

Securing Topsoil for Rehabilitation Using Fly Ash in Open-Cast Coal Mines: Effects of Fly Ash on Plant Growth

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<http://doi.org/10.29227/IM-2019-01-02>

Submission date: 11-07-2018 | Review date: 02-04-2019

Abstract

Rehabilitation is an important stage in mining operations for environmental conservation. However, the shortage of topsoil makes it difficult to achieve rehabilitation in open-cast coal mines. Securing topsoil by mixing soil with fly ash (FA), which is treated as an industrial waste, is expected to solve this issue in coal mines. While mixing soil with FA makes it possible to secure the topsoil and treat industrial waste simultaneously, the high alkalinity of FA and the dissolution of heavy metals from FA may inhibit plant growth. This study investigated the effects of FA in the topsoil on plant growth via vegetation tests with simulated topsoil mixed with FA using *Acacia mangium*, a species of flowering tree: the FA mixing ratios were set to 0%, 20%, 40%, 60%, and 100%. The growth of *Acacia mangium* was inhibited with increasing FA mixing ratio, especially from 60% to 80%. However, the growth rate of *Acacia mangium* in an FA mixing ratio of 100% was nearly comparable to that in a mixing ratio of 40%. Furthermore, there were no effects of the physical characteristics and pH conditions in the topsoil on the plant growth at any of the mixing ratios; meanwhile, the accumulated concentration of Al in the plant body increased significantly at an FA mixing ratio of 60%–80%. This suggests that the accumulation of Al, which inhibits plant growth, including root growth and its functions, in the plant body inhibited the growth of *Acacia mangium*. Therefore, the most important aspect in terms of rehabilitation concerning the use of FA for securing topsoil is not the mixing ratio of FA but the amount of Al in the FA and the accumulation of Al in the plant body.

Keywords: open-cast coal mine, fly ash, topsoil, rehabilitation, *acacia mangium*, aluminum (Al)

Introduction

Securing the topsoil is necessary for the rehabilitation of open-cast mines where disturbed mine soils inhibit plant growth (Eludoyin et al., 2017). Fly ash (FA), considered to be industrial waste, has been applied to topsoils to secure the amount of topsoil (Gorman et al., 2000; Truter et al., 2013). In addition, the application of FA to topsoils is expected to inhibit the effects of heavy metals on plant growth in soils by increasing the pH because FAs are highly alkaline (Basu et al., 2009; Shende et al., 1994). However, because several factors, such as high alkalinity and heavy metals in FA, may affect the growth of plants, the critical factors in the application of FA to topsoils should be determined in terms of their effects on plant growth (Pandey et al., 2010; Shende et al., 1994). Furthermore, the change in the physical characteristics of the topsoil after being mixed with FA plays an important role in the soil conditions for plant growth and, therefore, needs to be considered (Binal, 2016). Few studies have attempted to use FA to secure topsoil with a focus on the effects on plant growth taking into account multiple factors. This study discusses critical factors for the application of FA to securing topsoil in terms of the effects of FA on plant growth via a vegetation test with simulated topsoil with different FA mixing ratios from 0% to 100% in addition to some results that have already been reported (Hamanaka et al., 2017).

Materials and methods

Simulated soils with fly ash

The simulated soils were prepared by mixing decomposed granite produced by Gulin Kita-Kyu Co., Ltd. and Kyushu bentonite produced by Shinagawa Yogyo Co., Ltd. after screening based on soil texture, which was based on the post-mining land in a coal mine in Indonesia: the ratios of sand, silt, and clay were set to 51.1%, 25.2%, and 23.7%, respectively, which were within the range of the soil textures at the mine (sand: 22.0%–75.0%, silt: 8.0%–32.1%, and clay: 17.0%–48.0%) (Matsumoto et al., 2016; Matsumoto et al., 2018). The FA was sampled from a coal-fired power plant in Japan and was analyzed via X-ray fluorescence (XRF). Based on the chemical components of the XRF analysis, the FA was characterized according to ASTM C618-12a (ASTM C618-12a, 2012). The FA was homogeneously mixed with the simulated topsoil using the cone and quartering method at different mixing ratios (0% no FA, 20%, 40%, 60%, 80%, and 100%) and was labeled accordingly.

Material characteristics of the simulated soils with fly ash

To understand the physical characteristics of the simulated soils with FA, Atterberg limit tests were conducted based on the ASTM D4318-05 standard (ASTMD4318-05, 2005) in addition to particle-size distribution tests (ASTM D422-63, 2007), density tests (ASTM D854-14, 2014), and falling head permeability tests (ASTM D5084-10, 2010). Moreover, the

Tab. 1. Chemical composition of FA
Tab. 1. Skład chemiczny popiołów lotnych

Elements	SiO ₂	TiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MnO	MgO	CaO	Na ₂ O	K ₂ O
(%)	67.4	1.08	18.7	4.16	0.03	1.10	2.87	0.67	1.23
Elements	P ₂ O ₅	H ₂ O	SO ₃	V	Cr	Cu	Zn	Pb	As
(%)	0.33	1.97	0.22	0.022	0.004	0.009	0.015	0.006	0.002

Tab. 2. Physical characteristics of the simulated topsoils with FA
Tab. 2. Właściwości fizyczne badanych składów wierzchnich warstw gleby z popiołem lotnym

FA (%)	Sand (%)	Silt (%)	Clay (%)	Soil Type	Paste pH	Density (g/cm ³)	Permeability (×10 ⁻⁵ cm/s)	Liquid Limit (%)	Plastic Limit (%)	Plasticity Index (%)
0	51.1	25.2	23.7	CL	5.54	2.41	29.4	28.1	17.4	10.8
20	40.3	37.4	22.3	CL	10.49	2.28	19.3	23.5	14.8	8.8
40	31.1	48.3	20.6	SiCL	11.08	2.23	9.2	-	14.1	-
60	23.0	60.4	16.5	SiCL	11.72	2.18	5.3	-	-	-
80	17.9	65.1	17.0	SiCL	11.55	2.13	3.8	-	-	-
100	9.0	75.3	15.7	SiCL	11.67	2.07	2.5	-	-	-

change in the pH was reported as paste pH after 12 h of the dissolution process at a 1:2 of mixing ratio of the samples and deionized water to characterize the chemical characteristics of the simulated soils with FA (AMIRA, 2002). The effects of the material characteristics were inferred based on the results because such soil conditions are known to affect the growth of plants (Keller and Dexter, 2012).

Vegetation test with *Acacia mangium*

A vegetation test with *Acacia mangium* was conducted using the simulated topsoil with FA to determine the critical factor for the application of FA to topsoil securing in terms of the effect of FA on plant growth. Due to its high adaptability, *Acacia mangium* was used for the vegetation test; the goal was to obtain a clear distinction in the plant growth for various mixing ratios. The seeds were obtained in Japan. *Acacia mangium*, which grows in tropical forests in Southeast Asian countries, is considered to be an Al-excluder plant and has a resistance to poor soil conditions (Osaki et al., 1997). *Acacia mangium* has been applied successfully worldwide to the reclamation of post-mine lands containing bauxite, copper, coal, and iron.

The prepared simulated soils at different FA mixing ratios (0% no FA, 20%, 40%, 60%, 80%, and 100%) were evenly mixed in each flower pot at a constant rate to provide uniform physical conditions in each pot to be used for the vegetation test. The vegetation test was performed in the phytotron glass room G-9 in the Biotron Application Center, Kyushu University, under the following conditions: 30°C room temperature and 70% relative humidity assuming the local climate at the mine site in Indonesia. The seedlings were transplanted at approximately 40–120 mm into the pots with different FA mixing ratios. Five plants were planted in pots for each FA mixing ratio, and their heights and diameters were measured every week. A total of 500 mL of water was supplied to the pots every 3–4 days. A liquid fertilizer, HYPONeX-R (N-P-K = 6-10-5), diluted to 1,000 mg/L with water was added to the pots once a week. The test was continued for 133 days until a clear distinction was observed (Hamanaka et al., 2017). The results were analyzed via the F test with the means compared

via the Tukey test at a 5% level of probability (Dinardo et al., 1998). In addition, the pH and electric conductivity (EC) in the leachate from each pot were measured along with the measurement of the heights and diameters of the *Acacia mangium* plants.

At the end of the vegetation test, the accumulated concentrations of Al, As, B, Cd, Cr, Cu, Fe, and Zn in the *Acacia mangium* plants were determined by dissolving samples with acids. The plant samples were washed with deionized water using sonication (UT-106H, SHARP) at room temperature to remove soil particles. They were dried at 60°C for 72 h and pulverized using a mortar and pestle, including each part of the plants: leaves, stems, and roots. A total of 0.25 g of each part of the samples was digested by 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) until they were completely digested according to the description by Quadir et al. (2011). If the sample was not dissolved in the mixture, 1 mL of the mixture was added and the dissolution process was repeated. The volume of the solution was adjusted to 20 mL by adding deionized water, and the solutions were injected into an inductively coupled plasma-atomic emission spectrometer (ICP-AES, VISTA-MPX ICP-OES (Seiko Inst., Japan)) after filtration with a 0.45-μm membrane filter to measure the concentrations of Al, As, B, Cd, Cr, Cu, Fe, and Zn, which are thought to affect plant growth. The results were calculated in mg per dry unit weight (mg/g). In addition to the ratio of each part, the standard deviations of the accumulated ions in each part of the *Acacia mangium* plant were calculated.

Results and discussion

Table 1 shows the chemical composition of the FA, and Table 2 summarizes the material characteristics of the simulated soil with different mixing ratios of FA. The composition of SiO₂ has the highest value, followed by Al₂O₃ and Fe₂O₃. The CaO and MgO contents reflect the clay mineral content and the neutralization effects in the FA. Because the simulated soils with different FA mixing ratios can create alkaline soil conditions and provide dissolved metals, the effects of

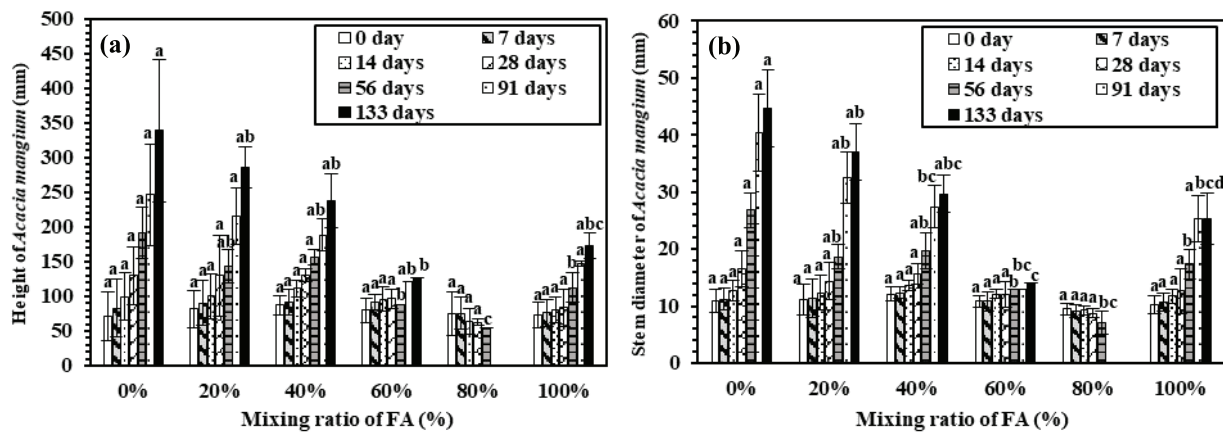


Fig. 1. Change in the (a) height and (b) stem diameter of *Acacia mangium* at different FA mixing ratios in the simulated topsoils. Means within the height and stem diameter of *Acacia mangium* with the same letter are not significantly different ($P < 5\%$) from each other according to the Tukey test

Rys. 1. Zmiana wysokości (a) i (b) średnicy *Acacia mangium* przy różnych dodatkach popiołów lotnych do badanych gleb.

W teście Tukeya różnice dla tych samych wysokości i średnicy łodyg nie przekraczają $P < 5\%$.

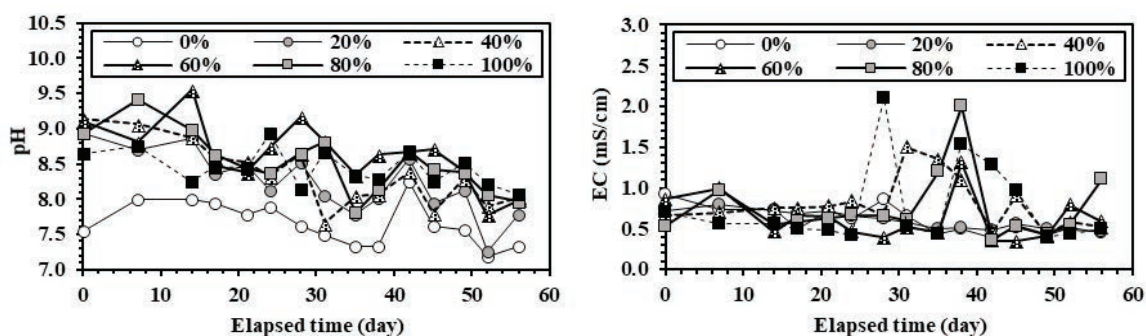


Fig. 2. Change in the pH and EC in the leachate during the vegetation test

Rys. 2. Zmiana pH i EC w odcieku podczas testu wegetacji

dissolved metals and the pH conditions on the plants needs to be investigated in this study. According to the classification by the chemical composition and the ASTM C618 standard, the FA in this study shows a similar chemical composition to class F, i.e., the $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$ (%) was 90.3% ($>70.0\%$ which is assigned to class F). Considering that FAs that are classified as classes C and F have up to 5.0% SO_3 , the level of SO_3 in the FA in this study had a relatively low value (Page et al., 1979). According to Table 2, the soil texture of the FA in this study primarily consisted of silt, decreasing the permeability with increasing FA mixing ratio in the simulated soil. The soil classification changed at 40% from CL to SiCL with a significant decrease in the permeability. Moreover, the liquid limits could not be measured at 40%, indicating that there was a significant change in the physical characteristics of the FA at a ratio of 40%. There was no remarkable change in the specific gravity of the simulated soil, which was 2.07–2.41 (g/cm^3). The paste pH changed from 5.54 at an FA mixing ratio of 0% in the simulated soil to 11.67 at an FA mixing ratio of 100% due to the alkalinity of the CaO in the FA. The rapid change in the paste pH at an FA mixing ratio of 20% indicated that the alkaline condition might affect the growth of plants at this mixing ratio. Because physical effects of the soil on the plant growth have been observed due to the higher subsurface water retention resulting from the high permeability in addition to the pH effects, the effects of the physical properties of the soils at 40% and that of the pH effects at 20% need to be

investigated during the vegetation test to determine if these are critical factors for the application of FA to topsoil in terms of their effects on plant growth (Korcak, 1996).

Figure 1 shows the change in the (a) height and (b) stem diameter of *Acacia mangium* at different FA mixing ratios in the simulated topsoils. The height and stem diameter decreased with increasing FA mixing ratio until 80%, and then they rose at 100%. After only 56 days at 80%, five plants withered during the vegetation test. These results suggest that the application of FA in the topsoil may inhibit the growth of *Acacia mangium*. Even though significant differences in the heights and stem diameters of *Acacia mangium* were not observed until 28 days for all mixing ratios according to the results of the F test, there were differences after 56 days for mixing ratios of 60% and 80%. This indicates that the mixture of FA affected the plant growth while the *Acacia mangium* plants were growing. Considering the significant change in the physical characteristics of the simulated topsoil with an FA mixing ratio of 40% and that of the paste pH at an FA mixing ratio of 20% in Table 2, the significant differences in the height and stem diameter of *Acacia mangium* after 56 days at FA mixing ratios of 60% and 80% suggest that the growth of *Acacia mangium* was not inhibited by the change in the physical properties and pH conditions in the simulated soil during the vegetation test. The change in the pH and EC in the leachate from 0 days to 56 days is plotted in Figure 2. The pH remained at approximately 8.0–9.0 at FA mixing

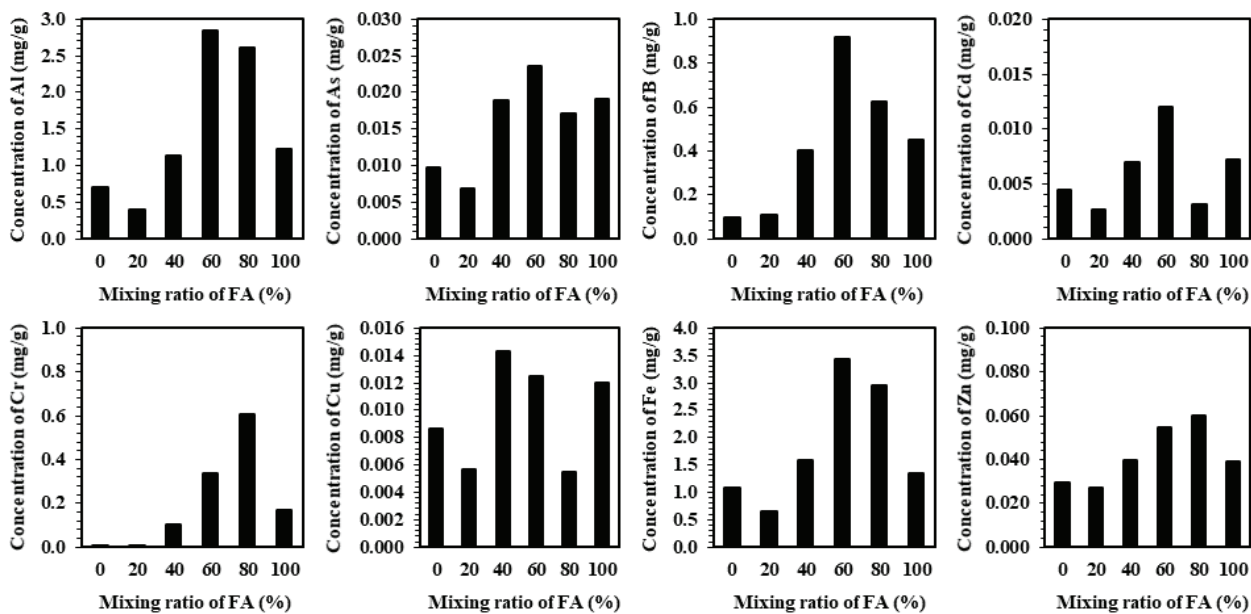


Fig. 3. Accumulated chemical concentrations in the body of *Acacia mangium*
Rys. 3. Skumulowane stężenia chemiczne w roślinie *Acacia mangium*

Tab. 3. Standard deviation of the accumulated ions in the body of *Acacia mangium*
Tab. 3. Odchylenie standardowe nagromadzonych jonów w *Acacia mangium*

FA (%)	Al	As	B	Cd	Cr	Cu	Fe	Zn
0	0.84	0.01	0.05	0.00	0.01	0.01	1.29	0.01
20	0.23	0.00	0.04	0.00	0.00	0.00	0.32	0.00
40	1.51	0.01	0.34	0.01	0.14	0.01	2.12	0.02
60, 80	-	-	-	-	-	-	-	-
100	1.84	0.02	0.46	0.01	0.41	0.02	1.32	0.03

ratios of 20–100% until 56 days, indicating that the growth of *Acacia mangium* was not affected by the alkaline conditions attributed to FA. The EC also remained at EC = 0.5–1.0 mS/cm at for all mixing ratios until 56 days, suggesting a continuous dissolution of ions from the simulated topsoil during the vegetation test.

Figure 3 summarizes the accumulated concentrations of Al, As, B, Cd, Cr, Cu, Fe, and Zn at the end of the vegetation test. High concentrations of Al and Fe were observed at FA mixing ratios of 60–80% compared to those at other FA mixing ratios: only Al and Fe showed more than 1.0 mg/g at FA mixing ratios of 60–80%. As Table 1 shows, the Al and Fe concentrations were attributed to the high content of Al and Fe in the FA. The lower concentrations of Al and Fe at an FA mixing ratio of 100% are thought to be due to the spatial distribution bias within the FA.

Tables 3 and 4 summarize the standard deviations of the accumulated ions in the body of *Acacia mangium* and the accumulated ions in each part of the body of *Acacia mangium*; the standard deviation could not be calculated at FA mixing ratios of 60% and 80% because they could not be separated into leaves, stems, and roots. The standard deviations of Al and Fe clearly increased with the FA mixing ratio and were greater than 1.0 at mixing ratios of 40% and 100%, showing an uneven distribution of Al and Fe in the plant body. In Table 4, approximately 90% of the Al and Fe accumulated in the roots (93–94% of the Al and 88–93% of the Fe at mixing

ratios of 40% and 100%). The suction of water from the roots caused the accumulation of the metals on the roots. Al easily dissolves as $\text{Al}(\text{OH})_4^-$ under alkaline conditions, inhibits nutrition absorption via nitrogen fixation on roots, and precipitates due to a detoxification mechanism of rhizotoxic mononuclear Al species, leading to a physical block to the transport of nutrients and other solutes into the apoplast necessary for root growth (Saifuddin et al., 2016; Stass et al., 2006). Even though As, Cd, Cr, and Cu accumulated on the roots at FA mixing ratios of 60–90%, the high concentration of Al (more than 2.0 mg/g at mixing ratios of 60–80%) indicates that the critical factor causing the inhibition of growth in *Acacia mangium* was the accumulation of Al derived from the FA in this study. In addition, the results show that the FA in this study can be applied to the simulated topsoil in ratios up to 40% in terms of plant growth; however, not only the FA mixing ratio but also the growth rate of plants and the accumulation of Al in the plants needs to be considered via continuous monitoring at mines. In addition, the effects of the application of FA to topsoil need to be investigated with other plant species without a resistance to poor soil conditions.

Conclusion

This study investigated the effects of FA on plant growth via vegetation tests with simulated topsoil mixed with FA using *Acacia mangium* plants; the FA mixing ratio was set to 0–100%. The growth of *Acacia mangium* was inhibited with

Tab. 4. Ratios of the accumulated ions in each part of the body of *Acacia mangium*
 Tab. 4. Udział zakumulowanych jonów w poszczególnych częściach *Acacia mangium*

FA (%)	(%)	Al	As	B	Cd	Cr	Cu	Fe	Zn
0	Leaf	5	22	29	11	9	11	6	25
	Stem	4	9	13	10	10	10	3	18
	Root	91	69	58	79	81	79	91	56
20	Leaf	18	41	63	34	11	22	18	33
	Stem	8	31	19	35	14	22	8	36
	Root	74	28	18	31	74	56	74	31
40	Leaf	4	35	73	27	3	14	4	28
	Stem	3	8	5	7	4	6	3	13
	Root	93	57	22	66	93	81	93	59
60, 80	-	-	-	-	-	-	-	-	-
100	Leaf	5	73	69	38	4	17	11	39
	Stem	1	5	3	4	0	2	1	5
	Root	94	22	29	58	95	81	88	56

increasing FA mixing ratio, especially from 60% to 80%. Conversely, the growth rate of *Acacia mangium* at an FA mixing ratio of 100% was nearly comparable to that at an FA mixing ratio of 40%. Furthermore, there were no effects of the physical characteristics and pH conditions in the topsoil on the plant growth at all the mixing ratios; meanwhile, the accumulating concentration of Al in the plant body increased significantly at FA mixing ratios of 60–80%. This suggests that the accumulation of Al, which inhibits plant growth, including root growth and its functions, in the plant body inhibited the growth of *Acacia mangium*. Therefore, the

most important aspect in terms of rehabilitation concerning the use of FA for securing topsoil is not the FA mixing ratio but the amount of Al in the FA and the accumulation of Al in the plant.

Acknowledgements

The authors are indebted to the Biotron Application Center, Kyushu University, for the vegetation test. This work was partly supported by JSPS KAKENHI Grant Number 17H07401. The authors would like to thank Enago (www.enago.jp) for the English language review.

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Przygotowanie wierzchniej warstwy gleby do rekultywacji przy użyciu popiołu lotnego w kopalniach odkrywkowych: wpływ popiołów lotnych na wzrost roślin

*Rekultywacja jest ważnym etapem w eksploatacji górniczej w zakresie ochrony środowiska. Brak wierzchniej warstwy gleby utrudnia osiągnięcie efektu rekultywacji w kopalniach odkrywkowych. Oczekuje się, że zabezpieczenie wierzchniej warstwy gleby przez zmieszanie gleby z popiołem lotnym (FA), który jest traktowany jako odpad przemysłowy, rozwiąże ten problem w odkrywkowych kopalniach węgla. Podczas mieszania gleby z FA możliwe jest jednoczesne zabezpieczenie wierzchniej warstwy gleby i utylizacja odpadów przemysłowych. Jednak wysoka alkaliczność FA i rozpuszczanie metali ciężkich z FA może hamować wzrost roślin. W artykule przedstawiono wpływ dodatku FA do wierzchniej warstwy gleby na wzrost roślin poprzez testy roślinności z symulowaną wierzchnią warstwą gleby zmieszaną z FA przy użyciu *Acacia mangium* i gatunków drzew kwitnących. Proporcje mieszanek FA zostały określone na 0%, 20%, 40%, 60% i 100%. Wzrost *Acacia Mangium* był hamowany wraz ze wzrostem zawartości FA, zwłaszcza od 60% do 80%. Jednak tempo wzrostu w mieszance FA w proporcji 40% było praktycznie takie jak bez dodatku FA. Ponadto nie zaobserwowano wpływu właściwości fizycznych i warunków pH w wierzchniej warstwie gleby na wzrost roślin w żadnej mieszance. Oznacza to, że skumulowane stężenie Al w roślinie znacznie wzrosło dla mieszanek z dodatkiem FA 60% -80%. Sugeruje to, że nagromadzenie Al, które hamuje wzrost roślin, w tym wzrost korzeni i jego funkcje, w organizmie rośliny hamuje wzrost *Acacia Mangium*. Dlatego najważniejszym aspektem rekultywacji z zastosowaniem FA do zabezpieczania wierzchniej warstwy gleby nie jest wielkość dodatku FA, lecz ilość Al w FA i akumulacja Al w roślinie.*

Słowa kluczowe: kopalnia odkrywkowa, popiół lotny, wierzchnia warstwa gleby, rekultywacja, Acacia mangium akacja mangowa, aluminium (Al)