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*The paper was published in the proceedings of the 11<sup>th</sup> Australia New Zealand Conference on Geomechanics and was edited by Prof. Guillermo Narsilio, Prof. Arul Arulrajah and Prof. Jayantha Kodikara. The conference was held in Melbourne, Australia, 15-18 July 2012.*

# Feasibility of using bentonite, lime and fly ash in permeable reactive barriers for acid sulphate soils

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## ABSTRACT

Acidic groundwater resulting from the poorly-planned use of acid sulfate soils has become a major environmental issue in coastal Australia. Use of permeable reactive barriers (PRBs) designed to generate alkalinity by promoting sulfate reduction has recently become popular as an alternative solution to this problem. Although different materials suitable for PRBs have recently been proposed and tested for remediation of acidic groundwater, there are still several aspects that need to be addressed. One of them is chemical precipitation and clogging of pore space that may significantly decrease the hydraulic conductivity of a PRB over a period of time. This study seeks to explore the feasibility of using bentonite in addition to fly ash and lime to form mixtures with a high buffer capacity and permeability that would enable groundwater flow through PRBs over a substantial period of time. To investigate the acid neutralization capacity of fly ash, lime, and bentonite mixtures, a series of laboratory tests were performed on different soil samples leached with solutions of sulphuric acid. It was found that the ability of mixtures to neutralize acidic fluids was mostly controlled by the content of lime. Laboratory data also indicated that an addition of bentonite to lime-fly ash mixtures may decrease the buffer capacity of soil. Results of hydraulic conductivity tests showed that the permeability of fly ash-lime specimens with bentonite may increase over a period of time, probably due to the changes in the diffuse double layer of bentonite particles.

*Keywords:* acid sulphate soil, permeable reactive barrier, buffer capacity, fly ash, bentonite

## 1 INTRODUCTION

Acid sulphate soils that contain iron sulfides (e.g. pyrite) pose a significant environmental, economic and social threat to coastal Australia. Although pyrite is relatively chemically inert when undisturbed and submerged under the watertable, it can result in the formation of sulphuric acid when exposed to oxygen. As the acid mobilizes large amounts of iron (Fe) and aluminium (Al) ions from the soil in the groundwater, it is an environmental hazard that will result in decreased agricultural and fishery productivity as well as cause damage to steel and concrete infrastructure (Golab et al 2006).

A few remediation techniques have been proposed and tested at different sites of coastal lowland in NSW, Australia (Indraratna et al 2002, 2006b). However, attention has recently been paid to the use of permeable reactive barriers (PRBs) due to several advantages such as no energy consumption, minimal operation and maintenance cost, and minimal disruption to the existing land (Indraratna et al 2010). A PRB can consist of a trench filled with reactive material and covered again with top soil to allow existing land use to continue (Golab et al 2006). When this barrier intersects the flow path of contaminated groundwater, the reactive material will increase the pH of the groundwater to a level that causes the iron and aluminium to precipitate out of solution. The reactive material is also expected to have a permeability that allows the groundwater to pass freely through the barrier.

A few studies have been performed to assess the capacity of different reactive material used in PRBs to neutralize acidity. Golab et al (2009) performed a number of laboratory tests to assess the suitability of fly ash, fresh recycled concrete, slag, lime and breccia in neutralizing acidity. Based on the obtained results, it was suggested that recycled concrete may be more preferable for a PRB compared to the lime and fly ash as the latter two typically have "excessively small grain size." Furthermore, Regmi et al. (2009) carried out a long-term (2 years) laboratory experiment to study the neutralizing capacity and permeability of recycled concrete while Indraratna et al (2010) tested the ability of recycled concrete to neutralize the acidity of groundwater in a large-scale field experiment. The results of these studies revealed that recycled concrete can successfully improve the pH of groundwater from acidic to mildly alkaline. However, the investigators noted that the armouring effect can occur in the PRB as a

result of precipitation of Fe and Al on the surface of reactive material, a process that can decrease the hydraulic conductivity and reduce the ability of PRB to neutralize acidity. Thus, there is still a need to find reactive material that can improve the long-term performance of PRBs.

Recent studies (Yeheyis et al 2010) related to mining waste management have shown that bentonite together with fly ash may be used to control the hydraulic conductivity of liners that contain mine wastes. It was also suggested that bentonite may improve the long-term performance of PRBs as the decrease in hydraulic conductivity of PRBs due to clogging can be offset by increases in pore space caused by reduction in the double layer thickness of bentonite particles. In addition, Kashir and Yanful (2001), who performed a series of hydraulic conductivity tests on bentonite permeated with acid mine drainage (AMD), reported that bentonite can retard metals such as Fe and Al, an advantage that can improve the overall performance of PRBs. However, it was also pointed out that bentonite did not seem to have a sufficient, long-term buffering capacity as the pH of the effluent liquid dropped to a low value of 3 after only 3 pore volumes of AMD permeation. It is clear that further research is needed to clarify the effect of bentonite on the performance of PRBs.

This study aims at determining whether mixtures of bentonite, fly ash and lime can be suitable for a permeable reactive barrier. To evaluate the potential of such mixtures to effectively remediate acidic water for a long period of time, buffer capacity and leaching experiments were carried out, and changes in pH and electrical conductivity were monitored. To study the permeability of the selected mixtures, hydraulic conductivity tests were performed using water and acidic fluids. This paper briefly presents and discusses the obtained results.

## **2 EXPERIMENTAL PROGRAM**

### **2.1 Material properties**

The materials used in this study were fly ash (FA), lime and bentonite. The Tarong fly ash, which is a residue of the coal fired power station industry, was selected for this research as it is relatively inexpensive and highly reactive due to its chemical composition. According to Zaeni et al. (2010), the fly ash mainly contains  $\text{SiO}_2$  ( $\approx 66.5\%$ ),  $\text{Al}_2\text{O}_3$  (28.9%),  $\text{Fe}_2\text{O}_3$  (0.6%), and  $\text{TiO}_2$  (1.8%). It is noted that no CaO was determined in this fly ash. In fact, the low content of CaO, typically ranging from 0 to 4%, is a common feature of all types fly ash available in Queensland, Australia (Nataatmadja and Morgan, 1999).

Commercially available lime was used as a neutralising agent due to the poor buffer capacity of the Tarong fly ash (results of the buffer capacity tests will be discussed in the following sections). Owing to its high calcium content, lime has been used in several studies to increase pH levels of groundwater (Ghosh and Subbarao 2006, Indaratna et al 2006a).

The final component tested for suitability was commercially available sodium bentonite. Due to its chemical composition, bentonite is a highly plastic and expansive clay, which is used in the construction industry for backfilling, seepage barriers and waste containment practises (Kashir and Yanful 2001).

Fly ash (FA) was mixed with different amount of lime (L) to form specimens (called FAL) with different lime content such as 2.5% (FAL-2.5), 5.0% (FAL-5), 7.5% (FAL-7.5), and 10% (FAL-10) by weight. To study the effect of bentonite on the long-term buffer capacity of the reactive material, 10% and 20% bentonite (by weight) were added to the FAL-5 and FAL-10 mixtures.

### **2.2 Test procedure**

#### **2.2.1 Buffer capacity tests**

Buffer capacity is the property of a soil to absorb and/or desorb  $\text{H}^+$  and  $\text{OH}^-$  ions, and thus it determines a resistance of soil to pH changes. In this work, the buffer capacity of studied mixtures was determined experimentally by titrating each clay sample with increasing concentrations of sulfuric acid (Yong et al. 1990, Gratchev and Towhata 2010). The acid solutions were first prepared at certain

concentrations, and then added to the soil at a ratio of 1:10 for soil : acid solution, using 4 grams of dry soil mixture and 40 grams of acid solution. The pH of the soil solution was measured after the soil suspension sample was thoroughly shaken and allowed to stand for at least 24 hours.

### 2.2.2 Leaching tests

The leaching tests were similar to the buffer capacity testing procedure, with two significant differences. Firstly, only two concentrations of acid were utilised, which were 10 and 20 grams of acid (out of 40 grams of acid solution), and secondly, the soil samples were tested repeatedly, each time with a new influent liquid. Mixtures of fly ash with different amounts of lime such as 2.5%, 5%, 7.5%, and 10% by weight were tested to determine the capacity of each mixture to neutralize acidity. To study the effect of bentonite on the long-term buffer capacity of the reactive material, 10 and 20% bentonite (by weight) were added to the lime-fly ash mixtures.

Similarly to the buffer capacity tests, a soil to liquid ratio of 1:10 was adopted, which resulted in a total of 4 grams of soil measured in dry weight, and 40 grams of acidic solution. The two variations of acidic solutions with acid content equal to 10 grams and 20 grams were prepared as the influent liquid, and the pH and electrical conductivity were measured prior to it being mixed with the soil mixture. After 24 hours, the effluent liquid was removed from the container, and the liquid's pH and electrical conductivity were measured. After the measurement, the container was re-filled with the acidic influent solution to maintain the desired value of pH throughout the test. This process was repeated until the soil samples lost most of their buffer capacity, and the pH values of influent and effluent liquids became very similar.

### 2.2.3 Hydraulic conductivity tests

A series of hydraulic conductivity tests were conducted to study changes in pH and hydraulic conductivity of fly ash-lime mixtures (FAL-10) with different content of bentonite. For each test, the specimen was first compacted in the mould in three layers at the optimum moisture content, and then subjected to a constant hydraulic flow of distilled water to obtain the reference hydraulic conductivity. After the specimen was brought to equilibrium using 3 L of water permeation, acidic solution with pH=3.0 was introduced into the specimen. The hydraulic conductivity, volume of permeated liquid, and pH of the effluent liquid were continually monitored and recorded. The coefficient of permeability was determined according to the "Falling head" method (AS 1289 6.7.2-2001).

## 3 TEST RESULTS AND DISCUSSION

### 3.1 Buffer capacity tests

The titration curves of pH against sulfuric acid input for the three soil suspensions are shown in Fig. 1. Also included in the figure is the titration curve of a blank (a solution in the absence of soil). The plot shows that the buffer capacity of fly ash sample is relatively poor. The pH of effluent liquid gradually declined from 4.9 to 3.6 with increasing acid concentration, reaching a low value when the amount of acid was about 3 grams.

In contrast, the lime exhibits a high buffer capacity. The specimen with distilled water returned an effluent pH value of 12.1, and each sample after that showed only a fractural decrease in pH with an increasing quantity of acid, with the final value of pH being as high as 11.9. The buffer capacity of bentonite was also high mostly due to its high cation exchange capacity (Gratchev and Towhata 2010).

### 3.2 Leaching tests

#### 3.2.1 Leaching tests on mixtures of fly ash and lime

The first series of leaching tests were conducted on mixtures of fly ash with varying lime content: 2.5%, 5%, 7.5%, and 10% by weight. The data obtained from these tests are presented in Fig. 2a in terms of the number of rounds of leaching against the pH of effluent liquid. It is evident from this plot that the samples with higher lime content have a greater long-term buffer capacity.

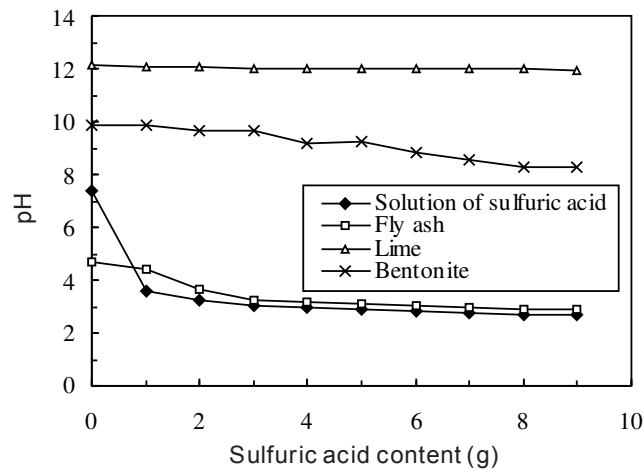


Figure 1. pH-titration curves of the Tarong fly ash, lime and bentonite.

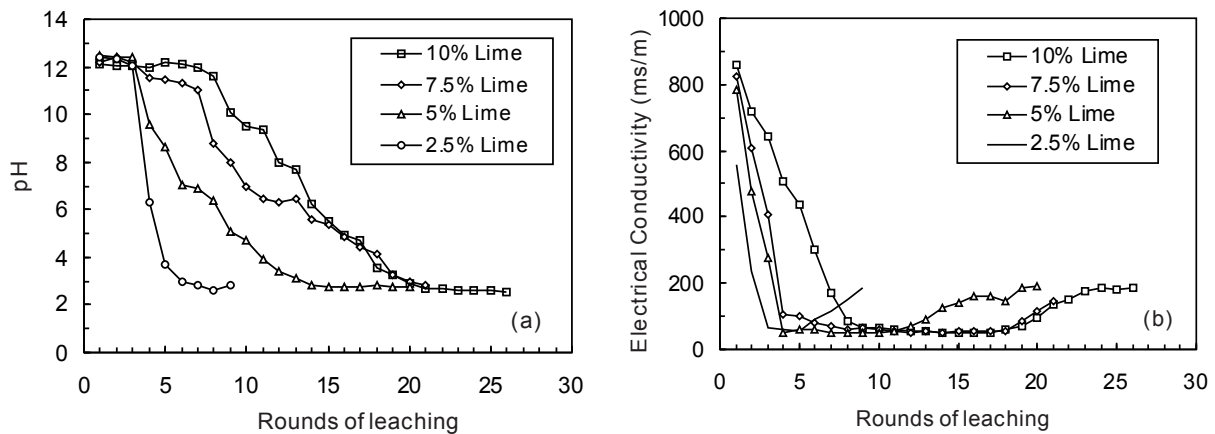


Figure 2. Results of leaching tests on mixtures of fly ash with different amount of lime plotted as rounds of leaching against pH (a), and electrical conductivity (b).

The electrical conductivity of the soil samples was also plotted against the number of rounds of leaching (Fig. 2b). It is interesting to note that the electrical conductivity decreases significantly before the pH values of the effluent liquid drops to a low level (Fig. 2a)

### 3.2.2 Leaching tests on mixtures of fly ash, lime, and bentonite

Results of leaching tests on the fly ash and lime mixtures with bentonite are presented in Fig 3. In Fig 3a, where 5% lime was used (FAL-5), the performance of mixtures with and without bentonite subjected to several rounds of leaching seems to be similar. However, when 10% lime was used in the fly ash-lime mixture (FAL-10), addition of bentonite seems to decrease the overall capacity of material to neutralize the acidic solution.

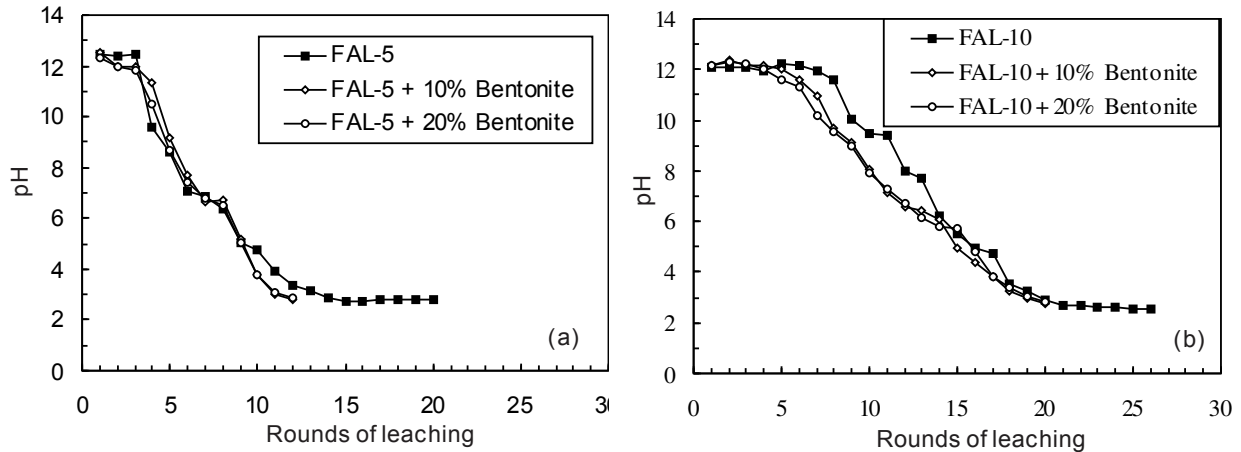


Figure 3. Results of leaching tests on mixtures of fly ash and lime with different amount of bentonite: (a) 10% and 20% bentonite was added to FAL-5; and (b) 10% and 20% bentonite was added to FAL-10.

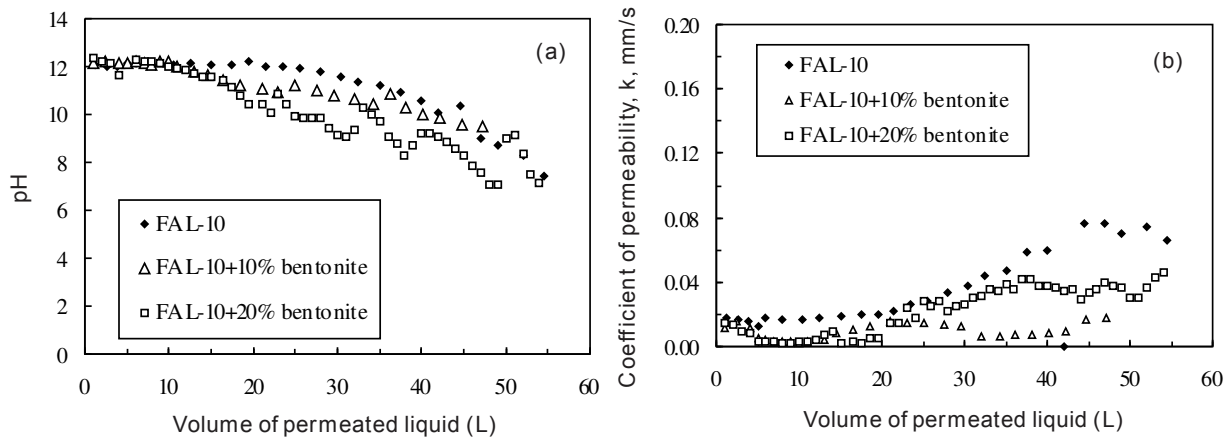


Figure 4. Results of hydraulic conductivity test on mixtures of fly ash and lime (FAL-10) with different content of bentonite: (a) effluent pH, and (b) coefficient of permeability of specimens permeated first with 3L of water and then acidic fluid.

### 3.2.3 Hydraulic conductivity test on lime-fly ash mixtures with bentonite

The results of hydraulic conductivity tests on mixtures of fly ash and lime (FAL-10), and FAL-10 with 10% and 20% bentonite permeated first with 3 L of distilled water and then with acidic liquid are presented in Fig. 4. The experiments were run for a total of approximately 55 L of water and acidic liquid permeation.

Figure 4a shows pH measurements as a function of effluent volume. First, all the samples exhibited essentially similar pH behavior during the first 10 L of water and acidic fluid permeation; that is, the initial effluent pH remained alkaline about 12. It is interesting to note that during the next 10 L of permeation, the effluent pH of FAL-10 remained almost the same while for the mixtures of FAL-10 with bentonite, the pH values decreased. It is evident from this figure that the effluent pH of FAL-10 with 20% bentonite decreased more quickly compared to that of FAL-10 with 10% bentonite. This trend continued until the end of the tests, with the FAL-10 showed the highest buffer capacity while the FAL-10 with 20% bentonite – the lowest. These results are in agreement with the data from the leaching tests (Fig. 3b), thus providing more support to the assumption that addition of bentonite may decrease the buffer capacity of lime-fly ash mixtures.

The results of “Falling head” tests on the mixtures of FAL-10, and FAL-10 with different content of bentonite are shown in Fig. 4b. It is clear from this figure that the average hydraulic conductivity of FAL-10 was initially about 0.017 mm/s within the first 3 L of water permeation. After the acidic liquid was introduced, the hydraulic conductivity first slightly increased to 0.02 mm/s after about 17 L, and then increased sharply to almost 0.08 mm/s. It is interesting to note that such an increase in the hydraulic conductivity correlates with a decrease in the pH of effluent liquid (Fig. 4a). It is believed that such changes in the hydraulic conductivity are mostly due to the leaching of calcium from the lime. For the mixtures of FAL-10 with bentonite, an increase in the hydraulic conductivity was also observed when the pH of effluent liquid began to drop. It is noted that a greater increase was found for the mixture of FAL-10 with 20% bentonite. This phenomenon can be attributed to the changes in the diffuse double layer of bentonite particles caused by high concentration of ions in the pore fluid.

#### 4 CONCLUSION

This paper presents the results of an experimental study aimed at determining the long-term buffer capacity of bentonite-lime-fly ash mixtures. Based on the obtained results, the following conclusions can be drawn:

- Increase in lime content results in a greater long-term buffer capacity of the Tarong fly ash;
- Addition of bentonite may decrease the long-term capacity of lime-fly ash mixture to neutralize acidity;
- An increase in the hydraulic conductivity of fly ash-lime mixtures containing bentonite and permeated with acidic liquid can occur due to the changes in the diffuse double layer of bentonite particles.

#### 5 ACKNOWLEDGEMENTS

The Tarong fly ash and bentonite were kindly provided for this study by Cement Australia and Sud-Chemie Australia Pty Ltd, respectively. This research was supported by the NRG grant from Griffith University, Australia.

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