITY FROM QUICKLIME TREATED SOILS IN A DIFFUSION CONTROLLED ENVIRONMENT

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Abstract : The effectiveness of quicklime-based stabilization/solidification (S/S) in immobilizing lead (Pb) was assessed by performing semi-dynamic leaching tests (ANS16.1). In order to simulate landfill leaching conditions, the ANS 16.1 test was modified by using 0.014 N acetic acid (pH = 3.25) instead of distilled water. Artificial soil samples as well as field soil samples contaminated with Pb were tested. The effectiveness of quicklime treatment was evaluated by determining diffusion coefficients (D_e) and leachability indices (LX). A model developed by de Groot and van der Sloot was used to elucidate the controlling Pb leaching mechanisms. Overall, upon quicklime treatment Pb leachability was significantly reduced in all of the samples tested. The mean LX values were higher than 9 for an artificial soil sample containing 30% kaolinite treated with 10% quicklime and for a field soil sample treated with 10% quicklime, which suggests that S/S treated soils can be considered acceptable for "controlled utilization". Moreover, quicklime treatment was more effective in artificially contaminated soil with high kaolinite content (30%), indicating the amount of clay plays an important role in the success of the treatment. The controlling Pb leaching mechanism was found to be diffusion, in all quicklime treated samples.

Key Words : lead (Pb), semi-dynamic leaching test (ANS16.1), stabilization/solidification (S/S), quicklime, diffusion coefficient (D_e), leachability index (LX), diffusion

INTRODUCTION

Lead (Pb), which is listed as a priority pollutant by the EPA (EPA code D0008)¹⁾, has been identified as one of the most toxic elements to human health and is a widespread contaminant in many hazardous wastes sites.²⁾ It has been reported that Pb can cause damage to the human nervous system, blood vessels, kidneys, the brain, red blood cells and digestive systems.^{2,3)} Human exposure to Pb is intensified because of industrialization. This exposure is

primarily due to leaded gasoline, Pb storage batteries, leaded paints, Pb-ammunitions, leaded solder in electronics and plumbing equipments, and Pb-contaminated food cans.

Due to this widespread use of Pb over a long period of time, the Pb loading rate in soil is approximately 20 times (or higher than) its natural removal rate.⁴⁾ As a result, the risk of groundwater due to leaching of Pb from landfill areas and industrially contaminated land has received increasing attention.⁵⁾

Stabilization/solidification (S/S) process is one of the most effective methods to treat soils with heavy metal contamination.^{6,7)} By applying S/S process, the hazardous potential of waste materials can be minimized by converting the

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contaminant into forms which are less-soluble, less mobile or less toxic and encapsulating the waste within a monolithic solid of high structural integrity.⁷⁾ Various stabilizing agents have been used by numerous researchers in the treatment of soils contaminated with Pb. Li et al.⁸⁾ investigated Pb immobilization using Portland cement (OPC) and pulverized fly ash (PFA). Thevenin and Pera¹⁾ studied Pb interactions with OPC and ground-granulated blast furnace slag (GGBFS). Long and Zhang³⁾ used cement in combination with various additives such as lime, ash, clay, apatite, and silicate for treating Pb contaminated soils. Wang and Vipulanandan⁹⁾ used Type 1 OPC and class C fly ash as a stabilizing agent to evaluate Pb leachability. Dong Jin Lee used OPC and calcite to evaluate the leachability of Pb-doped solidified waste forms.¹⁰⁻¹⁴⁾

In this study, quicklime was used as the main stabilizing agent rather than cement and hydrated lime because: a) of its economic advantage over cement and hydrated lime b) of the increase in temperature due to the heat of hydration results in an accelerated rate of reaction c) of the limited information available to date on quicklime-based S/S for Pb.

Upon quicklime treatment of Pb contaminated soils, there are three possible Pb immobilization mechanisms: precipitation, inclusion and sorption. Several researchers have shown that Pb immobilization can be achieved by the formation of insoluble precipitates. Palomo and Palacios¹⁵⁾ have suggested that the formation of lead silicate (Pb_3SiO_5) controls the immobilization of Pb in contaminated soils. Moulin et al.¹⁶⁾ also suggested that Pb can be retained in the solid through the formation of Si-O-Pb bonds. Inclusion can be defined as either physical inclusion or chemical inclusion. Physical inclusion can be achieved by creating a monolithic solid, while chemical inclusion can be achieved at high pH (approximately 12.8) conditions through incorporation of Pb into newly formed pozzolanic reaction products, such as calcium silicate hydrates (CSH) and calcium aluminate hydrates

(CAH).¹⁷⁾ At high pH conditions the solubility of alumina and silica in clay minerals is greatly increased and alumina and silica are available for reaction with water and calcium derived from lime to form various pozzolanic reaction products. Sorption on clays and CSH/CAH can also be considered as a possible immobilizing mechanism for Pb.

The effectiveness of the treatment can be assessed by performing leaching tests. Leaching is known to be a complex process because several factors may influence the release of specific constituents from a waste over a period of time.¹⁸⁾ These factors include: major element chemistry, pH, redox potential, complexation reactions, liquid-to-solid ratio, and contact time etc.¹⁸⁾ Moreover, since very little is known about the chemical species in different waste forms, as well as their behavior with respect to time, the long-term performance of S/S-treated waste has been difficult to predict.

The objectives of this study are: 1) to evaluate the effectiveness of quicklime treatment in increasing the degree of Pb immobilization 2) to determine the controlling Pb leaching mechanisms (e.g. wash off, diffusion and dissolution) in quicklime treated soils 3) to validate results derived from artificially contaminated soils, by applying the same quicklime treatment to Pb contaminated field soils.

REVIEW OF DIFFUSION MODEL

ANS model, Diffusion Coefficient (D_e) and Leachability Index (LX)

The leachability of Pb from quicklime treated soils was evaluated using the ANS 16.1 model.¹⁹⁾ This model was established based on Fick's diffusion theory and standardized by ANS.¹⁹⁾ This model can be used to determine the cumulative fraction of Pb leached against time. It has been widely reported that the leaching of the contaminants from cement-treated waste is mostly a diffusion-controlled process.^{20,21)} Due to the slow diffusion rate of contaminants, it can be assumed that a quicklime-treated waste

form is a semi-infinite medium, much like the cement-treated waste examined in previous studies.²¹⁻²³ This result implies that the release of the contaminants from the treated waste is negligible compared to its total mass. According to preliminary column percolation tests performed on specimens identical to those used in this study, the hydraulic conductivity values were low (between 2×10^{-6} and 5×10^{-10} cm/sec). As a result, diffusion is expected to be the controlling leaching mechanism in soils treated with quicklime. The diffusion coefficient (D_e) and the leachability index (LX) are the most important parameters to assess leaching behavior of contaminants in the treated soil. The D_e of the leached samples was determined based on the following ANS 16.1 model¹⁹:

$$D_e = \pi \cdot \left[\frac{\left(\frac{a_n}{A_0} \right)}{(\Delta t)_n} \right]^2 \cdot \left[\frac{V}{S} \right]^2 \cdot T_n \quad (1)$$

where a_n = the activity of a metal ion released from the specimen during the leaching interval n , A_0 = the total activity of a given metal ion in the specimen at the beginning of the first leaching interval, $(\Delta t)_n = t_n - t_{n-1}$, V = the volume of specimen in cm^3 , S = the geometric surface area of the specimen as calculated from measured dimensions in cm^2 , and T_n is the elapsed time to the middle of the leaching period n in seconds. D_e is the effective diffusion coefficients (cm^2/sec). The D_e values from equation (1) are termed "effective" because diffusion occurs in the liquid filling the interstitial spaces of a porous body.

An evaluation of the effectiveness of quicklime-based S/S was assessed by determining the leachability index (LX). According to Environment Canada²⁴, LX can be used as a performance criterion for the utilization and disposal of S/S waste. When LX values are higher than 9, a treatment process can be

considered effective and S/S wastes could be used in "controlled utilization". Examples of controlled utilization are quarry rehabilitation, lagoon closure, and road construction etc. When LX values are higher than 8, S/S wastes can be used in segregated or sanitary landfills. S/S waste with an LX value lower than 8 is not considered appropriate for disposal. The leachability index is defined using the following formula:

$$LX = \frac{1}{m} \cdot \sum_{i=1}^m [-\log(D_e)]_n \quad (2)$$

where n is the number of the particular leaching period, and m is the total number of individual leaching periods.

Determination of Controlling Pb Leaching Mechanisms

In order to determine the controlling Pb leaching mechanisms, a model developed by de Groot and van der Sloot²⁵ was used. The controlling Pb leaching mechanism was determined based on the slope of the plot of the logarithm of cumulative fraction release, $\log(B_t)$, versus the logarithm of time, $\log(t)$.²⁵ If diffusion is the dominant leaching mechanism, theory suggests the following relationship:

$$\log(B_t) = \frac{1}{2} \cdot \log(t) + \log \left[U_{\max} \cdot d \cdot \sqrt{\left(\frac{D_e}{\pi} \right)} \right] \quad (3)$$

where D_e = the effective diffusion coefficient in cm^2/sec for component x (Pb in this study), B_t = the cumulative maximum release of the component in mg/m^2 , t = the contact time in seconds, U_{\max} = the maximum leachable quantity in mg/kg , d = the bulk density of the product in kg/m^3 .

According to de Groot and van der Sloot²⁵, if the slope from Equation 3 is close to 0.5, Pb release will be slow and diffusion will be the controlling mechanism. At a slope of approxi-

mately 1, the controlling Pb leaching mechanism is dissolution. In this case, the dissolution of material from the surface proceeds faster than diffusion through the pore space of the soil matrix. Occasionally, a soluble layer exists on the surface of the material. During the beginning of the leaching tests, most of the soluble material on the surface will dissolve. This process, known as surface wash-off, typically results in a slope close to 0. Both dissolution and surface wash-off processes will result in the release of highly soluble compounds. However, the material is not expected to be depleted during the dissolution process.

EXPERIMENTAL METHODOLOGY

Reagents and Materials

Analytical grade lead oxide (PbO) was used as the source for Pb and provided by Fisher Scientific Company (Suwanee, GA, USA). Kaolinite was obtained from Dry Branch Kaolin (Dry Branch, GA, USA). Chemical grade quicklime (CaO) powder (95% CaO) was provided by the Bellefonte Lime Company (Naperville, IL, USA). The chemical and physical properties of kaolinite and quicklime are presented in Table 1.

Table 1. Summary of chemical and physical properties

	Kaolinite	Quicklime
SiO ₂ (dry wt %)	45.7	1.2
Al ₂ O ₃ (dry wt %)	38.5	-
Iron Oxide (dry wt %)	0.4	-
CaO (dry wt %)	0.2	95.4
MgO (dry wt %)	0.1	0.85
SO ₃ (dry wt %)	-	0.012 (as S)
Na ₂ O (dry wt %)	0.04	-
K ₂ O (dry wt %)	0.1	-
TiO ₂ (dry wt %)	1.4	-
Surface area (m ² /g)	66	40.0-41.5
pH	4.0-6.5	12.9

Sample Preparation

In this study, clay and fine quartz sand were

combined in order to prepare artificial soils. Mixtures of clay and sand were used rather than pure clay to provide samples with a gradation more comparable to those of natural soils. Moreover, these mixtures are easier to compact than pure clay. Kaolinite was used as the clay source and the amount of kaolinite present was varied in order to evaluate its relative contribution to Pb immobilization. Lead oxide (PbO) was added to the kaolinite-sand mixes at a concentration level of 7,000 mg lead (Pb²⁺) per kg of solid of untreated soils. This initial Pb²⁺ concentration was used in accordance with the sponsor's (US Department of Energy) request.

A field soil sample collected from the Cataract Creek tailing facility, located in Montana, USA, was also tested in this study. Its total Pb concentration and pH value were 3,530 mg/kg and 5.9, respectively. It was classified as a uniformly graded fine-silty sand.

Six types of specimens were prepared; K15L0, K15L10, K30L0, K30L10, TL0 and TL10. The letters in the specimen designation reflect the sample mineralogy, i.e., K: kaolinite, T: Cataract Creek tailings and L: quicklime. The numbers following the letters indicate the weight percent of the preceding attributes. For artificial soils, since the same type of fine quartz sand was added in all mixes, sand was not included in the specimen designation. Sand content is always complimentary to the kaolinite content on a 100% w/w basis. For example, the specimen designation K15L10 stands for 15% kaolinite and 85% fine quartz sand at a treatment level of 10% quicklime added on the basis of the total weight (kaolinite-sand). The same treatment level of 10% quicklime was used in a field soil sample.

ANS 16.1 Tests and Analysis

The ANS 16.1 method was modified by using a 0.014N acetic acid solution (pH = 3.25) instead of distilled water. This modification was conducted in order to simulate the landfill leaching conditions of S/S treated waste being disposed of in a landfill environment. In this

study, specimens with a 4.0 ± 0.4 cm height and a 4.70 ± 0.05 cm diameter were prepared by compaction (in accordance with ASTM D1557-91) at optimum water content.²⁶⁾ Following compaction, all specimens were cured for 28 days in sealed sample bags at 20°C. Prior to the initiation of the specimen leaching test, the solids were immersed in distilled water for 30 seconds to rinse out any loose particles from the specimen surface. A nylon mesh harness was used in order to suspend each specimen near the centroid of the acetic acid solution in a polyethylene container. As specified by the ANS 16.1 method, the ratio of leachant volume (V_L) to the specimen external surface area (S) was retained at 10 ± 0.2 cm. This ratio is maintained in order to minimize leachant composition changes as well as provide an ample concentration of extracted species for analysis.¹⁹⁾ The leachate was collected and entirely replaced at designated time intervals (2, 7, 24, 48, 72, 96, 120, 456, 1128, and 2160 hours). The sampled leachate was separated using a 0.4 μ m pore-size membrane filter. The concentrations of soluble Pb were analyzed with a Zeeman Furnace Atomic Absorption Spectrometer (AAS) (Varian SpectraAA- 400).

RESULT AND DISCUSSION

Cumulative Release of Pb with and without S/S Treatment

The cumulative fraction of Pb leached (%) from untreated and quicklime treated specimens

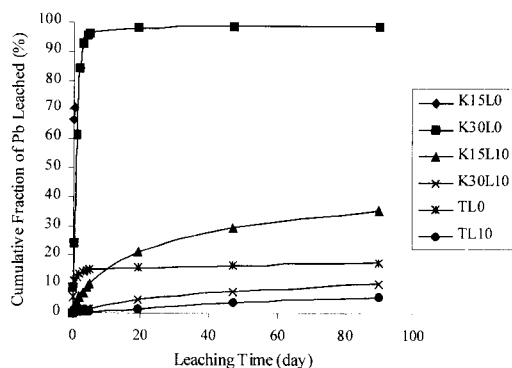


Figure 1. Cumulative fraction of Pb leached (%) as a function of time.

for artificial and field soil samples are plotted as a function of leaching time in Figure 1. The ultimate cumulative fractions of Pb leached (%) from all samples upon test completion (90 days) were summarized and presented in Table 2.

Table 2. Cumulative fraction of Pb leached (%) following test completion

Sample	Cumulative fraction of Pb leached (%)
K15L0	70.1*
K30L0	98.9
K15L10	35.3
K30L10	9.99
TL0	17.50
TL10	5.44

*Note: K15L0 sample disintegrated after 7 hours of testing

In untreated artificial soil samples, an increased amount of kaolinite (from 15% to 30%) led to decreased amounts of Pb leached (Figure 1 and Table 1). The K15L0 sample released 71% of the total lead within the first 7 hours of testing and then disintegrated, so that no more results are available for this sample. The K30L0 sample retained more lead in the matrix, releasing only 24% in the first 7 hours and maintained structural stability until the end of the test, but a total of 99% of Pb was released when the experiment concluded after 90 days. This indicated that sorption on kaolinite is considered the main mechanism responsible for Pb immobilization in untreated samples. However, kaolinite is known as inert clay in terms of cation exchange capacity (CEC) and surface area. Therefore, the sorption capacity for Pb immobilization is limited as shown in Figure 1; approximately 99% of Pb leached out from the K30L0 sample after test completion. Upon quicklime treatment, Pb leachability was significantly reduced in all samples. Even though the K15L0 sample disintegrated after 7 hours of testing, results from the K15L10 sample, within the same time period, showed significant Pb reduction (more than 65%) (Figures 1 and Table 2). Moreover, Pb leachability was reduced by approximately 90% in the K30L10 sample, as compared to the K30L0 sample (Figure 1 and

Table 1). Among all treated artificial soil samples, Pb immobilization was most effective in the K30L10 sample (9.99% Pb release) and least effective in the K15L0 sample (70.1% Pb release within 7 hours of testing) (Figure 1 and Table 1).

The untreated field soil sample (TL0) leached 17.5% of the cumulative Pb fraction. This is mainly due to the low pH condition (pH = 5.9). Therefore, the leachable fraction of Pb was most probably leached out in the field so that only 17.5% could be leached out at pH values less than 4.5. However, the leachable Pb fraction was further reduced to 5.4% upon quicklime treatment (Figure 1 and Table 2). This further reduction is most probably due to pozzolanic reaction products such as CSH and CAH and insoluble Pb precipitates such as lead silicate. However, the formation of pozzolanic reaction products should be limited because of the small amount of clay. This indicates that Pb immobilization by quicklime treatment can be successful unless it is limited by low clay content.

Overall, sorption in artificially contaminated soils was found to be the main mechanism in untreated samples at low pH. However, upon quicklime treatment the significant reduction in Pb release was probably due to the formation of pozzolanic reaction products and insoluble Pb precipitates such as CSH and CAH, and lead silicate, respectively. These mechanisms were more pronounced in samples containing higher kaolinite percentages (30%) since high silicate and alumina sources can be obtained from kaolinite at high pH. However, this is not the case in the field soil samples due to the limited amounts of clay.

The Effect of Leachant pH on Pb Leachability

The leachant pH was monitored at designated time intervals in accordance with ANS 16.1 method. The leachant pH values of untreated and treated artificial and field soil samples versus leaching time (days) are presented in

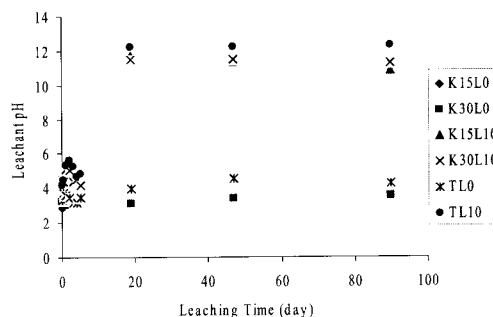


Figure 2. Leachant pH during semi-dynamic leaching tests.

Figure 2. The leachant pH values were less than 4.5 for all untreated samples. However, upon quicklime treatment the leachant pH values were significantly increased (higher than 11) after 5 days of testing. This is due to the calcium consuming the buffering capacity of the liquid after 5 days of testing. At high pH values (greater than 12), the predominant lead species are anionic such as $\text{Pb}(\text{OH})_3^-$ and $\text{Pb}(\text{OH})_4^{2-}$.²⁷ In the same pH region, kaolinite particles are negatively charged.²⁸ Therefore, the adsorption capacity of Pb is very limited and Pb immobilization is mainly due to the formation of pozzolanic reaction products and insoluble Pb precipitates.

The Controlling Leaching Mechanisms and Effectiveness of Quicklime Treatment

The controlling leaching mechanisms were evaluated by using a diffusion model (Equation 3) developed by de Groot and van der Sloot.²⁵ The cumulative release of Pb from samples K30L10, TL0, and TLI plotted as a function of time (Figure 3), as an example. All slope and R^2 values obtained from the diffusion model are presented in Table 3.

For untreated artificial soil samples, K15L0 and K30L0, slope values were 0.05 and 0.18, respectively (Table 3). This shows that surface wash-off appeared to be the main controlling leaching mechanism. Upon quicklime treatment, slope values for K15L10 and K30L10 samples were 0.51 and 0.59, respectively (Table 3).

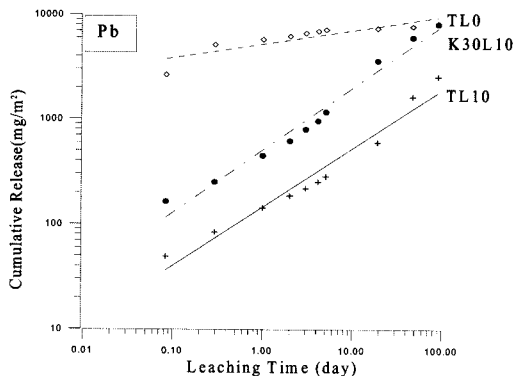


Figure 3. Logarithm of the cumulative release of the Pb versus the logarithm of time for sample K30L10, TL0 and TL10.

Table 3. Regression analyses results for Pb release

Sample	Slope	R ²
K15L0	0.05*	-
K30L0	0.18	0.86
K15L10	0.51	0.99
K30L10	0.59	0.98
TL0	0.13	0.76
TL10	0.56	0.96

*Note: K15L0 sample disintegrated after 7 hours of testing

These slope values suggest that Pb release was controlled by diffusion.

Similarity, in field soil samples, the slope value for the untreated sample (TL0) was 0.13 and it increased to 0.56 upon quicklime treatment. This indicated that diffusion is the controlling Pb leaching mechanism. Overall, the results obtained from artificial and field soils indicate that Pb release from quicklime treated samples was mainly controlled by diffusion. Numerous researchers have previously reported this conclusion. Côté et al.²²⁾ confirmed that diffusion was the main controlling leaching mechanism for Pb release in fly ash-lime treated waste. Andrés et al.²³⁾ have also demonstrated that diffusion was the dominant mechanism governing Pb release in stabilized steel foundry dusts.

Diffusion coefficients (D_e) and LX values (defined by Equations (1) and (2), respectively) for all artificial and field soil samples were computed and are listed in Table 4. The diffusion coefficients (D_e) from treated samples

Table 4. Mean effective diffusion coefficients (D_e) and Leachability indices (LX)

Sample	Mean D_e (cm ² /sec)	Mean LX
K15L0	6.16E-06*	6.04*
K30L0	6.07E-07	7.73
K15L10	1.02E-08	8.07
K30L10	3.62E-10	9.57
TL0	4.71E-08	8.66
TL10	1.03E-10	10.2

*Note: K15L0 sample disintegrated after 7 hours of testing

were significantly lower than those from untreated samples. Upon quicklime treatment, mean D_e values for K15L10 and K30L10 samples were 1.02×10^{-8} cm²/sec and 3.62×10^{-10} cm²/sec, respectively (Table 4). These diffusion coefficients fall in the general range of 10^{-5} cm²/sec (very mobile) to 10^{-15} cm²/sec (immobile).²⁹⁾ Therefore, it can be concluded that Pb mobility was significantly reduced upon quicklime treatment. More specifically, the K15L10 and K30L10 samples showed a D_e decrease of two and three orders of magnitude, compared to the K15L0 and K30L0 samples, respectively. The increased amount of kaolinite used also resulted in a reduction of D_e values due to its contribution to the adsorption capacity of the sample and the higher cementing action it provided. Specifically, the 30% kaolinite sample showed D_e values two orders of magnitude less than their 15% counterpart upon quicklime treatment. Similar results were obtained from the field soil samples. Since LX values from samples K30L10 and TL10 were higher than 9, these two samples can be considered acceptable for “controlled utilization”. The LX value of sample TL10 was higher than 9 due to the low pH conditions which resulted in low leachable Pb fraction. Therefore, effective immobilization was achieved even though very small amount of clay was present. The LX value of the K15L10 sample was approximately 8.1. This result indicates that even though the K15L10 sample was treated with quicklime, it is not suitable for “controlled utilization”. Therefore, the amount of clay is an important factor in the immobilization

of Pb using quicklime.

CONCLUSIONS

Pb leachability in artificial and field soil samples treated with quicklime was evaluated by performing semi-dynamic leaching tests. The specific conclusions pertaining to the results presented herein can be drawn as follows:

1. In untreated artificial soil samples, the amount of kaolinite appears to be an important factor affecting Pb leachability. This result indicated that sorption on kaolinite particles was the prevailing mechanism for Pb immobilization. However, the capacity of sorption onto kaolinite was limited due to low CEC and surface area.
2. The mobility of Pb was reduced significantly in the quicklime treated soils (reduced D_e and increased LX), thus allowing only trace levels to be released.
3. Upon quicklime treatment, the controlling leaching mechanism of Pb from artificial and field soil samples appeared to be diffusion. In untreated artificial and field soil samples, however, the controlling leaching mechanism was found to be wash-off.
4. The sample containing 30% kaolinite treated with 10% quicklime and the field soil sample treated with 10% quicklime can be considered acceptable for "controlled utilization".
5. In quicklime treated samples, the amount of clay also plays an important role in the success of the treatment. This result indicated that Pb immobilization can be significantly enhanced only in the presence of high clay content. Even though the quicklime treated field sample can be considered acceptable for "controlled utilization", this type of sample is not appropriate for quicklime treatment unless the clay content is high or the leachable fraction of Pb is low.

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