# RECLAMATION OF ABANDONED COAL MINE WASTES USING LIME CAKE BYPRODUCTS IN KOREA[1]

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**Abstract.** In Korea, hundreds of abandoned and closed coal and metallic mines are present in the steep mountain valleys due to the depression of the mining industry since the late 1980s. From these mines, enormous amounts of coal waste were dumped on the slopes, which causes sedimentation and acid mine drainage (AMD) to be discharged directly into streams causing detrimental effects on soil and water environments. A limestone slurry by-product (lime cake) is produced from the Solvay process in manufacturing soda ash. It has very fine particles, low hydraulic conductivities ($10^{-8}$ ~ $10^{-9}$ cm/sec), high pH, high EC due to the presence of CaO, MgO and $CaCl_2$ as major components, and traces of heavy metals. Due to these properties, it has potential to be used as a neutralizer for acid-producing materials. A field plot experiment was used to test the application of lime cake for reclaiming coal wastes. Each plot was 20 x 5 m (L x W) in size on a 56% slope. Treatments included a control (waste only), calcite ($CaCO_3$), and lime cake. The lime requirement (LR) for the coal waste to pH 7.0 was determined and treatments consisted of adding 100%, 50%, and 25% of the LR. The lime cake and calcite were also applied in either a layer between the coal waste and topsoil or mixed into the topsoil and coal waste. Each plot was hydroseeded with grasses and planted with trees. In each plot, surface runoff and subsurface water were collected. The lime cake treatments increased the pH of coal waste from 3.5 to 6, and neutralized the pH of the runoff and leachate of the coal waste from 4.3 to 6.7. Surface cover of seeded species was significantly increased with lime cake and the 25% LR plots were sufficient to neutralize the acidity in the coal waste.

Additional Key Words: abandoned mine land, acid mine drainage, runoff, revegetation, water quality

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## Introduction

In Korea, over 300 coal mines have been closed or abandoned due to the depression of the mining industry since the late 1980s (Coal Industry Promotion Board, 2000). Many of them are located in steep mountain valleys and the enormous amounts of coal mine waste have been abandoned on slopes and acid mine drainage (AMD) from either portals or waste piles have been discharging directly to streams, causing detrimental effects on soil and water qualities. The environmental disruptions caused by the closed mines are very serious in Korea.

The wastewater from the portals of closed coal mines and the leachate from the waste piles are low in pH and contain high concentrations of Fe, Al, and Mn (Choi, et al., 1998; Kim et al., 1999; Park et al., 2001, 2002; Yang et al., 2002). Major environmental problems caused by the acid mine drainage and waste piles are the coatings of oxidized iron precipitates and aluminum precipitates on riverbeds and soils.

The Coal Industry Promotion Board (CIPB, 2000) in Korea has spent over $15 million dollars (US) annually to remediate the mine-related damages and to improve the environment. Most of the costs are directed to the wastewater treatment and forest restoration, such as to install passive AMD treatment systems (e.g., Successive Alkaline Producing System: SAPS) and revegetation of the forest. However, the investment has not been very effective due to the large number of sites, large amounts of AMD, and budget limitations.

A lime by-product (lime cake) is waste material produced from the Solvay process in manufacturing soda ash. In Korea, over 3 million tons of lime cake are stock piled. This material has been used in the past to reclaim disturbed lands, but due to recent concerns from environmental groups the lime cake has not been used in reclamation and remains in stockpiles with no plan for proper disposal. The lime cake has very fine particles, low hydraulic conductivities ($10^{-8}$ ~ $10^{-9}$ cm/sec), high pH and high EC due to the presence of CaO, MgO, $CaCl_2$ and NaCl as major components (Yang et al., 2002; Yang, 2003). Due to these physical and chemical properties, the lime cake has potential to be used as a neutralizer for acid-producing materials.

The objectives of this research were to measure the acid-neutralizing capacity of the lime cake, apply varying amounts of lime cake to coal waste plots by either layering or mixing the cake into the coal waste, collect and analyze runoff and leachate from treated coal waste piles, and access ground cover of seeded plants. If the lime cake application proves successful, this research will help in properly disposing of this material by reclaiming abandoned lands.

## **Materials and Methods**

Ten treatments were installed on a large, abandoned coal waste pile to test the application of the lime cake for reclamation of these piles. The slope of the coal waste site was 29.2° (56%). Each plot was 20 x 5 m (L x W) in size (Fig. 1) and was separated by plastic boundaries. Treatments include the control (coal waste alone), calcite ($CaCO_3$) as a reference, and lime cake (Table 1). The lime requirement (LR) for the coal waste to pH 7.0 was determined by titration to determine the amount of lime cake application. Lime cake treatments consisted of 25%, 50% and 100% of the LR (as $CaCO_3$).

The lime cake and calcite were either layered between the coal waste and topsoil or mixed with coal waste and topsoil. Each plot was hydroseeded with grasses and planted with trees. Surface coverage by grasses was determined by the computer image analysis.

In each plot, the flume and gutter were connected to a water reservoir to collect all the runoff and leachate from each plot (Fig. 2). Three pipes, 5 cm in diameter, were buried in each plot and connected to the reservoir to collect the leachate. Chemical properties such as pH and ion concentrations of the runoff and leachate were analyzed periodically. Efficiency of the lime cake in coal waste reclamation was assessed based on surface cover by plants, neutralization of runoff and leachate, and soil quality.

Table 1. Treatment design and revegetation on the coal waste pile.

| Plot Number | Treatments | Lime Treatment Methods | Vegetation† |
|---|---|---|---|
| 1 | Coal waste only | | Grass and trees |
| 2 | Coal waste + Lime cake (LR 100%) | Mixed | Grass and trees |
| 3 | Coal waste + $CaCO_3$ + topsoil | Layered | Grass and trees |
| 4 | Coal waste + $CaCO_3$ + topsoil | Mixed | Grass and trees |
| 5 | Coal waste + Lime cake (LR 100%) + topsoil | Layered | Grass and trees |
| 6 | Coal waste + Lime cake (LR 100%) + topsoil | Mixed | Grass and trees |
| 7 | Coal waste + Lime cake (LR 50%) + topsoil | Layered | Grass and trees |
| 8 | Coal waste + Lime cake (LR 50%) + topsoil | Mixed | Grass and trees |
| 9 | Coal waste + Lime cake (LR 25%) + topsoil | Layered | Grass and trees |
| 10 | Coal waste + Lime cake (LR 25%) + topsoil | Mixed | Grass and trees |

† Grasses: Orchard grass (*Dactylis glomerata* L), Kentucky Bluegrass (*Poa pratensis* L.) and Eulalia (*Miscanthus sinensis* Anderss); Trees: Pine tree (P*inus densiflora* S. et Z.), White birch (*Betula platyphylla* var. *japonica*) and Alder tree (A*lnus firma* S. et Z).

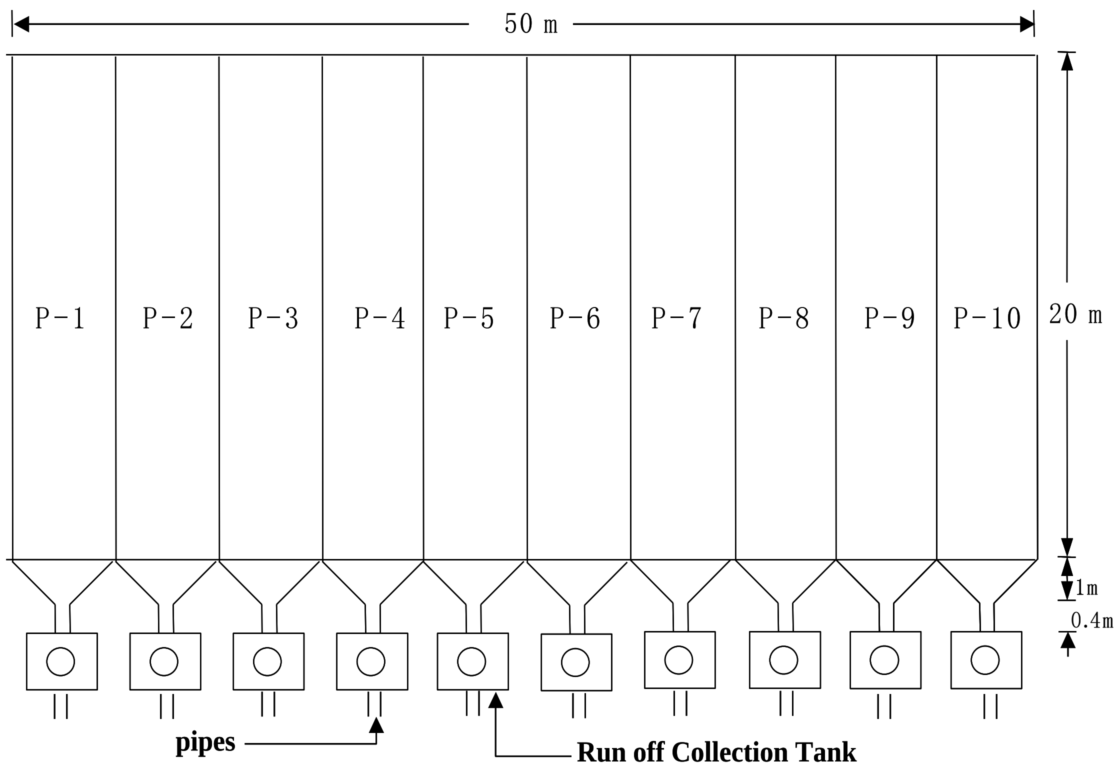


Figure 1. Layout of the coal waste plots with runoff and leachate collection reservoirs.

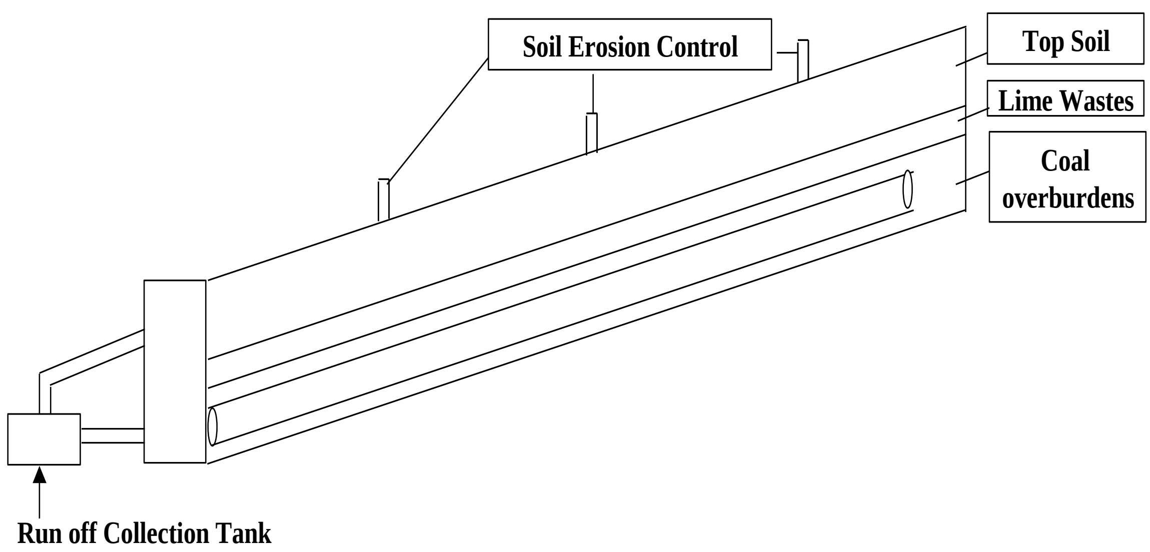


Figure 2. Side view of the experimental coal waste plots.

## Results and Discussion

Chemical Properties of the Coal Waste, Lime Cake and Topsoil

The pH of coal waste was 3.5 and about 17 Mg of $CaCO_3$ per ha were needed to adjust the pH to 7.0. The lime cake was high in bases such as Ca, Mg and Na with a high pH (11.2) and high electrical conductivity (EC: 19.6 dS $m^{-1}$). The topsoil was obtained from the road cut and was low in fertility.

Table 2. Chemical characteristics of the lime cake, coal wastes, and topsoil.

| Sample | pH (1:5) | EC (1:5) | OM | $P_2O_5$ | LR | Exchangeable | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | | | | | Ca | Mg | K | Na |
| | | dS $m^{-1}$ | g $kg^{-1}$ | mg $kg^{-1}$ | Mg $ha^{-1}$ | ----- $cmol_c$ $kg^{-1}$ ----- | | | |
| Lime Cake | 11.24 | 79.55 | 8.3 | 7.92 | - | 233.8 | 50.54 | 2.29 | 77.92 |
| Coal Waste | 3.50 | 0.23 | 16.5 | 9.13 | 16.5 | 3.9 | 0.29 | 0.06 | 0.07 |
| Topsoil | 6.51 | 0.03 | 0.8 | 15.75 | 0.37 | 4.5 | 0.54 | 0.06 | 0.12 |

OM : Organic matter based on the loss on ignition (LOI)

LR : Lime Requirement (as $CaCO_3$)

Table 3 shows the elemental composition of the coal waste and lime cake analyzed by the X-Ray Fluorescence (XRF) method. The coal waste was composed mainly of $SiO_2$, $Al_2O_3$, $Fe_2O_3$ and $K_2O$. These four elements comprised 66.5%. Major components of the lime cake were CaO, MgO, $Na_2O$, $SiO_2$ and Cl, of which composed 66%. X-ray diffraction analysis indicates that major minerals in lime cake were calcite, quartz and plagioclase. The particle size analysis indicated that percentages of clay, silt and sand were 8.6, 53.1 and 38.3, respectively. The toxic heavy metal contents in lime cake and coal wastes were very low based on the 0.1M HCl extraction method (data not shown). Based on the chemical properties as shown in Tables 2 and 3, the lime cake seemed to have potential to neutralize the acidic coal waste.

Effects of Lime Cake on pH of Coal Waste

Fig. 3 shows the pH changes at each treatment plot. The pH of the control (coal waste only) was 3.5 but increased to 7.5 when mixed with lime cake without the topsoil (plot 2). However pHs of the plots treated with $CaCO_3$ and lime cake either layered or mixed with the lime requirement of 25%, 50% and 100% (plot 3 to 10) were stabilized at about 6.0 irrespective of the amounts of lime cake. This might be due to the pH buffer capacity of the topsoil. The neutralizing effects of lime cake were equivalent to the calcite. This result indicated that coal waste had less buffering capacity for pH than the topsoil. Thus, the combined treatment of lime cake with topsoil was suggested to neutralize the acidic coal waste.

Table 3. The elemental compositions of the lime cake and coal waste.

| Element Composition (%) | Lime Cake | Coal Wastes |
|---|---|---|
| $Na_2O$ | 2.30 | 0.14 |
| MgO | 11.45 | 0.34 |
| CaO | 39.29 | 0.22 |
| $SiO_2$ | 2.83 | 37.11 |
| $P_2O_5$ | 0.04 | 0.15 |
| $K_2O$ | 0.15 | 3.47 |
| Cl | 10.19 | ND |
| $Fe_2O_3$ | 1.03 | 4.76 |
| $Al_2O_3$ | 1.31 | 21.15 |
| $Cr_2O_3$ | ND | ND |
| MnO | ND | ND |
| SrO | 0.04 | 0.01 |
| Water Content and Loss on Ignition (LOI) | 31.07 | 23.35 |

Effects of Lime Cake on Chemical Properties of Runoff and Leachate

Fig. 4 shows the pH of the runoff and leachate collected in the tanks at the bottom of experimental plots. Data were averaged over measurements from April to August. The runoff pH of the coal waste was 4.3 but increased significantly to the range of 6.7 to 7.1 with treatments of calcite and lime cake. There were no significant differences in runoff pH among treatments of calcite and lime cake, layered or mixed, or amounts of lime cake. This might be due to the combined effects of lime cake and buffering capacity of the topsoil.

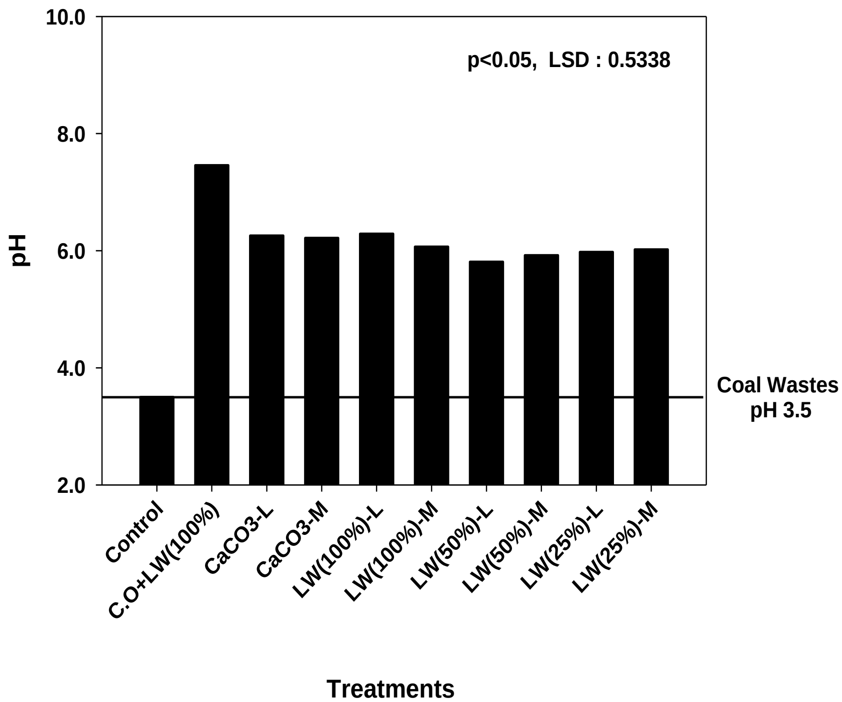


Figure 3. Soil pH in each treatment plot (Control, Coal Waste Only; C.O, Coal Waste; LW, Lime Cake; L, Layered; and M, Mixed)

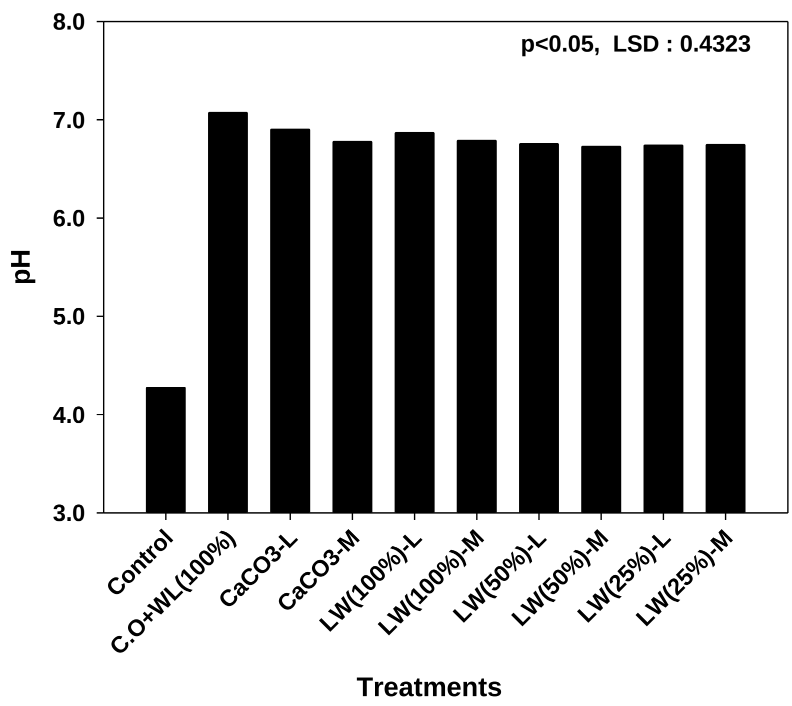


Figure 4. Runoff and leachate pH of each treatment plot (Control, Coal Waste Only; C.O, Coal Waste; LW, Lime Cake; L, Layered; and M, Mixed)

Fig. 5 shows sulfate ion concentrations in the runoff and leachate at each treatment plot. The sulfate concentration in the runoff was highest in the control but decreased significantly with the lime cake treatment. When the coal wastes were treated in combinations of lime cake and topsoil, the sulfate concentrations in the runoff were slightly decreased. The sulfate ion in runoff from abandoned piles and mines in Korea has been shown to be up to several thousand ppm and was known to increase the acidity of the AMD (Choi et al., 1998; Kim et al., 1999). The decreases in sulfate ion with lime cake treatments might play a positive role in alleviating the environmental impacts of the coal wastes.

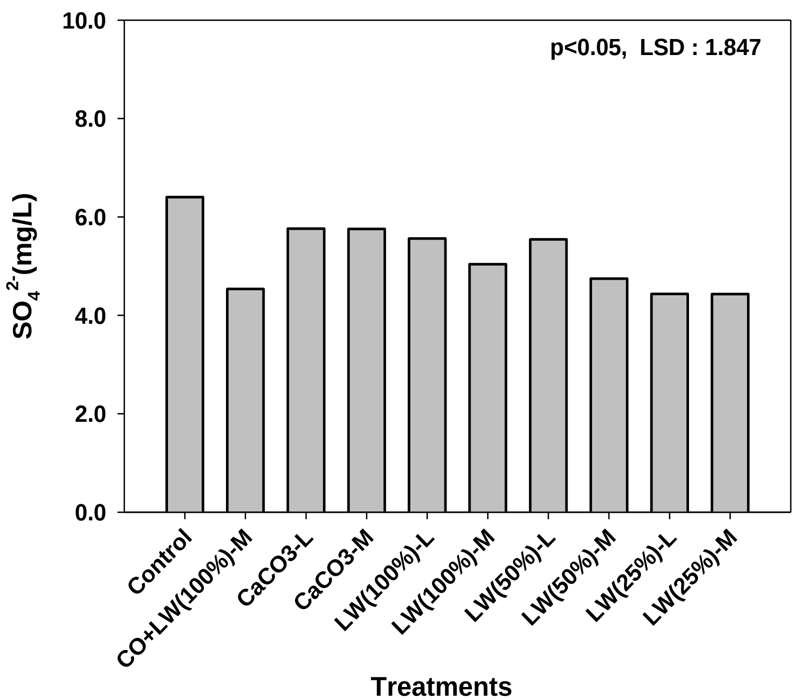


Figure 5. Sulfate ion concentrations in runoff and leachate in each treatment plot (Control, Coal Waste Only; C.O, Coal Waste; LW, Lime Cake; L, Layered; and M, Mixed).

Fig. 6 represents the Al and Fe concentrations in the runoff and leachate from plots as a function of date and precipitation. Al and Fe from the coal wastes are the major ions deteriorating the water quality in abandoned coal mine areas in Korea (Choi et al., 1998; Kim et al., 1999). The initial Al concentrations in the runoff were in the ranges of 30 to 60 mg/L, but those were sharply decreased with time. There were no significant interactions among treatment plots, dates and precipitation on the Al concentrations in the runoff. However, concentrations of Fe in the runoff fluctuated with date and precipitation, but showed a tendency to decrease its concentration with lime cake treatments. In the experimental site, the yellowboy symptom due to the oxidized Fe coatings on soils and rock fragments were evident prior to the lime cake treatment, but this phenomenon disappeared with treatment. This might be related to the decreases in Fe concentrations in the runoff and leachate from the coal wastes. The Fe concentrations in the runoff seemed to increase with the amount of precipitation.

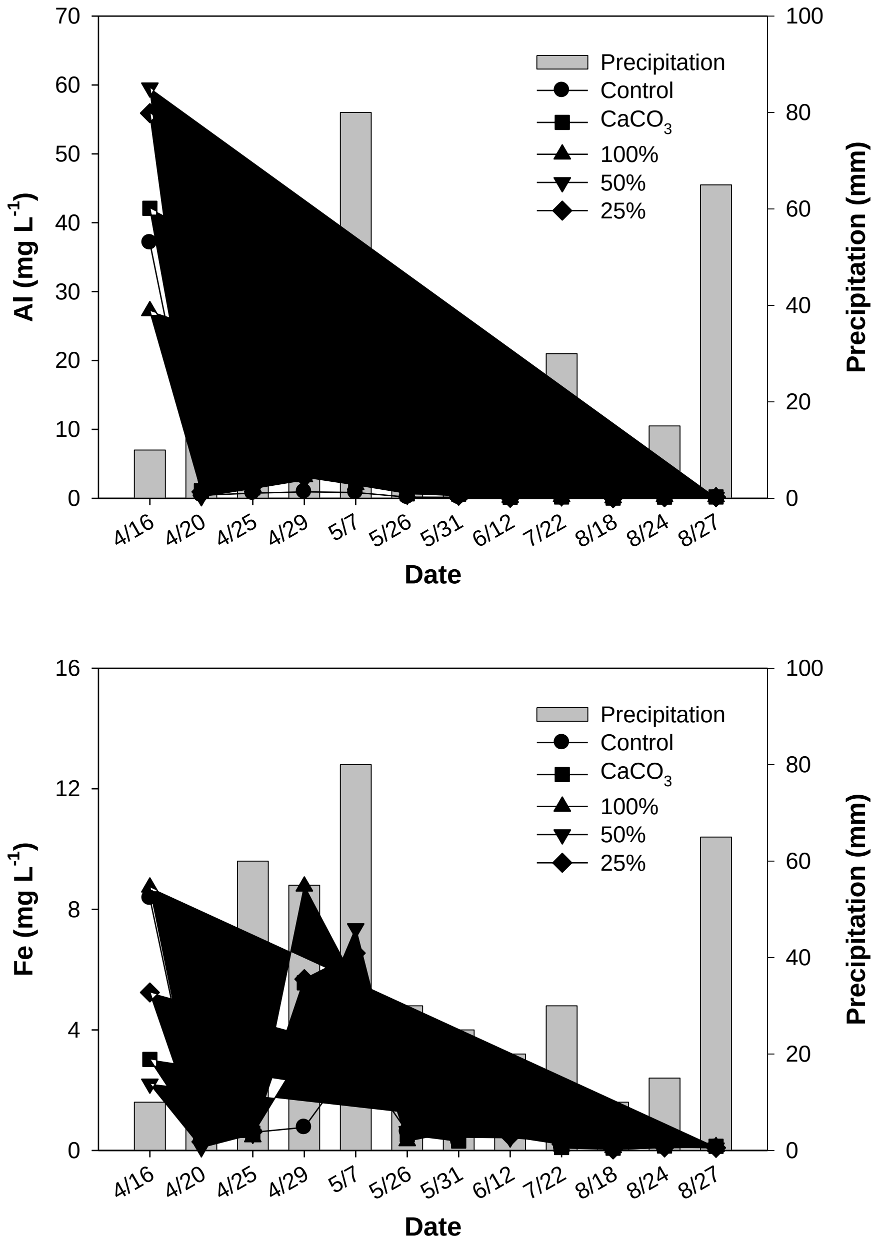


Figure 6. Al and Fe concentrations in runoff and leachate as affected by lime cake treatment and precipitation.

Effects of Lime Cake on Revegetation of the Coal Wastes

Table 4 shows the percentage of the grass cover and the surface coverage ($cm^2$) in each treatment plot during June to August. Due to the temperature in Korea, grasses such as orchard grass (*Dactylis glomerata* L), Kentucky bluegrass (*Poa pratensis* L.) and Eulalia (*Miscanthus sinensis* Anderss) were hydroseeded at the end of May. The grasses covered only 15.5 % of the coal waste plot at the early stage but the cover increased with time to 33% in August. Growth of grasses was enhanced with the combined treatments of lime cake and topsoil (plots 5 to 10), which resulted in increased surface cover by grasses. The increase in surface cover from June to August was higher with lime cake treatments. The cover percentages and surface coverage were highest when the lime cake was applied at 25 % of the lime requirement. This might be related to the high salt content of the lime cake. Bioassay tests in the greenhouse revealed that seed germination of these grasses was highest when lime cake was applied at 25 % of the lime requirement (LR) but germination was significantly suppressed at the 50% and 100% treatments. The results suggest that high salts content of the lime cake might be a limiting factor in revegetation of coal waste.

Table 4. Vegetation cover percentage and surface coverage ($cm^2$) at each treatment plot during June to August.

| Month | | Treatment Plots* | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 |
| June | %# | 15.5 | 13.2 | 14.5 | 14.6 | 15.8 | 25.6 | 22.4 | 30.3 | 25.5 | 25.6 |
| | $cm^{2\#}$ | 310.9 | 246.9 | 256.0 | 259.0 | 287.7 | 509.3 | 471.6 | 537.9 | 494.0 | 510.8 |
| July | % | 25.9 | 23.2 | 26.1 | 22.8 | 21.0 | 30.9 | 29.3 | 37.9 | 36.3 | 37.2 |
| | $cm^2$ | 531.9 | 529.6 | 553.7 | 537.0 | 477.7 | 604.8 | 575.2 | 758.9 | 739.6 | 752.9 |
| August | % | 33.4 | 27.5 | 46.3 | 45.7 | 40.4 | 37.5 | 36.9 | 45.6 | 52.6 | 61.2 |
| | $cm^2$ | 680.3 | 547.4 | 920.1 | 913.8 | 797.0 | 783.2 | 760.1 | 908.2 | 1016.3 | 1175.8 |

*Refer to Table 1 for the treatment combination.

#Percentages and surface coverage were averaged over 5 measurements.

## Summary

Field plots were used to test the effects of the lime cake on the reclamation of coal wastes by examining the chemical qualities soil and water (runoff and leachate) and surface cover of grasses. Lime cake treatments increased the pH of the coal waste from 3.5 to 6, and raised the pH of runoff and leachate from coal waste from 4.3 to 6.7. Concentrations of sulfate, Al and Fe in the runoff and leachate were significantly decreased with lime cake. Surface cover by orchard grass, Kentucky bluegrass, and Eulalia on coal waste was significantly increased with the lime cake. The amount of lime cake at 25% of the lime requirement was sufficient to neutralize the acidic coal waste and allowing germination of grasses. Either layering the lime cake between the coal waste and topsoil or mixing with coal waste could be adopted as reclamation methods. The combined treatment of lime cake and topsoil is recommended for revegetation in the coal waste piles. Results demonstrated that the lime cake from soda ash production has good potential for reclaiming abandoned coal waste piles and to alleviate the environmental problems associated poor soil and water quality from coal wastes.

## Acknowledgements

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