



Amelioration of acidic soil using fly Ash for Mine Revegetation in Post-Mining Land

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Abstract

This paper described the use of fly ash for soil amelioration of acidic soils to promote plant growth. In mining sites, acid sulfate soils/rocks, which contain sulfide minerals (e.g. pyrite FeS_2), have appeared as a result of overburden excavation. The excessively acidic condition inhibits plant growth due to the dissolution of harmful elements, such as Al, Fe, and Mn. Fly ash, an alkaline byproduct of coal combustion generated in thermal power plants is expected to be adopted to ameliorate acidic soils. However, the mixing ratio of fly ash must be considered because excessive addition of fly ash can have a negative impact on plant growth due to its physical/chemical properties. The pot trials using *Acacia mangium* demonstrate the evolution of plant growth with a 5%–10% addition of fly ash into acidic soil. When the acidic soil has a high potential for metal dissolution, the metal ions leached from the acidic soil are large, making it difficult to improve plant growth due to osmotic and ionic stress. This work suggests that the effects of fly ash on metal ions leached from the soil have to be considered for the amelioration of acidic soil.

Keywords Acid Soil · Coal Ash · Metal Accumulation · Plant Growth

1 Introduction

There is concern that the development of open-pit mines has significant impacts on the surrounding environment, such as the loss of tropical forests and pollution of rivers. For sustainable resource management that minimizes the impact on the surrounding environment, it is necessary to consider appropriate rehabilitation processes for such mines from their early stages of development. Acid mine drainage (AMD) is a serious environmental problem that has been reported in many mining sites. The high concentrations of metal ions in AMD and their low pH not only adversely affect the water environment in surrounding rivers but also inhibit plant growth in mining areas. Several studies have reported that plant growth is inhibited in acidic

environments created by the acidification of sulfide minerals (Matsumoto et al. 2017; Takeuchi and Shimano 2009). Simultaneously, the disposal of large amounts of coal (fly) ash, an industrial waste generated from coal-fired power plants, has become an issue. In general, coal ash is disposed of in landfills. However, considering the shortage of landfills, a more effective utilization of coal ash must be studied. Currently, most of the coal ash generated is reused as a raw material for cement. However, due to its chemical composition, coal ash is projected to be used in agricultural fields as a soil amendment and a source of trace elements (Kaur and Goyal 2016; Parab et al. 2015; Shaheen et al. 2014; Yunusa et al. 2012). However, the effects of the accumulation of toxic ions must be considered when planning such measures (Manoharan, Yunusa, Loganathan, Lawrie, Murray, et al., 2010; Manoharan, Yunusa, Loganathan, Lawrie, Skilbeck, et al., 2010). Some studies have focused on the application of coal ash for solving environmental issues and promoting plant growth in mining fields (Kumar and Singh 2003; Matsumoto et al. 2016, 2019; Jayasinghe et al. 2008, 2009) examined the amelioration of Ultisols, (low productivity acidic soils) by adopting synthetic soil aggregates including fly ash. They reported that fly ash improved the physical and chemical properties of the soil and helped increase crop

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growth (Jayasinghe et al. 2008, 2009) Tsadilas et al. 2009 investigated the application of fly ash with sewage sludge in acidic Alfisols (Tsadilas et al. 2009). It has also been reported that adding fly ash to soil amendments yielded a positive impact as it increases the pH of acidic soils, which improves the adsorption of heavy metals. Similarly, Parab et al. (2015) established that bacterial activity was improved by adding fly ash to acidic soil. Although some researchers have studied the improvement of soil fertility and neutralization of acidic soil by adding fly ash, few studies have utilized fly ash with pyritic soil. In this study, we focused on alkaline coal ash, which is an industrial waste material, as a material to ameliorate an acidic mixture of granitic soil and pyrite.

2 Materials and methods

2.1 Preparation of soils for pot trials

In this study, two types of acidic soils (acidic soils 1 and 2) were prepared using decomposed granite soil with minimum neutralizing capacity and pyrite, the main cause of AMD, obtained from Furikusa, Aichi Prefecture. For fly ash, we used the raw powder produced at a coal-fired power plant in Japan. The particle size composition, paste pH, and paste EC of each soil sample were measured. The particle size was determined using a sieving process and categorized as follows: clay less than 0.005 mm, silt from 0.005 to 0.075 mm, and sand from 0.075 to 2 mm. Paste pH, an indicator of the soil acidity, was measured after mixing soil and deionized water at a ratio of 2:5, which dissolved the water-soluble components of soil (Kabala et al. 2016). Paste EC, an indicator of metal dissolution, was measured after mixing soil and deionized water at a 1:5 mixing ratio (Slavich and Petterson 1993). The pH and EC were assessed using a portable multi-water quality meter (WM-32P; DKK-TOA CORPORATION).

Next, acidic soils 1 and 2 were mixed with fly ash at ratios of 0%, 5%, 10%, 20%, and 50% to evaluate their applicability as a planting base. Paste pH, paste EC, leached metal ion analysis, and permeability tests were conducted on each soil sample. The leached metal ions, Al, Fe, and Mn, the main dissolution elements in AMD, were analyzed using the leachate from the paste EC test. Inductively coupled plasma-mass spectrometry (ICP-MS; Agilent 7500c; Agilent Technologies Japan, Ltd.) was used in this study.

Hydraulic conductivity was measured based on falling head permeability tests (ASTM D5084-10 2010).

2.2 Laboratory pot trials

To investigate the effects of fly ash application on plant growth in the acidic soils, pot trials were conducted using the prepared acidic soils 1 and 2. *Acacia mangium*, a widely planted forest tree in tropical regions, was used for the growth test. *Acacia mangium* is fast-growing and can develop even in acidic, nutrient-poor soils. For the test, plastic pots with a diameter of 15 cm were filled with each soil sample, and seedlings were planted at a height of approximately 13 cm. After planting, the pots were placed in the Phytotron Glass Room A-1, the artificial climate chamber at the Center for Bio-Environmental Sciences, Kyushu University. The plants were maintained at 30 °C and 70% relative humidity; irrigated with 500 mL of water per pot every 3 to 4 days; and fertilized with a liquid fertilizer (N-P-K=6-10-5) weekly. For the test, five seedlings were planted in one soil sector, and growth progress was observed for 11 weeks. The dry weight of the whole plant was measured at the end of the growth test. The plant samples were washed with deionized water at room temperature using sonication (UT-106 H, SHARP) to remove soil particles, and then they were dried at 60 °C for 72 h. Additionally, the plant samples were pulverized using a mortar and pestle and completely digested using 5 mL of a mixture of 61% nitric acid (HNO₃) and 35% hydrochloric acid (HCl) at a ratio of 3:1 at 110 °C in a DigiPREP Jr. (SCP Science, Quebec, Canada) (Quadir et al. 2011). The solutions were subjected to ICP-MS after filtration through a 0.45 μm membrane to quantify the amounts of Al, Fe, and Mn.

3 Results and discussion

3.1 Evaluation of soils as planting basement

Table 1 lists the fundamental properties of each sample. Acidic soils 1 and 2 were categorized as sandy loam, while fly ash was categorized as silt loam due to its abundant silt content. The results showed that acidic soil 2 had a lower pH and higher EC than acidic soil 1. In addition, the pH of fly ash shows alkalinity.

Table 2 shows the variation in soil composition under different mixing ratios of acidic soils and fly ash. The silt

Table 1 Soil compositions, Paste pH, Paste EC

Sample	Sand (%)	Silt (%)	Clay (%)	Soil Texture	Paste pH	Paste EC (mS/cm)
Acidic soil 1	84.5	7.3	8.3	Sandy Loam	3.70	1.32
Acidic soil 2	85.8	7.2	7.0	Sandy Loam	3.10	3.50
Fly ash	9.0	75.3	15.7	Silt Loam	12.60	3.20

Table 2 Soil compositions of mixed soils

Sample	Fly ash (%)	Sand (%)	Silt (%)	Clay (%)	Soil texture
Acidic Soil 1	0	84.5	7.3	8.3	Sandy Loam
	5	81.1	10.3	8.6	Sandy Loam
	10	77.7	13.3	9.0	Sandy Loam
	20	70.9	19.3	9.8	Sandy Loam
	50	50.7	37.3	12.0	Loam
Acidic Soil 2	0	85.8	7.2	7.0	Sandy Loam
	5	82.4	10.2	7.4	Sandy Loam
	10	78.9	13.2	7.9	Sandy Loam
	20	72.0	19.2	8.7	Sandy Loam
	50	51.4	37.3	11.3	Loam

and clay contents increased as the ratio of fly ash in the mixture increased. The soil texture changed from “Sandy Loam” to “Loam” because of the increase in fine content. This increase in the fine content caused a decrease in soil permeability, thereby inhibiting plant growth. Thus, addition of excessive fly ash in the mixture should be avoided.

Table 3 shows the results of each test and the standard values for the planting base. Table 3 shows that the paste pH increased along with the amount of fly ash, thereby improving the acidic soil. However, in acidic soil 1, the paste pH became alkaline when the amount of fly ash mixed was 50%, suggesting that excessive amounts of fly ash caused alkalinization of the soil. In acidic soil 1, the paste EC decreased when the amount of fly ash was 5% and 10%. In the case of acidic soil 2, paste EC was decreased by increasing the amount of fly ash up to 50%, but the improvement was not sufficient compared with the standard values (Research Committee of Japanese Institute of Landscape Architecture 2000). In addition, the hydraulic conductivity decreased as the amount of fly ash increased in both acidic soils, suggesting that an excessive amount of fly ash may cause the planting base to have a poor permeability.

Table 4 shows the dissolved metal ions in the leachate of the paste EC test. Excessive Fe ion elution was detected in the acidic soil, which tended to decrease as the amount of fly ash increased, that is, as the pH of the elution solution

became higher. This indicates that the metal ion elution due to AMD generation can be reduced with the addition of fly ash, but alkalization from adding excessive amounts must be considered. Additionally, compared with acidic soil 1, acidic soil 2 showed more elution of all metal ions.

3.2 Plant growth

Figures 1 and 2 show the increase in plant height and basal diameter after 1, 4, 8, and 11 weeks in each soil sample. For the control, soil conditions results without pyrite and fly ash are also shown. In the acidic soil 1, the plant height and basal diameter increased with elapsed time with the addition of 5%, 10%, and 20% fly ash. Improvement in plant growth could not be confirmed for the other cases because the plants died. At 11 weeks, plants in acidic soil 1 supplemented with 5%, 10%, and 20% fly ash showed height increments of 40.6 mm, 47.3 mm, and 34.0 mm, respectively, compared with the 110.5 mm in the control pot. The basal diameters showed a similar tendency as the plant height. At 11 weeks, plants in acidic soil 1 showed basal diameter increments of 0.86 mm, 0.90 mm, and 0.58 mm with 5%, 10%, and 20% of fly ash addition, respectively, compared to the 1.80 mm in the control pot. Slight increments or no improvements in plant height and basal diameter were found in acidic soil 2. Overall, the growth of plants in acidic soil 1 was improved

Table 4 Dissolution of dominant metal ions in AMD

Sample	Fly ash (%)	Al (ppm)	Fe (ppm)	Mn (ppm)
Acidic Soil 1	0	34.1	298	14.2
	5	5.4	143	8.1
	10	15.7	161	9.3
	20	17.6	143	9.3
	50	14.1	17	0.2
Acidic Soil 2	0	20.2	1067	20.7
	5	24.6	1167	23.5
	10	14.4	814	17.8
	20	31.8	705	16.2
	50	26.2	187	6.4

Table 3 Paste pH, Paste EC, hydraulic conductivity of mixed soils

Sample	Fly ash (%)	Paste pH	Paste EC (mS/cm)	Hydraulic conductivity (cm/s)
Acidic Soil 1	0	3.70	1.32	5.70×10^{-4}
	5	5.90	1.06	2.16×10^{-4}
	10	6.35	1.20	1.34×10^{-4}
	20	8.44	1.33	1.13×10^{-4}
	50	11.22	1.49	1.56×10^{-5}
Acidic Soil 2	0	3.10	3.50	7.43×10^{-4}
	5	4.83	3.30	2.84×10^{-4}
	10	5.64	2.80	1.45×10^{-4}
	20	6.13	2.80	1.33×10^{-4}
	50	7.87	2.40	3.34×10^{-5}
Standard		4.5–7.5	0.1–1.0	1.0×10^{-4} – 1.0×10^{-1}

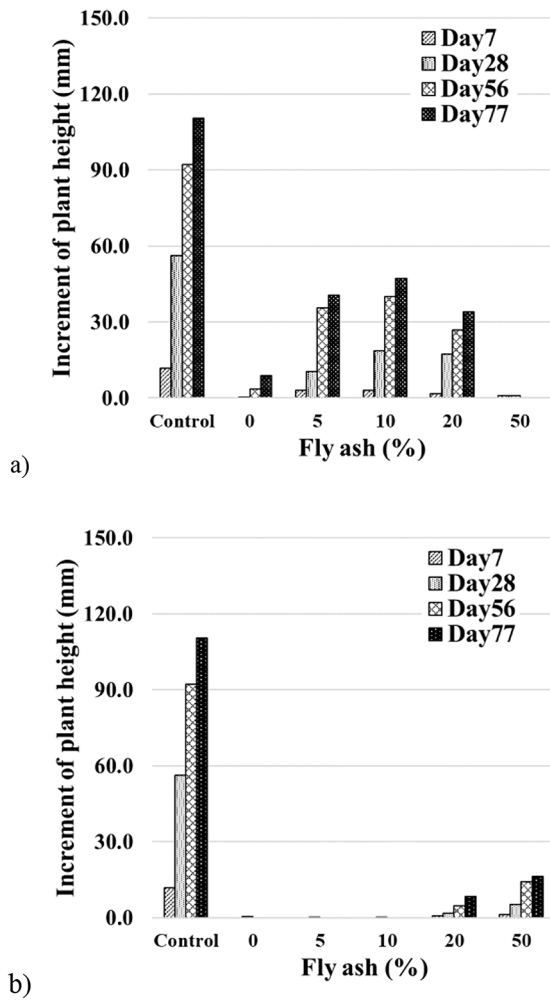


Fig. 1 Increment of plant height: **a** Acidic soil 1, **b** Acidic soil 2

at a certain ratio of fly ash, while the growth of plants in acidic soil 2 did not improve at any ratio of fly ash.

Since seedling growth is related to plant height, number of leaves, basal diameter, and volume of roots, it is necessary to examine the differences in plant growth between the soils using dry weight. Figure 3 shows the seedlings' dry weights measured at the end of the test. The Tukey-Kramer method (significance level $\alpha=0.05$) was used to identify significant differences in the test results; these differences have been indicated in the figures using different letters for the same item. Here, "significant difference" means that there is a difference between the groups that cannot be considered to have occurred by chance.

The results confirmed a difference in the growth of seedlings in acidic soil 1 under different treatments. This difference was more pronounced when the amount of fly ash was 5–10%, suggesting that the difference in the growth of

seedlings was due to the difference in the soil type. According to the values of paste EC shown in Table 3, acidic soil 1 containing 5–10% of fly ash had the lowest EC value among the soils because the paste pH was almost neutral. This indicates that plant growth can be promoted by adding fly ash at a suitable ratio to neutralize soil paste pH. However, excessive amounts should be avoided because large amounts of metals be eluted from fly ash. Several researchers have suggested that fly ash should be added to soil within an allowable range (Matsi and Keramidas 1999; Mishra & Shukla, 1986). There was no observable growth of seedlings when the amount of fly ash was 0% or 50%. When the amount of fly ash was 0%, plant root growth was inhibited due to metal dissolution from the soil under acidic conditions. When the amount of fly ash was 50%, nutrient absorption was inhibited as excessive amounts of fly ash caused alkalization of the soil and decreased its permeability.

There was no significant difference in acidic soil 2 regardless of the ratio of fly ash because no improvement in growth was observed upon mixing fly ash. The growth of seedlings was inhibited by osmotic stress and ionic stress due to the high soil paste EC. Osmotic stress is caused by high salt concentrations in the soil; it inhibits the ability of plants to absorb water and causes a water deficit (Bernstein, 1975). The soil EC of acidic soil 2 exceeded the standard value according to Table 3, indicating that the osmotic stress inhibited the water uptake by roots. Ionic stress is caused by excessive concentrations of certain ions contained in the plant; it disrupts the nutrient balance of plants, causing leaf death and growth inhibition.

Table 5 shows the metal accumulation in the plant body, which was derived by investigating the seedling after the pot trials had finished. The metal accumulation in the plant body was consistent with the dissolution of metal ions from the soils, as shown in Table 4, indicating that the plant in acidic soil 2 contained much higher Al, Fe, and Mn concentrations than that in acidic soil 1, especially in the 0–10% mixing ratios. In summary, both osmotic and ionic stresses had adverse effects on plant growth in acidic soil 2. The contents of Al and Fe were 172 mg/kg and 561 mg/kg, respectively, for acidic soil 1 containing 5% fly ash and 227 mg/kg and 425 mg/kg for control, respectively, indicating that these elements are comparable. Considering that Al has an adverse impact on plant growth and Fe is a major element eluted from pyrite, plant growth in acidic soil 1 with 5% fly ash was improved compared to that in the other fly ash ratio due to the reduction in Al dissolution and inhibition of Fe uptake into the plant body. The content of Mn was 571 mg/kg in acidic soil 1 and 39 mg/kg in the control, suggesting that Mn did not significantly affect the plant growth of *Acacia mangium*.

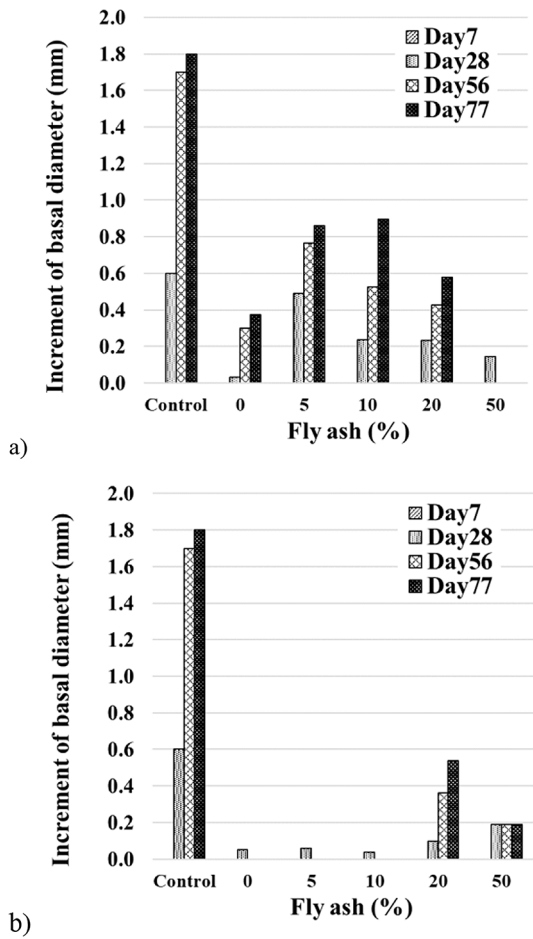


Fig. 2 Increment of basal diameter: a Acidic soil 1, b Acidic soil 2

Table 5 The metal accumulation in the plant body

Sample	Fly ash (%)	Al (mg/kg)	Fe (mg/kg)	Mn (mg/kg)
Acidic Soil 1	0	1837	3911	3410
	5	172	561	571
	10	762	2283	704
	20	552	1776	182
	50	542	919	29
Acidic Soil 2	0	13,463	110,482	5932
	5	23,413	137,257	14,008
	10	7416	26,840	7474
	20	1057	6404	2115
	50	2526	4140	428
Control	-	227	425	39

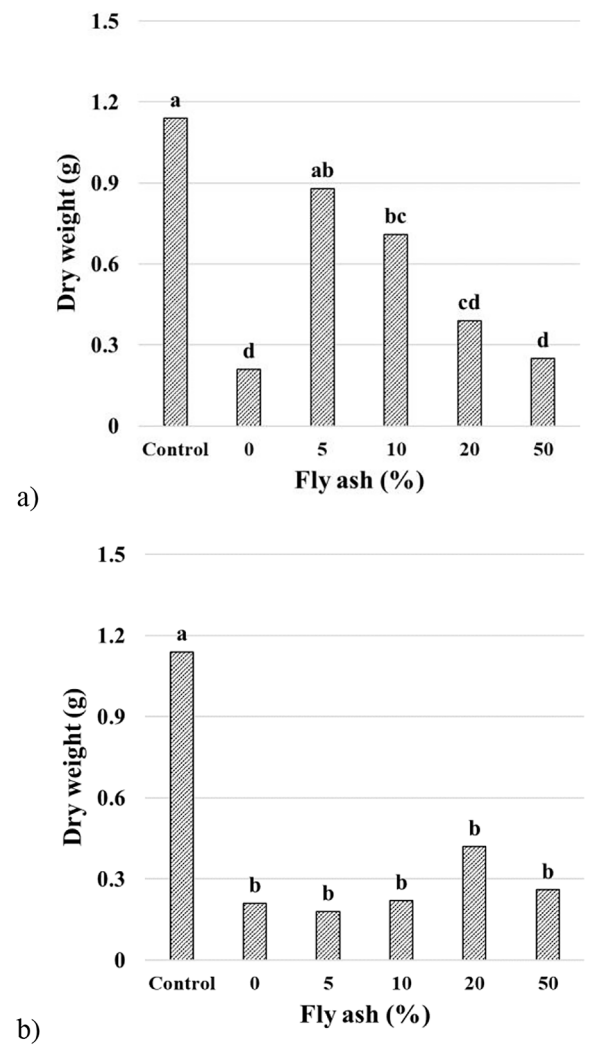


Fig. 3 Dry weight of *Acacia mangium*: a Acidic soil 1, b Acidic soil 2

4 Conclusions

In this study, we aimed to use fly ash, an industrial waste, as an acid-neutralizing material for acidic soils. We conducted potted plant trials using *Acacia mangium*. It was found that it is possible to improve plant growth using fly ash when the amounts of metal ions leached from the acidic soil were small. However, it is impossible to improve plant growth when the acidic soil has a higher potential to leach metal ions. This means that it is difficult to improve plant growth when acidic soils have a high metal ion leaching potential due to osmotic and ionic stresses. Therefore, the effects of fly ash and the metal ions leached from the soil must be considered along with the soil pH when fly ash is applied to acidic soils.

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Availability of data and materials The data used to support the findings of this study are available from the corresponding author upon request.

Declarations

Conflict of interest The authors declare no conflict of interest.

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