

Mitigation of Rainfall Erosion in Coal Mine Overburden via Microbial Calcification

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Abstract: In spite of renewable sources, many countries still mostly depend on coal for electricity generation. It accompanies massive dumping of coal mine overburden further accumulated and formed large heap during open cast coal mining. These coal mine overburden occupy not only huge land but also create many environmental challenges. Water and soil pollution due to leachate during rainfall and air pollution due to dust during erosion are two major environmental challenges confronted in daily basis. The present study focuses to evaluate the feasibility of enzyme induced calcite precipitation to mitigate water/rainfall induced erosion of coal mine overburden. For this, the jack bean urease was augmented with two different concentration ratios of urea and calcium chloride dihydrate viz 1:1 and 1:1.25 by spraying method. After 10 days of curing, the rainfall erosion test concluded that the erosion rate decreased significantly in the biotreated sample due to precipitation of calcium carbonate biomineral. Also, an increase in the strength was observed in the biotreated sample via pocket penetrometer. Further, the microscopic characterization proves the carbonate biomineral precipitation and its linkage formation inside the sample pore which is the reason of erosion resistance and strength gain.

Keywords: Coal mine overburden; rainfall erosion; enzyme induced calcite precipitation; Jack bean urease; biomineral

1. Introduction

Coal mine overburden is a major threat to coal workers and nearby regions. The study of atmospheric contaminations associated with coal overburden dust has become the primary focus of geo- environmental studies (Gabarrón et al., 2017; Khorshid and Thiele-Bruhn, 2016; Mirzaei Aminiyan et al., 2017). The main cause of dust emission in coal mining practices and processing are drilling, blasting, loading and unloading, and transportation (Baur 2011; Fabiano et al. 2014). Poor air circulation due to dust emission leads to low visibility and exposure to workers for a longer period and higher intensity at mining sites. Exposure for a longer period and higher intensity to dust environment lead to severe health and other ecological/environmental problems (Lashgari and Kecojevic 2016; Li et al. 2016; Lin et al.2017; Mandal et al. 2012). Studies have proved that prolonged exposure to excessive amounts of respiratory coal overburden dust can lead to fatal lung diseases, namely Pneumoconiosis, chronicobstructive pulmonary disease, tuberculosis, chronic bronchitis, emphysema, and other lung diseases. These lung diseases are generally common in coal mine workers, leading to higher mortality rates. Defining health as a state of complete physical, mental, and social well-being of an individual, and not

merely the absence of diseases. Currently, dust is considered the world's largest single environmental health risk. Globally, premature deaths and morbidity occur maximum due to ambient and indoor air pollution (Zhang et al., 2016; Lim et al., 2012). Most health problems in coal mining regions are caused by site dust and water erosion. The land, bodies of water, air, and environment become contaminated from dust generation by blasting, excavation, and mine waste dumping. The concentration of particulate matter is higher in mining areas, and respiratory diseases are more common in mining communities (Saha et al., 2011). One of the most hazardous occupations in the world is mining, both in terms of short-term injuries and fatalities and long-term human health problems such as cancer and respiratory conditions (Stephens and Ahern,2001). The study on coal mining has proved that people living in coal mining communities are more prone to cancer, increased risk of developing heart and lung diseases, hypertension, and kidney diseases, and mortality rates are also higher in nearby mining regions (Hendryx and Ahern, 2008). Furthermore, people in these mining regions consume water mixed with chemical waste and debris from mining activities. Apart from chemical hazards, high temperatures and humidity may cause heat-related illness; long-term exposure of mineworkers to heat stress renders them less productive (Maurya et al.,2015). Coal dust plays a significant role in climate change and other atmospheric processes. Coal dust is one of the primary sources of potentially harmful elements contaminated in the air (Rout et al., 2015; Sun et al., 2014). Agriculture and water contamination are severely harmed by wind erosion. It has a variety of effects on agriculture. It might lower the nutritional value and yield of crop plants. It badly affects plants by reducing results or degrading agricultural products' quality (Spash, 1997). The resulting economic loss from dust and erosion has become an issue of political and scientific concern worldwide. Coal dust associated with potential harmful elements like gaseous sulfur, nitrogen compounds, photochemical oxidants, ozone, and SPM is more damaging to agriculture. Wind erosion reduces the growth and economic yield of a wide range of major crop species in North America and Europe (Holland et al., 2006).

Metals present in the coal mine overburden get to contaminate the soil, and the erosion brings to the nearby water bodies, which may reach the human body either through agricultural products or in drinking water (Rulkens et al., 1998; Sponza and Koraoglu, 2002; Haroon et al., 2010). During the rainy season, heavy metal leaching occurs and contaminates the groundwater. According to Hamdam et al. (2016), a chemical dust suppressant, a sprinkle of water, synthetic polymers, and biopolymers applied to the soil surface are among the traditional ways of managing dust that has been successful. The sprinkling of water has less effectiveness in arid and semi-arid regions due to the evaporation rate of water. Using chemical suppressants for dust stabilization negatively affects vegetation, groundwater, equipment, and vehicles in mining areas. Synthetic polymers and biopolymers are highly productive and longer lasting than water and chemicals suppressants but tend to be very expensive. Recently, researchers realized that Enzyme induced calcite precipitation is much more environmentally friendly and has become the center of focus.

Hamdam et al. (2016), reported that enzyme-induced calcite precipitation (EICP) is a novel technique in civil and geoenvironmental engineering. In EICP, free urease enzymes are used as urea hydrolysis catalysts. The product calcium carbonate can reduce soil porosity and act as a bridge to connect soil particles. Neupane et al. (2015), have studied that various combinations of different urea, calcium chloride dihydrate, and urease can produce the optimum amount of calcium carbonate precipitation. EICP has outstanding performance in controlling surface erosion for building up soil strength, as observed by Putra et al. (2016) and Sun et al. (2020). A large volume of overburdened soil generates considerable dust, washed away by rainfall from slopes, that further causes dust pollution.

The purpose of present study is to determine the effectiveness of enzyme induced calcite precipitation (EICP) as a mean of fugitive dust control technique. In a laboratory experiment, model dust slope were prepared, and rainfall was simulated to evaluate the treatment effect. Therefore, EICP is used to control dust slope erosion, and rain is simulated to evaluate the treatment effect in coal mine overburdened soil.

2. Materials and Methods

2.1 Materials and Characterization

The coal mine overburden soil utilized in this work was sampled using a disturbed sampling method below a depth of about 1.5 m at Katras area, BCCL. The physical characteristics related to grain size distribution curve is shown in Fig1. and the specific gravity of coal mine overburden soil is 2.37. The chemical characteristics of coal mine overburden by XRF analysis shows the elements that contains C is 15%, Ca is 0.13%, Mg is 0.5% and the ph is 7.2. The EICP solutions employed for this study are made up of jack bean urease, urea, and calcium chloride dihydrate. Sumner et al. (2013) introduced urease, an enzyme extracted from jack beans (*Canavalia ensiformis*) that catalyzed the hydrolysis of urea to form ammonia and carbon dioxide. Equimolar concentration of urea-calcium chloride dihydrate (0.5M and 0.625M) with 4.8 g/l urease were used in both the treatment process.





Fig. 1. Grain Size Distribution Curve

2.2 Methods

Batch study preparation

The preferred concentration of the constituent chemicals producing high precipitation ratio and high precipitation efficiency was determined by conducting forty-eight batch precipitation tests. The preferred concentration in terms of precipitation ratio is defined as the ratio of measured precipitation mass ($M_{\text{Experimental Mass}}$) to the maximum theoretical mass precipitate ($M_{\text{Theoretical mass}}$) of EICP solution.

Precipitation ratio (%) =
$$\frac{M_{\text{Experimental Mass}}}{M_{\text{IM}}}$$
 Theoretica mass × 100 (1)

$$M_{\text{Theoretical mass}} = C \times V \times M \tag{2}$$

 $M_{\text{Experimental Mass}}$ = Mass of the precipitated materials CaCO₃ evaluated from the tests in grams.

 $M_{\text{Theoretica mass}}$ = Theoretical Mass of CaCO₃ in grams.

- C= Concentration of equimolar concentration of reactant in moles/litre.
- *V*= Volume of solution in litre.
- M= Molar mass of calcium carbonate (CaCO₃) = 100.087 g/mole

Tests were conducted in 50 ml Falcon tubes containing 30 ml EICP solution without any soil. Based upon the previous research in EICP solution, there were two sets, i.e., the equimolar concentration of urea and calcium chloride dihydrate and the non-equimolar concentration of urea and calcium chloride dihydrate

The urease enzyme was dissolved into deionized water first for 15min and filtered using filter papers to remove the undissolved particles of urease. The filtered urease was put in falcon tubes, and then urea and calcium chloride dihydrate were added to prepare EICP. The falcon tubes were kept aside for 30° Celsius in an incubator shaker at 150 rpm for seven days. Simultaneously measure Ph for initial and seven days of incubation. The precipitated calcium carbonates settled at the bottom of the falcon tubes within a few hours. After seven days, the solution is filtered through filter paper. Drying the amount of precipitation deposited on the filter paper and in the tubes was measured after one day of drying at 100 degrees Celsius. Combining the dried filter paper with the precipitation present in the falcon tube allowed us to calculate the total amount of calcium carbonate precipitation. Each of these tests is conducted twice to check the reproducibility.

Table 1. represents the batch precipitation test we selected out of 48 falcon tube tests, i.e., the equimolar concentration of urea-calcium chloride dihydrate (0.5 M) and non-equimolar concentration of urea-calcium chloride dihydrate (0.5M) with 4.8 g/l urease for the treatment process.

Equimolar/Non-	Urea	Calcium chloride	Urease	Precipitation Ratio	Ph
Equimolar		dihydrate			
concentration					
1:1	0.5M	0.5M	4.8g/l	77.9	7.57
1:1.25	0.5M	0.625M	4.8g/l	93.7	7.35

Table 1. Batch Precipitation Test

Rainfall erosion test

The rainfall model consisted of a metal frame that functioned as the support for the model slope, a mild steel container with a length of 16.7 cm, a width of 15.8 cm, and a height of 10.98 cm—fixing the slope angle at 35°. Rainfall is simulated by a rainfall device containing a sprinkler, a water faucet, and a sprayer head. The sample size used in this study was small; all samples were prepared under identical conditions to compare the testing results. The sprayer head was an adjustable spray sprinkler, placed around 43cm above the model surface, providing a uniform rainfall covering the entire surface area of the model slope. Controlling the rainfall intensity at 30mm/hr, purposely selected to stimulate rainfall condition. The water used in simulating rainfall was tap water. The collection vessel collected dust soil washed out from the model slope. The video recorder continuously captured the surface erosion pattern of the model slope. The set up for rainfall erosion test used in the study is similar to Jiang et al. (2019).

Sample preparation and treatment methods

The samples for rainfall erosion test were prepared using a mild steel container size (18 x 15.6 x 5) cm³. First, the samples were oven dried for a day and subjected to minor compaction to attain field criteria. Subsequently, constant pore volume was added to a particular model, and water was sprayed uniformly over the entire area. The sample sprayed with tap water was considered as the control sample (S0). Further, the EICP solution was prepared by dissolving urease powder in tap water to a specific urease concentration of 4.8g/l and filtered through filter paper. Then, adding 0.5M equimolar concentration of urea-calcium chloridedihydrate is named S1. The non-equimolar concentration of urea-calcium chloridedihydrate has been discarded for the rainfall erosion test. The latter is discussed in microstructural analysis in results and discussion. After spraying, these trays were exposed to sunlight, and the weight of the samples was measured for seven days before being subjected to a rainfall erosion test.

2.2.4 Microstructural Analysis

To characterize the precipitated material for the baseline study, FESEM are done. Field Emission Scanning Electron Microscope (FESEM) represents the morphology investigation of precipitated CaCO₃crystal growth in urea-calcium chloride dihydrate solution from the test tube experiments. Energy dispersive spectroscopy (EDS) was used for elemental identification of the precipitate CaCO₃ from the test tube experiments. The samples were sputter-coated with platinum- palladium (Pt- Pd).

3. Results and Discussion

3.1 Results

Batch study results

Okwadha and Li (2010) reported that the increase in calcium carbonate precipitation is due to the production of ammonium ions in urea hydrolysis and the presence of calcium ions from calcium chloride dihydrate. With 4.8g/l urease, the equimolar concentration of urea-calcium chloride dihydrate shows 78% precipitation efficiency because the urea gets hydrolyzed through ureolysis to produce carbonate ion and ammonium ion to form calcium carbonate based on the amount of calcium in the treatment solution. This reaction also produces hydroxide ions which leads to an increase in alkalinity. Hamdam et al. (2016) studied that in equimolar concentration, the stoichiometric with urea produces excess ammonia while consuming approximately 97% of the available Ca^{2+} and 100% of urea. With 4.8g/l urease, the non-equimolar concentration of urea-calcium chloride dihydrate (0.5M-0.625M) shows the best precipitation i.e 93% in comparison to equimolar concentration. The non-equimolar concentration may increase or decrease ammonia (NH₃) release, affecting the chemical environment's alkalinity (pH). Hamdam et al. (2016) observed that a high urea concentration leads to increased alkalinity due to the abundance of NH₃ in the absence of Ca^{2+} ions. Thus, Ca^{2+} becomes the limiting reagent in the reaction. While comparing with less urea concentration which leads to a reduction in alkalinity with high calcium chloride dihydrate

concentration and there may be the availability of excess Ca^{2+} ion in the reaction with a limited amount of carbonate ion.

Rainfall erosion test

After treatment, the surface erosion pattern of model slopes differed significantly—the slopes under simulated rainfall for 2, 5, 10, 20 and 30 mins. With control treatment, slope S0 showed a collapsing phenomenon and observed surface erosion at the top of the slope at 5 min. A minor amount of dust was washed away after 10 mins. At 20 mins , a significant amount of soil flowed into the collection vessels from the model's slope's top. Less than half of the soil was still in the container after 30 mins.

In contrast to the treated slope S0, up to 10 mins, slope S1 did not develop any collapse characteristics, and the amount of eroded soil was low. However, after 20 mins, due to the soil erosion, the damage began to spread from the top of the slope. Interestingly, at the end of 30 mins of testing, damage in the soil sample did not show any significant changes see (Fig.1) represents the pictures of model slope under rainfall test.



(A. Control Sample S0 before rainfall)



(C. EICP Sample S1 before rainfall)



(B. Control Sample S0 after rainfall)



(D. EICP Sample S1 after rainfall)

Fig. 1. Picture of the sample under Rainfall Erosion Test

Microstructural Analysis

According to Ni et al. (2008), Calcium carbonate precipitation appears with different crystalline phases (polymorphs of vaterite, aragonite, and calcite in increasing order of thermodynamic stability). Rhombohedral shape for calcite is the most desired polymorph as it is thermodynamically stable. In contrast, the spherical shape for vaterite and needle-like shape for aragonite is metastable material that can quickly transform into a calcite state. The calcium carbonate precipitation with a particle size $\leq 425 \mu m$ was used to examine the polymorphs in the Scanning electron microscope. While increasing urease concentration, calcite precipitation will increase, which will highly influence CaCO3 morphology.Sondi et al. (2001) reported that urease concentration depends upon the different amino acid groups forming calcium carbonate. Through SEM images, calcite polymorph was visible for all the test tube samples. Results observed in the equimolar concentration of 0.5M urea and calcium chloride dihydrate with urease of 4.8g/l showed a mixture of rhombohedral and spherical-shaped crystal form, typical calcite morphology. The crystals exhibited smooth crystal faces with sharply defined edges. Still, most of the crystals had only partly developed smooth faces and incompletely formed edges and appeared as clusters of intergrown crystals. In a MICP process, ureolytic bacteria produce calcite spherulites, which Al-Thawadi and Cord-Ruwisch examined in 2012. They hypothesized that initially, spherical-shaped vaterite crystals developed, but they eventually broke down and changed into rhombohedral calcite (after9 hours), inheriting the initial vaterite spheres' spherical shape. The crystal shapes obtained from the SEM analysis is shown in (Fig. 2). In non-equimolar concentrations of 0.5M urea and 0.625 M calcium chloride dihydrate with urease of 4.8g/l showed vaterite formation. The amount of vaterite gradually declines with an increase in enzyme concentration, whereas the converse occurs for calcite. The formation of CaCO3 is dependent on the concentration of the enzyme (Li et al., 2013). Almajed et al.(2018) stated that rapid precipitation of CaCO₃ in EICP, compared to MICP, can be disadvantageous because it can sometimes result in the formation of unstable vaterite. For this reason, non-equimolar concentration has been discarded for the rainfall erosion test.



A. Equimolar concentration of 0.5 M urea – 0.5M calcium chloride dihydrate



B. Non equimolar concentration of 0.5M urea - 0.625M calcium chloride dihydrate

Fig.2. SEM images of CaCO3 crystals formed in Equimolar concentration and Non-Equimolar concentration

4. Conclusion

Laboratory experiments were conducted to demonstrate the feasibility of enzyme-induced calcite precipitation to improve rainfall erosion resistance. The optimum ratios differed with different

concentrations of reactants in the cementation solution. With an equimolar concentration i.e., 0.5M of urea and calcium chloride dihydatre showed better rainfall erosion resistance; moreover, the surface strength of slopes increased, unlike the control test. The presented results indicate a promising potential technique for dust control and leachate prevention.

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