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In-Situ Lime Stabilization of Ponded CCRs – Results of an Ongoing Laboratory Feasibility Study

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ABSTRACT

In response to a number of high profile releases of coal combustion residues (CCRs) at surface impoundments over the last 6 years, EPA and states have embarked on rule making efforts designed to address CCRs management with an emphasis on risks associated with impoundments. With the final EPA rule on CCR management having just been published, electric utility companies are exploring and assessing management options to significantly minimize economic impacts and environmental risks associated with CCRs management. This paper presents results of an ongoing feasibility study which evaluates the practical and economical prospect of using in-situ lime stabilization of ponded CCRs. Specifically this paper presents results of a laboratory characterization of ponded CCRs samples collected from active sites in southeastern and Midwestern U.S.

Given that CCRs are pozzolanic in nature, the addition of lime triggers cementitious reactions that can potentially result in a material with more favorable geotechnical and chemical properties. Moreover, the hydration reaction of quicklime (CaO) to hydrated lime ($Ca(OH)_2$) in wet CCRs is a drying/dewatering process. This study examines the effects of quicklime addition on mechanical strength, compactibility, and heavy metal leachability of ponded CCRs. Basic geotechnical index tests including gradation, crushed and uncrushed specific gravities, Atterberg limits, and Eades and Grimm test were performed to characterize the samples. Compaction tests were conducted to obtain the optimum moisture contents and maximum dry densities of the ponded CCRs samples. Unconfined compressive strength tests were conducted on both treated and untreated ponded CCRs specimens. Initial results show a time-dependent increase in unconfined compressive strength consistent with ongoing cementitous reaction. In addition to the physical tests, the toxicity characteristic leaching procedure (TCLP), as described by EPA method 1311, was performed to assess the leaching potential of the untreated and quicklime treated ponded CCRs samples.

INTRODUCTION

Based on U.S. Energy Information Administration data (2015), coal remains one of the largest fuel sources of electricity generation in the U.S. accounting for approximately 35.8 % of the total net generation for electric utility. This large amount of coal utilization for electric generation produces huge quantity of coal combustion residues (CCRs) that requires handling and management. Of the over 114 million tons of total CCRs produced in 2013, only about 44.8 % is reused/recycled according the American Coal Ash Association (ACAA, 2015). A large portion of the remaining CCRs waste is managed wet through sluicing into surface impoundments/containment ponds for storage. In recent coal ash impoundment assessment study, the EPA reports approximately 676 such above ground impoundments containing wet CCRs from coal-burning steam electric power generating facilities in the U.S.

In response to a number of high profile releases of coal combustion residues (CCRs) at surface wet CCRs impoundments and similar management units over the last eight years, EPA and State Regulatory agencies have embarked on rule making efforts designed to address CCRs management with an emphasis on risks associated with impoundments. Apart from structural integrity concerns associated with each surface impoundment or similar management unit, several exceedances of state groundwater standards in down gradient wells have been reported for several unlined ash ponds. While the EPA rules have not yet been published, coal-burning electric utility companies are proactively exploring and assessing management options to significantly minimize economic impacts and environmental risks associated with CCRs management. Common closure or management options frequently proposed include: capping of ash basins with compacted clay liner or geosynthetic/geomembrane liner, dewatering, excavation and removal of CCRs from basins to a lined facility, and/or construction of alternative caps that would control long-term releases of constituents of concern while meeting site structural integrity standard.

Except for alternative cap/encapsulation, all the other closure or management options are technically challenging, rift of safety concerns and may not be economical. For example, in a pilot field testing study, Delaney et al. (1985) described using a vacuum wellpoint dewatering system to improve the condition of the wet CCRs facility where dewatering was carried out to the extent that the CCRs could be excavated and hauled out for disposal. In this case, dewatering the ash served to minimize risk associated to handling and transporting saturated, unstable ash which was difficult to excavate, to transport and place in a disposal site. This pilot field study, although successful, provides limited data which impedes extrapolation to predict the overall performance of similar dewatering schemes applied at a larger scale and for different project-specific conditions or to develop and propose generalized recommended procedures to apply this method to other ash ponds. This paper presents an alternative management approach involving in-situ lime stabilization of ponded CCRs materials which could be used in combination with other strategies. This ongoing study evaluates the practical and economical prospect of in-situ lime stabilization of ponded CCRs through laboratory characterization of ponded CCRs samples collected from active sites in southeastern and Midwestern US. The goal of this research is to demonstrate the structural stability and environmental benefits of in-situ treatment of wet or ponded with quicklime. The specific objectives of study is to demonstrate the impact of quicklime in the drying/dewatering process of wet or pond CCRs through hydration reaction to hydrated lime ($Ca(OH)_2$), to evaluate the effects of quicklime addition on mechanical strength, hydraulic conductivity, compactibility, and heavy metal leachability of ponded CCRs.

BACKGROUND OF LIME STABILIZATION

The use of lime to strengthen weak soils has been successfully employed for many decades (Vorobieff and Murphy, 2003, Castel and Arulanandan, 1979, Little, 1999, Rogers and Glendinning, 2000, Thompson, 1964). Considerable research and successful case histories have been reported on the improvement of density, stiffness, and bearing capacity of weak ground conditions using different ground improvement techniques including in-situ lime stabilization by various techniques such as dry or wet soil mixing.

The application of in-situ lime stabilization towards improving engineering properties of CCRs materials is relatively new and only a few lab-based studies have been reported in the literature. Most of these studies have not involved ponded fly ash. One of the first studies involving lime stabilization of CCRs was presented by Gosh and Subbarao (2001). This study presented evidence of microstructural developments at the particle level that explained the observed engineering improvement of strength and reduction of permeability of lime-treated CCRs. Gosh and Subbarao (2007) and Chand and Subbarao (2007) provided laboratory experimental evidence documenting considerable shear strength gains measured in a Class F CCRs treated with lime. Gosh and Subbarao (2007) reported unconfined compressive strength increases of 134 and 225 % after a 28 day curing period at 30 ºC for lime contents of 4 and 6 %, respectively. The unconfined compressive strength values were reported to continue to increase with curing time. For a 90 day curing period, this study reported an increase of the unconfined compressive strength with respect to the control samples with no lime addition of 598 and 1720 % for 4 and 6 % lime contents, respectively. Chand and Subbarao (2007) tracked strength and durability values for a pond ash treated with lime contents of 10 and 14 % and curing periods that extended up to 180 days. These levels of lime content yielded unconfined compressive strengths higher than 1.2 MPa after 28 days of curing, requiring the use of test methods commonly used in rock mechanics. For example, the unconfined compressive strength values obtained for ponded CCRs samples treated with 10 and 14 % lime after 28 days of curing were 38 and 41.5 times higher than the average value obtained from untreated pond ash samples. Zhou et al. (2002) presented results of an experimental feasibility study conducted to assess lime for improvement of weak ash ground. The lime contents considered in this study were

5, 10, 15, 20, and 25 % by dry weight of CCRs. The curing conditions of the specimens involved using a box at a relative humidity of about 90 % and a temperature of about 24 ºC. This study showed similar trends as the ones mentioned above. In general, UCS increased with curing time and with lime content. However, these authors found that when the lime content is 10 % or higher the strength gains are substantially higher.

EXPERIMENTAL METHOD

Materials

CCRs samples were obtained from two separate confidential active CCRs basins located in the Southeastern and Midwestern US, hereon referred to as sites 1 and 2 samples. Bulk wet samples were collected from approximately 3 – 5 feet of the pond surface at several different locations (up to 10 sampling locations). Though ponded CCRs from each site were sampled at different locations of the pond, elemental composition analysis using X-Ray Fluorescence (XRF) and particle size distribution showed that elemental composition and particle size were similar at each sampling location. Owing to the large quantity of sample materials required for the experimental plan and the equivalent characteristics of the samples, all bulk samples from each pond were mixed together and homogenized in the lab using a mortar mixer to form the sample material used for this study. Lime used in study is the fine size quicklime (CaO > 90 %, MgO < 5 %, $CaCO_3$ < 3 %, and SiO_2 < 2 %) supplied by Lhoist North America.

Material Characterization

(a) Elemental composition of CCRs

Elemental compositions of the two CCRs samples used in this study were performed using XRF for elemental compositions (Table 1) at the Lhoist material lab. The materials consist of high amounts of oxides of silicon and aluminum with low amounts of calcium oxide.

(b) Mineralogical characterization of the CCRs

The mineralogy of the CCRs samples was determined using X-ray diffraction (XRD). Though the diffractograms of the samples are not presented in this paper, Table 2 summarizes the identified mineral phases and the fraction of these phases in the CCRs samples. The two main crystalline phases identified in the CCRs samples are quartz (SiO_2) and mullite ($Al_6Si_2O_{13}$). These two mineral phases are among the principal minerals found in coal fly ash (Rattanasak and Chindaprasirt, 2009). Magnetite/Iron Oxide (Fe_3O_4), hematite (Fe_2O_3), Aluminum Silicate Hydroxide ($Al_3Si2O_7(OH)_3$) and Corundum (Al_2O_3), are the other mineral phases present in the CCRs samples.

Table 1: Chemical composition of the CCRs samples (mass %)

	Site 1 CCRs Sample	**Site 2 CCRs Sample**
CaO	0.57	0.92
MgO	0.836	0.617
SiO2	53.8	47.5
TiO2	1.25	1.11
Al2O3	25.9	23.5
MnO	0.0299	0.0237
Fe2O3	5.98	8.24
P2O5	0.144	0.188
Na2O	0.281	0.293
K2O	3.36	2.30
LOI 600 °C (1000 °C)	9.08 (9.31)	14.9 (15.3)

Table 2: Mineral phases, chemical formula and fractions of the CCRs samples

Compound Name	**Chemical Formula**	**Quantity (%)**	
		Site 1	**Site 2**
Quartz	SiO_2	19.5	32.8
Mullite, syn	$Al_6Si_2O_{13}$	30.1	25.4
Iron Oxide	Fe_3O_4	5.5	7.6
Hematite, syn	Fe_2O_3	6.9	4.8
Aluminum Silicate Hydroxide	$Al_3Si_2O_7(OH)_3$	28.1	21
Corundum, syn	Al_2O_3	9.9	8.4

(c) Particle size distribution (PSD)

Laser diffraction particle size analyzer was used to determine the PSD of CCRs samples from sites 1 and 2 (Figure 1). The instrument uses the principle of light scattering to determine the particle size distribution of sample in powder form.

From Figure 1, the CCRs sites 1 and 2 have particles that range in size from 0.4 μm to 300 μm and 0.6 μm to 300 μm, respectively.

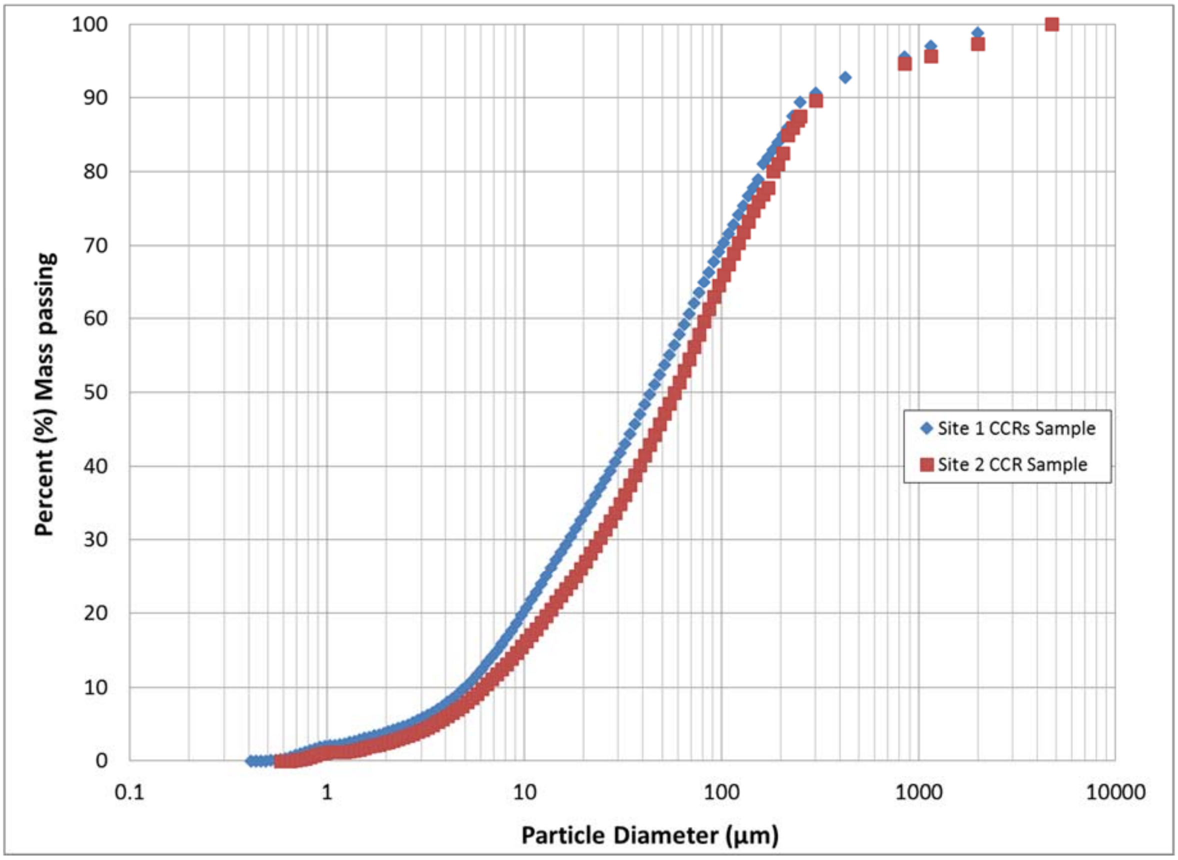

Figure 1: Particle size distribution of Site 1 and 2 CCRs samples

(d) Geotechnical index properties and characterization

Specific gravities of the CCRs samples were determined using standard test methods by water pycnometer in accordance with the ASTM D854-14. Compaction tests were performed in the laboratory on oven dried samples using standard energy effort to establish the maximum dry densities and optimum moisture contents of the CCRs (ASTM D698-12). Compaction tests were performed using two types of molds (standard Proctor mold of 0.0333 ft^3 (943.0 cm^3) and mini Harvard mold of 0.0024 ft^3 (67.96 cm^3)). Oven dried samples were thoroughly mixed at a predetermined molding DI-water contents and covered to mellow for approximately 24 hours prior to performing compaction tests. The mellowed samples were placed into the molds in 3 layers, subjecting the CCR material to a total compactive effort of approximately 112400 ft-lbf/ft^3 (600 kN-m/m^3). The number of hammer blows, hammer drop height, and the hammer mass to achieve the compactive effort are summarized in Table 3. The plot of the dry density – moisture content relationships obtained from compaction tests are presented in Figures 2 and 3. The physical and geotechnical index properties of the CCRs samples are summarized in Table 4.

Table 3: Summary table of compaction test features and characteristics

Compaction Features and Components		Mold Type	
		Standard Proctor	**Mini Harvard**
Mold Dimension	Diameter (in)	4.	1.31
	Height (in)	4	2.81
	Volume (ft^3)	1/30	1/454
Hammer	Drop Height of Hammer (in)	12	4
	Hammer Weight (lb)	5.5	2.324
Compaction	# of layers	3	3
	# of blows/layer	25	12
	Total Energy (ft-lbf/ft^3)	12,400	12,661

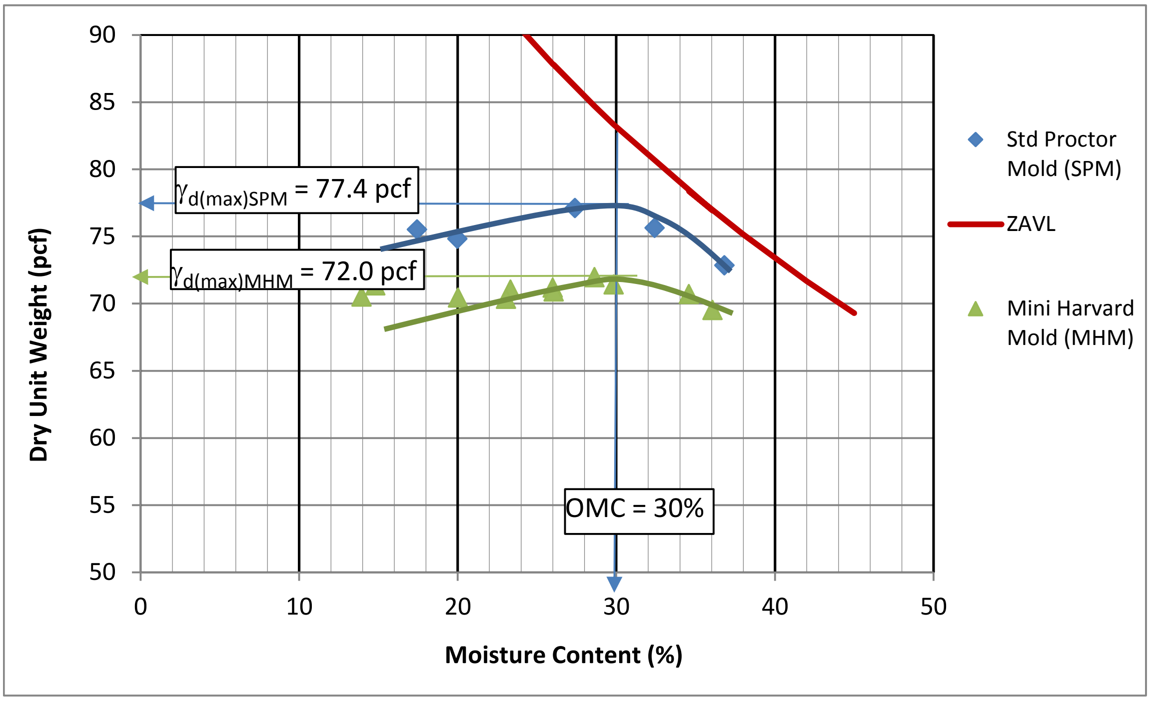

Figure 2: Density – Moisture Content Relationship of Site 1 CCRs Sample

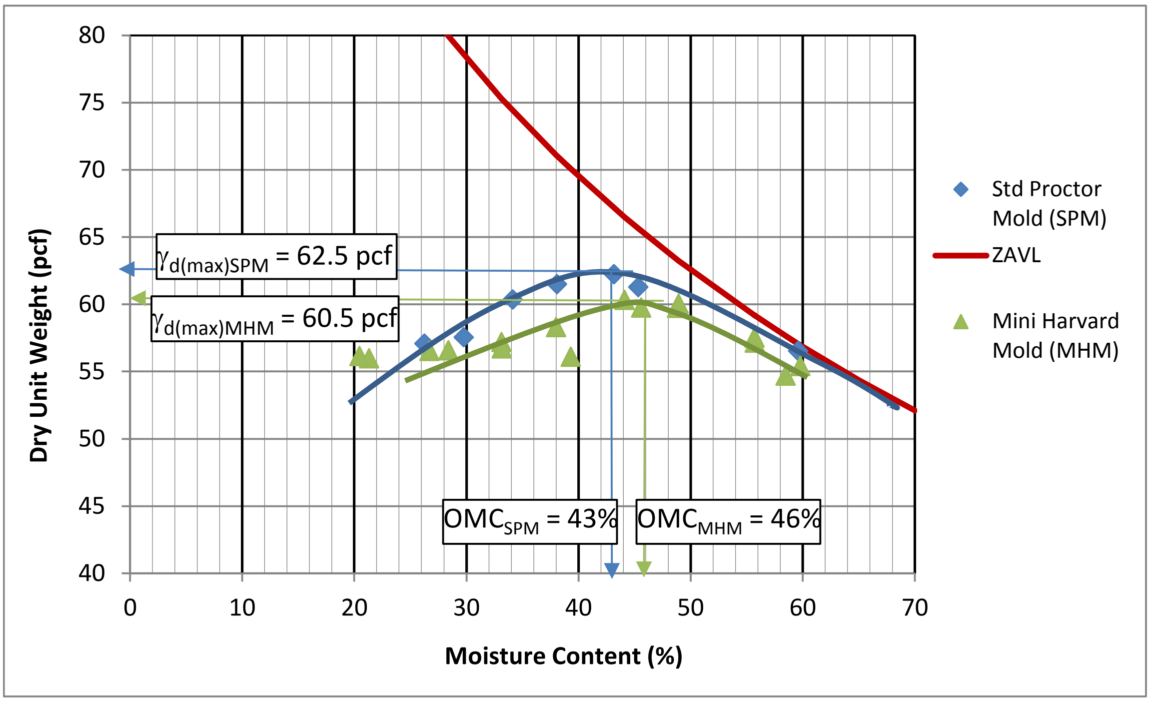

Figure 3: Density – Moisture Content Relationship of Site 2 CCRs Sample

Table 4: Summary of Physical and Geotechnical Index properties of CCRs samples

	Site 1	**Site 2**
C_c	0.84	1.13
C_u	13.2	14.2
D60 (μm)	64.9	86.5
D50 (μm)	43.4	57.8
D30 (μm)	16.3	24.4
D10 (μm)	4.9	6.1
Gs	2.22	2.01
Max Dry Density (pcf)	72.0	60.5
Optimum Moisture Content (%)	30	46

(e) Leachability characteristics

The Toxicity Characteristics Leaching Procedure (TCLP) test (EPA Method 1311) was performed to assess the mobility of inorganic elements from the CCRs samples. For the test, dried granular samples of the materials were leached for 18 hours in an acidic solution of pH 2.88 or 4.93 (prepared with acetic acid). The leachates were filtered (using 0.45 filter paper) and analyzed in ICP at the Lhoist materials lab in Irving, TX for the following TCLP cations (Ag, As, Ba, Cd, Cr, Pb and Se) (Table 5).

Table 5: ICP analysis of the leachate of the TCLP tests performed on the untreated CCRs samples

Sample Type	Elements (mg/L)						
	Ag	As	Ba	Cd	Cr	Pb	Se
Site 1, untreated	<0.01	0.106	1.689	<0.01	0.049	0.913	0.365
Site 2, untreated	<0.01	0.158	1.404	<0.01	0.062	0.884	0.407
RCRA Regulatory Levels	5.0	5.0	100	1.0	5.0	5.0	1.0

Determination of Minimum Lime Content for Stabilization

The minimum lime content to initiate stabilization was determined in accordance with the ASTM 6276-99a (Eades and Grimm's pH test) for soil stabilization. The goal of this test was to determine the minimum lime content to achieve a pH of 12.4. Lime content in the study varied between 1 to 6 % by mass of dry CCRs. 0 % and 100 % lime contents were also conducted as control. Low lime content (Hydrated lime or quicklime) at approximately 2 % is sufficient to achieve a pH of 12.4, the minimum content required for stabilization.

Selecting Effective Lime Content for this Stabilization of Pond CCRs

Several factors influence the determination of the effective quick lime content to stabilize ponded CCRs in-situ. Key factors include:

1. Sufficient drying of wet CCRs without resulting to expensive dewatering and achieving a range of moisture contents that permit compaction of treated CCRs to achieve acceptable dry density.
2. Adequate strength gain to provide effective stabilization/solidification and significant permeability reduction to minimize infiltration through the treated ponded CCRs.
3. Satisfactory encapsulation without negatively affecting the release potential of constituent of concerns.

The following experiments were designed to investigate the potential impacts of quicklime treatment of ponded CCRs samples on the factors highlighted above.

a. Assessing drying potential of wet CCRs with quick lime

In assessing quicklime drying potential of wet CCRs, different contributing factors are postulated. During the hydration reaction of quicklime (CaO) to hydrated lime ($Ca(OH)_2$)

some quantity of water from wet CCRs is utilized during the process. The amount of water involved in this reaction process can be determined based on stoichiometry. Further hydration and pozzolanic reactions can result in further utilization of some amount of available water. However, significant moisture loss to evaporation will occur during the mellowing period if treated CCRs are unsealed. Drying or moisture loss to evaporation is highly dependent on climatic conditions and thickness of the treated layer.

In order to evaluate the feasibility of using quicklime to dry wet CCRs, samples of known moisture contents from each site (Site 1 sample at 45 % and Site 2 sample at 62 %) was mixed with dry quick lime at x different dosages: 1, 2, 3, 4, 4.5, 5 and 6 % (these dosages represent the ratio of dry weight of quick lime over the dry weight of fly CCRs) and left to mellow sealed and unsealed. The sealed samples were designed to assess the required water to hydrate quicklime to hydrated lime, which can be estimated following stoichiometry. The unsealed samples stimulate additional moisture loss due to evaporation and other climatic factors within the lab environment. Mellowed unsealed samples were remixed, and sampled for moisture content determination. The data from drying potential test of quicklime are presented in Figures 4 and 5 for wet sites 1 and 2 samples, respectively.

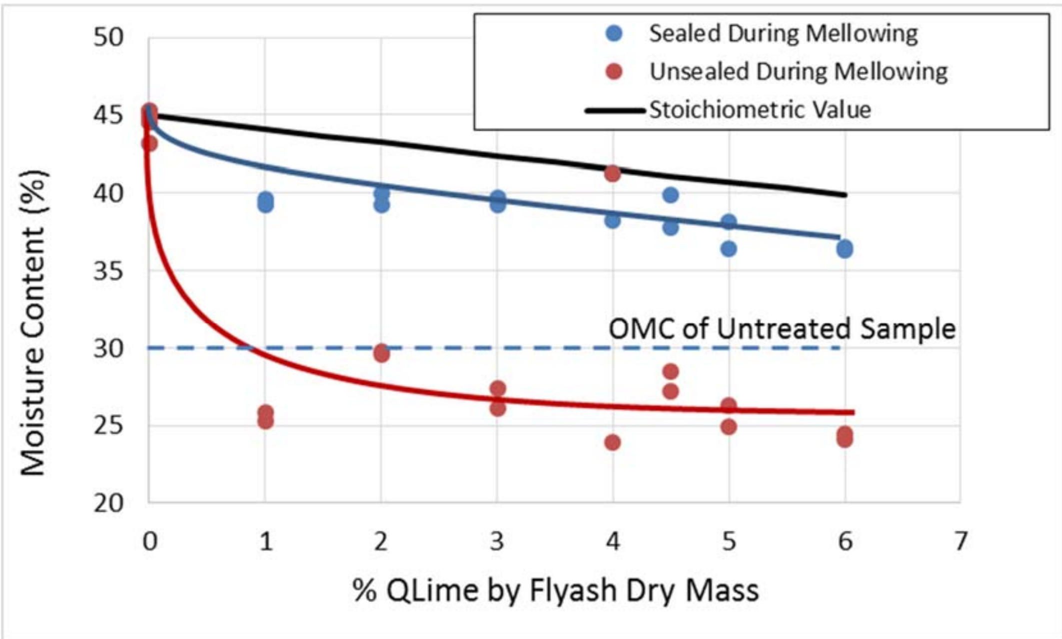

Figure 4: Drying Potential of Quicklime Treatment of wet Site 1 CCRs Samples

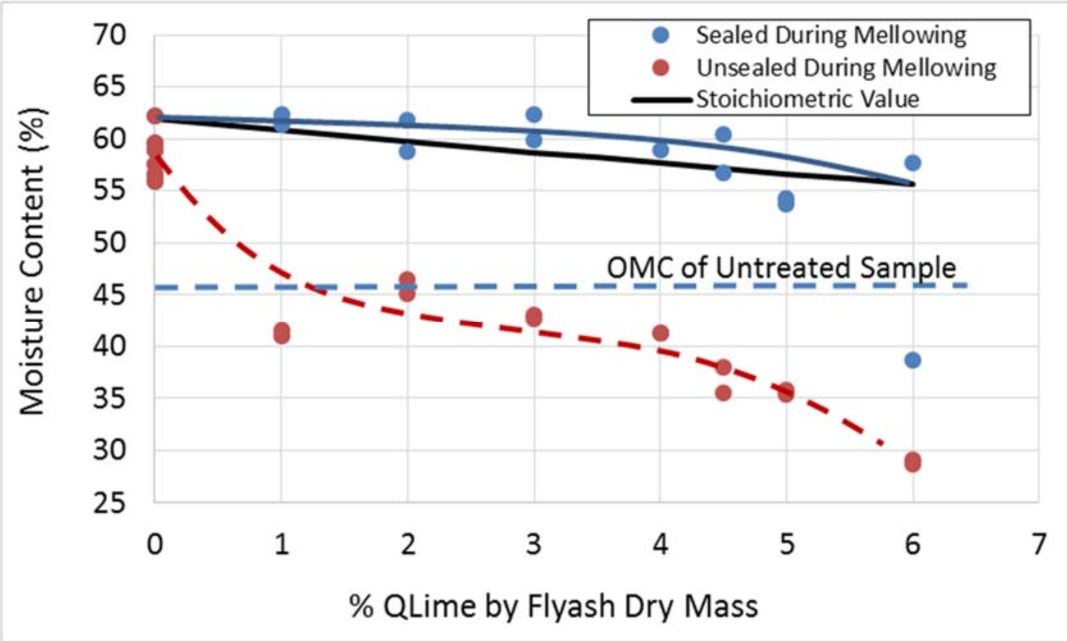

Figure 5: Drying Potential of Quicklime Treatment of wet Site 2 CCRs Samples

b. Unconfined compression tests on wet CCRs treated with different quicklime contents

Triplicate samples of compacted CCRs from sites 1 and 2 were treated and mellowed under unsealed conditions to assess the drying potential of ponded CCRs. Samples were compacted in three layers in a Mini Harvard mold at compactive effort of 12,661 ft-lbf/ft^3 (613 kN-m/m^3). The compacted CCRs samples of sites 1 and 2 were cured at 40 ºC for 2 days in an oven. The samples were kept in sealed Ziploc® bags containing moisture sponges to provide humidity and prevent drying of samples during hydration. After accelerated curing, the samples were conditioned to room temperature for 2 hours in an environmental chamber set to 23 ºC. Samples were tested in general accordance with ASTM D2166 (ASTM 2006). The test matrix of the test results discussed in this paper are summarized in Table 5 below. All unconfined compression tests were strain controlled carried out at a strain rate of 0.5 %/min. The peak stress of the stress-strain curve was taken as the unconfined compressive strength (UCS).

RESULTS

Selection of an Optimal Content of Quick Lime

Based on initial considerations, economics and practical details, an initial set of unconfined compressive tests was carried out to help determine an adequate quick lime content for both CCRs materials. The range of quick lime contents evaluated were 1, 2, 3, 4, 4.5, 5, and 6 % by dry mass of CCRs. The comparison of unconfined compressive strengths for the different quick lime contents was done from samples tested after a 2 day curing period at 40 °C (i.e. accelerated curing). Both pond CCRs sites showed an increasing unconfined compressive strength values with increasing quick lime content for the range of quick lime contents considered. The average trends of increase of UCS as a function of quick lime content for the pond CCRs of Sites 1 and 2 are shown in Figure 6. This figure shows the average trend lines and presents the associated linear regression equation. Based on these results, a 5% quick lime content was selected for the additional unconfined compression, permeability and leachability laboratory tests for both sites.

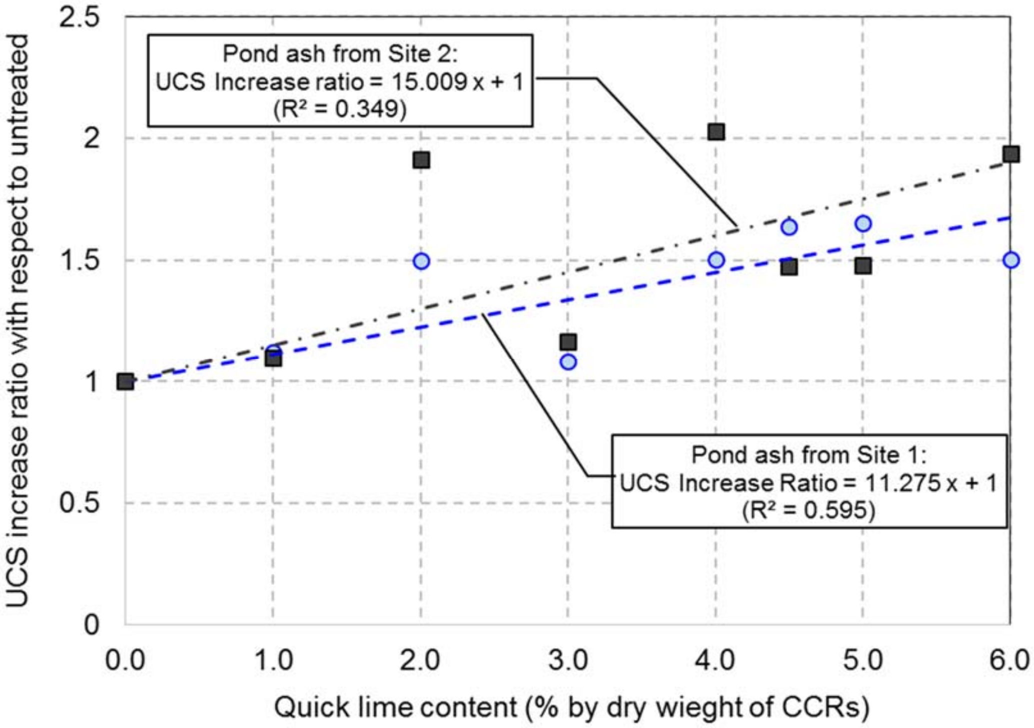

Figure 6: Unconfined compressive strength of 2-day - 40 °C cured compacted pond CCRs versus quick lime content

Effect of Curing Conditions and Curing time on UCS for Pond Samples Treated with 5 % Quick Lime Content

Each wet ponded CCRs was treated with quicklime ratio of 5 % and mixed using a mechanical mixer at a controlled speed of about 111 rpm and for a time period of 2 minutes. The mixed samples were allowed to mellow unsealed for 24 hours and remix. After thoroughly remixing each pond CCRs-lime mixture was compacted into the mini Harvard mold, using a total compactive effort of 12,661 ft-lbf/ft^3. A series of unconfined compression tests were carried on untreated and lime-treated compacted pond CCRs samples to assess strength variation as a function of lime content, curing conditions and curing time. Unconfined compressive strength of lime stabilized geomaterials (for example soils) cured at room temperature of 23 ± 2 °C (73 ± 4 °F) in a moist chamber for either 7, 28 or 90 days are typically used for construction quality control (ASTM D5102-09). However, accelerated curing at elevated temperature of 40 °C (105 °F) or higher but not to exceed 49 °C (120°F) is also commonly performed when it is expedient to simulate long-term field conditions. For this study, triplicate samples of both pond CCRs samples treated with 5 % quicklime content were cured at normal room temperature of 23 ± 2 °C for 2, 7, 14, 28 and 90 days and at accelerated temperature of 40 °C at 2, 4, 6 and 8 days as per ASTM D5102-09 standard.

As expected, both CCRs samples treated at 5 % lime show increasing UCS values with curing time for both room and elevated temperature curing conditions. Samples treated at 5 % lime content and cured for 2 days at accelerated and normal room temperatures possess the same UCS values for each CCRs site. It appears that up to 2 days of curing may not be sufficient to cause accelerated CCRs–lime pozzolanic reactions at elevated temperature of 40 °C for the CCRs tested in this study. Curing at an elevated temperature of 40 °C for a period of 8 days caused UCS values to grow from about 1.1 times at 2 days to 7.1 times at 8 days than the samples cured at room temperature for site 1. Similarly for site 2, the strength increased from about 1.3 fold at 2 days to about 5.9 fold at 8 days curing than same samples cured at room temperature. In Figures 7 and 8, the strength increase as a function of curing period for samples cured at elevated temperature appears to follow an exponential function.

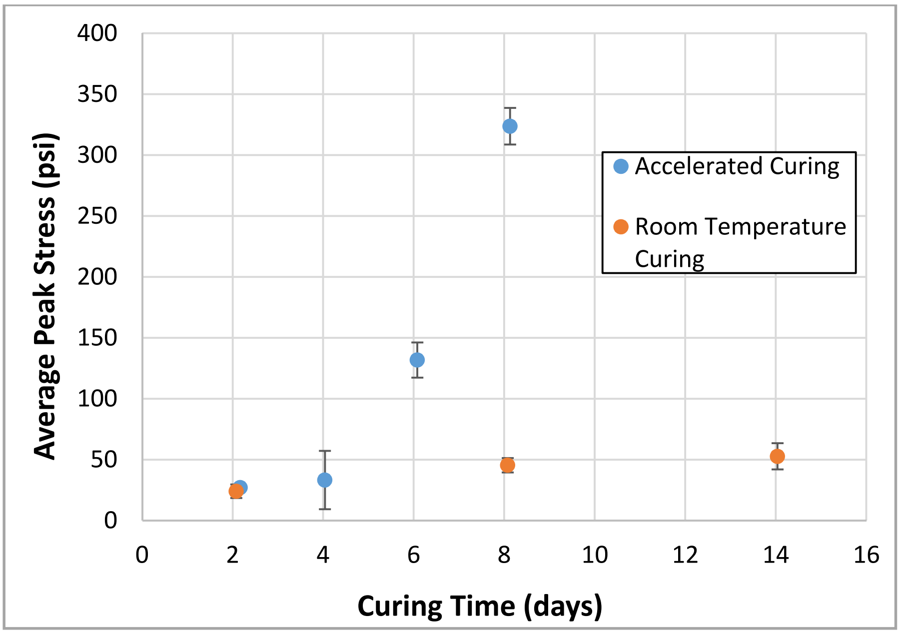

Figure 7: UCS of Site 1 Sample with 5 % Quick Lime Content

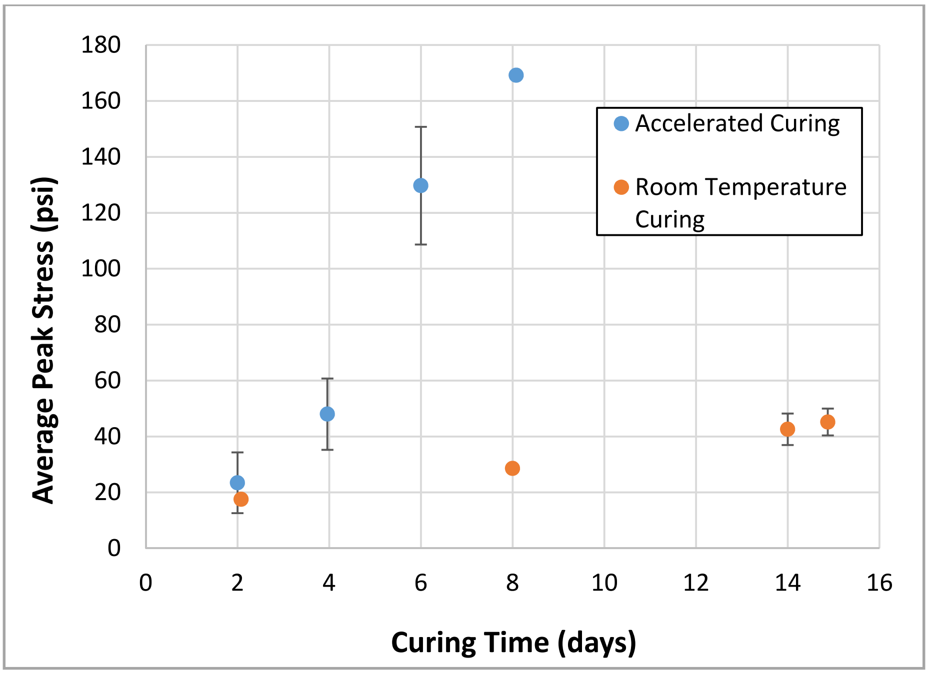

Figure 8: UCS of Site 2 Sample with 5 % Quick Lime Content

This behavior was also observed by Rao and Asha (2012), however, the authors concluded that the compressive strength of lime-fly CCRs is controlled uniquely by the mass of the reacted lime for a given curing condition and that at elevated temperatures larger amounts of lime were reacted.

Impact of 5 % Quicklime treatment on leachability of CCRs samples

TCLP leaching test was performed on crushed UCS samples of 5 % quick lime treated CCRs for both sites. The leaching test was performed as per EPA Method 1311 for the samples cured under different conditions and time durations. Table 6 presents summary results of ICP analysis performed on leached sample cured at elevated and normal room temperatures for a curing duration of 2 days. No significant leached values difference exists for each sites between the curing conditions for the curing duration of 2 days. The released concentrations of all the elements analyzed are significantly below the regulatory levels of the Toxicity Characteristics Leaching Procedure (TCLP) and slight reduction in amount leached with strength gain/pozzolanic reaction is observed for some elements (Figures 9-11).

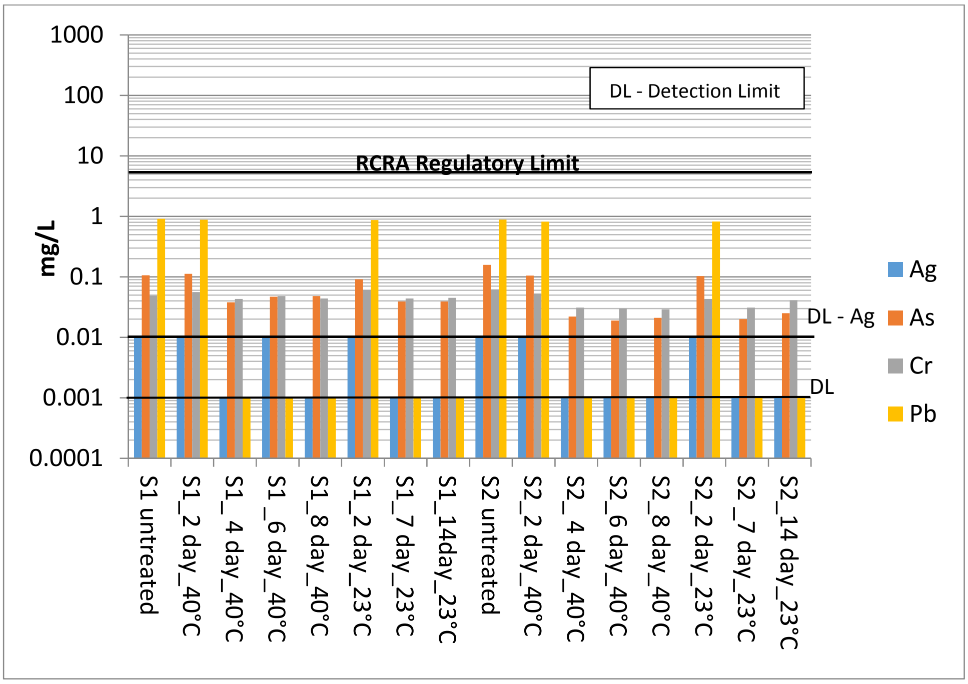

Figure 9: ICP analysis of the leachate of TCLP tests performed on site 1 and 2 CCRs samples treated with 5 % quicklime; As, Cr, Pb, Ag

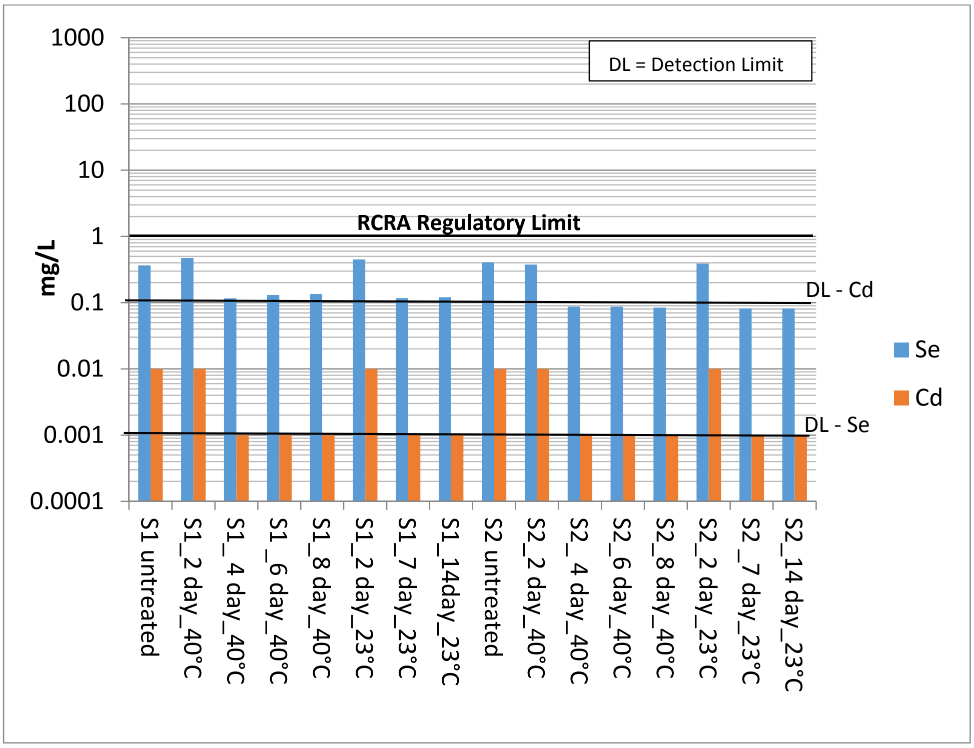

Figure 10: ICP analysis of the leachate of TCLP tests performed on site 1 and 2 CCRs samples treated with 5 % quicklime; Se, Cd

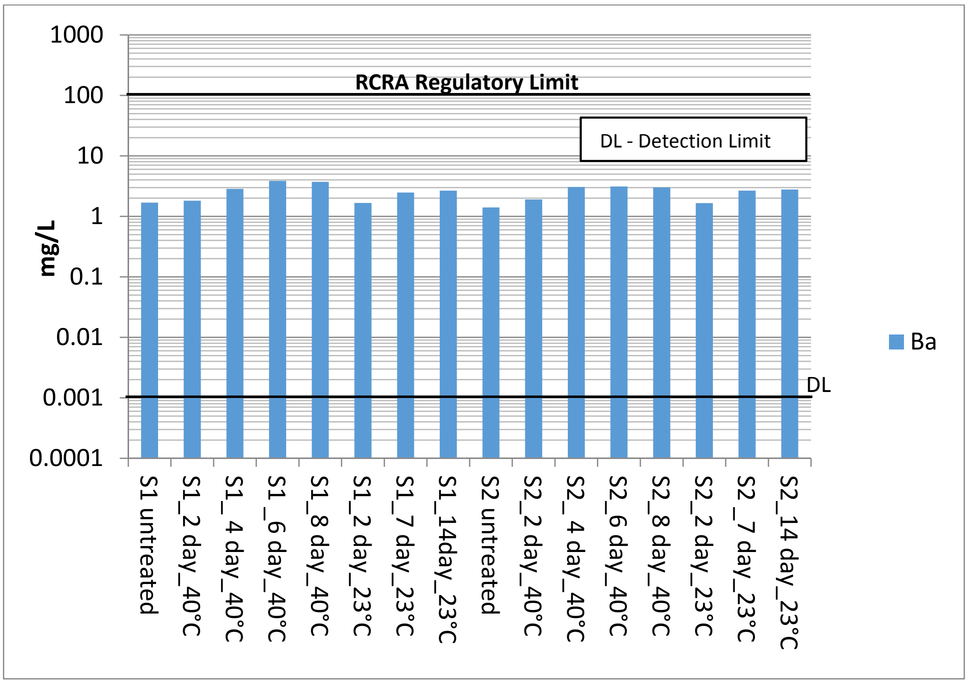

Figure 11: ICP analysis of the leachate of TCLP tests performed on site 1 and 2 CCRs samples treated with 5 % quicklime; Ba

SUMMARY AND CONCLUSIONS:

Engineering properties of ponded CCRs samples stabilized with lime (1 to 6 %) were investigated through laboratory experiments. The experimental study included characterization of two ponded CCRs, compaction tests for standard Proctor energy using two molds, chemical analyses, unconfined compression tests, TCLP leachability tests, and assessment of the hydraulic conductivity of treated and untreated ponded CCRs using flex wall permeability tests and 1-D consolidation tests. The ongoing study aim is to evaluate the practical and economic prospects of in-situ lime stabilization of ponded CCRs materials in wet storage facilities. The results to date show improvements in strength that suggest it could be a cost-effective methodology to achieve structural integrity requirements. Furthermore, the leachability and permeability test results also suggest this methodology meets environmental benefit requirements. Quick lime treatment at 5 % ratio of dry mass of wet ponded CCRs does not adversely impact on the release potential of TCLP heavy metals (Ag, As, Ba, Cd, Cr, Pb and Se). The released concentrations were significantly lower than the RCRA regulatory levels for the untreated and treated CCRs. Results from this exploratory and laboratory study suggests that lime treatment of CCRs improves geotechnical properties of the material while not adversely impacting, and in some cases reducing the release of some of the metals from these materials. Given the promise of this bench scale testing, future field-relevant pilot or demonstration study is recommended for in-situ evaluation and verification of current results. Compare to other alternative ponded CCRs closure/management options, in-situ lime stabilization of ponded CCRs is predicted to be less expensive, more economical and safe option.

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