Effects of Electrokinesis on Arsenic Mobility and Accumulation in Mott Dwarf Napier Grass for Treatment of Mine Tailings

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Research

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Abstract

This research firstly aimed to study the potential of electrokinetics (EK) on arsenic (As) mobility in gelatin (agar) medium with different containers (spherical or rectangular), electrodes (graphite or stainless-steel grade 316L), and applied electric field (0, 1, and 2 V cm⁻¹) by dividing the area into nine parts to ascertain the As movement (horizontally and vertically) after 72 h. Secondly, utilizing the optimal conditions for As mobility from the first part, to ascertain the effect of EK with Mott Dwarf Napier Grass (*Pennisetum purpureum* cv. Mott) for the co-treatment of mine tailings at an applied electric field of 0, 1, 2, and 4 V cm⁻¹ to examine the As accumulation in the plants and mine tailings after 15, 30, 45, 60, 75, 90, 105, and 120 d. The results showed that As had the highest mobility in agar in the spherical container using graphite electrodes with an applied electric field of 2 V cm⁻¹. The areas around the cathodes had lower As levels than that around the anode, with a net movement from cathode to anode areas both horizontally and vertically. For the co-treatment of mine tailings with EK and *P. purpureum* cv. Mott cultivation, the applied electric field provided better As treatment results than without an electric field, and the plants accumulated As in their roots to a greater extent than in the stem and leaves. The highest As absorption capability in the roots of *P. purpureum* cv. Mott (7.69 ± 0.16 mg kg⁻¹) was obtained after 90 d with an applied electric field of 1 V cm⁻¹.

Introduction

Heavy metal contamination is an important environmental issue, where in many countries around the world it is causing an increasing danger to ecosystems, including human life. The main cause of such contamination is frequently human activities created in response to the need to improve economic systems for better living. These processes, such as mining, tend to be focused on profit with deficiencies in management, and so result in heavy metal contamination in the environment (Xiao et al., 2017), in terms of the soil, surface water, groundwater, and the mine tailings that are stored in the tailing storage facility (TSF).

Mine tailings that are stored in the TSF are a by-product of mineral processing and are mostly stored in general, open storage ponds without any protection. This can result in the transport of small contaminated particles by wind and water, and in some cases, leakage of heavy metals into the groundwater when there is poor maintenance of the pond. Most mine tailings consist of small sand particles, unused minerals, and heavy metals (Santibañez et al., 2012). It has been reported that the release of toxic metals from contaminated mines around the world amounts to 10⁹ tons per year (Adiansyah et al., 2015). Currently, China has more than 8,000 mines owned by the state and 110,000 belonging to private companies, with a total accumulation of mine tailings between 2009–2014 of over 10⁹ tons (Pan et al., 2014).

In Thailand, heavy metal contamination occurs in many areas, such as arsenic (As) contamination at Ron Phibun District, Nakhon Si Thammarat Province, lead contamination in Klity Creek Thong Pha Phum District, Kanchanaburi, and cadmium contamination in the Mae Tao River Basin, Mae Sot District, Tak Province (Pollution Control Department, 2016). The most common type of pollutant that is considered to be harmful to living organisms is As, mainly caused by gold mining activities. These gold mining sites are associated with As contamination in agricultural areas around the mining area (Kumarathilaka et al., 2018). Gold mining in Thailand currently generates around 750,000 tons of mine tailings per year with a lifetime concession estimated as 19 million tons (Changul et al., 2009). In addition, the mine tailings produced from mining gold ore have As concentrations of more than 50 mg kg⁻¹ (Sampanpanish and Suwattiga, 2017). Note that As is an extremely toxic and dangerous element for living things, including human health. In general, As contamination in the soil and mine tailings can be treated in a variety of ways depending on technological differences, such as excavation, capping, solidification and stabilization, soil washing, vertical engineering barriers, phytoremediation, and electrokinetics (EK) (Khalid et al., 2017; Liu et al., 2018; Sampanpanish and Tantitheerasak, 2015).

In general, ex-situ technologies often have a high operation cost resulting in the need for improvement and development of in-situ technologies to be more efficient. One of the safe treatment technologies that does not destroy the topsoil and has a low cost (Khalid et al., 2017), is phytoremediation. This uses plants to treat and restore areas with soil contamination, including mine tailings (Wang et al., 2017). However, there are many plants that can absorb pollutants and accumulate them in different parts of the plant, which vary according to the increase in biomass (Afzal et al., 2014). This technology can be used to treat both organic and inorganic pollutants (Dary et al., 2010; Glick, 2010) that are in the soil, water, and air (Ali et al., 2013), such as benzene, ethylbenzene, xylene, radionuclides, and heavy metals (Sampanpanish, 2015). Although phytoremediation technology has many advantages, its principal limitation is it requires a long treatment period for a high efficiency. This has led to the development of other techniques to be used in conjunction with phytoremediation for a more effective treatment. One of those co-treatment technologies is EK.

The use of in-situ EK can remove As from the soil and contaminated mine tailings (Baek et al., 2009; Yuan and Chiang, 2007), is suitable for use in sites where the majority of the soil elements are clay and sand, and can be utilized to treat a wide area to reduce the extent of contamination (Suied et al., 2018; Weng et al., 2007). The principles of EK treatment technology are the use of a direct current electrical field with electrodes implanted into the contaminated area to induce an electric current flow through the ground particles between those electrodes and so cause movement of various substances (mostly charged) to the respective electrode. By using electric currents, water, ions, and small particles in the water between the particles of the earth move to the electrodes. In other words, it is the application of an electric potential through a series of cathodes and anodes, where the input electric field has a potential difference that will induce reactions in the soil and move the contaminants towards and concentrate at the cathode or anode. The electrode areas can then be subjected to other contamination treatments (Rosa et al., 2017).

This research used phytoremediation and EK together to treat As that had accumulated and contaminated mine tailings stored in a TSF within a gold mine. The aim was to reduce the toxicity value using *Pennisetum purpureum* cv. Mott, a biomass plant that has received attention for the production of renewable energy (Ana et al., 2017), and has a low cost of planting, a fast growth rate, high yield per 1,600 m², and is easy to maintain (Sarwanto and Tuswati, 2018).

Materials And Methods

Preparation of mine tailings and *P. purpureum* cv. Mott.

The mine tailings used in this study were taken from the TSF at a depth of 0–30 cm. Wood, stones, and unwanted plants were removed) and then it was assayed for basic physical and chemical characteristics, with the results summarized in Table 1.

The plant used in this study was *P. purpureum* cv. Mott, which was obtained from the Nakhon Ratchasima Animal Food Research and Development Center, Pak Chong Sub-district, Pak Chong District, Nakhon Ratchasima Province (Thailand). However, the plants used in the experiment come from stem cutting method. The plants were selected to be of the same age, size, and fresh (wet) weight. A total of plants (0.5 g plants/soil, volume adjustment 50 ml, limit of quantification As is 0.50 mg kg⁻¹) were examined prior to the experiment to determine their As level, with none being detected in the studied plants.

Experimental design

The experiment was divided into two parts. In the first part, the effect of EK on As mobility in agar medium was examined for different shaped containers (spherical or rectangular), electrodes (graphite or stainless steel grade 316L), and the applied electric field (0, 1, and 2 V cm⁻¹). The agar samples were collected for analysis after 72 h. The conditions that resulted in the highest As mobility were then selected for use in the second part, to test the ability of co-treatment of mine tailings with *P. purpureum* cv. Mott together with EK at different applied electric fields (0, 1, 2, and 4 V cm⁻¹) for 3 h d⁻¹, in terms of the As level in the plant tissues and mine tailings at 15, 30, 45, 60, 75, 90, 105, and 120 d. These two parts are outlined in more detail below.

Effect of different EK factors on As mobility in agar

The agar used in this study was food grade and had a resistance (R) of 56 Ω and no detectable As. The agar was prepared at 15 g l⁻¹ water containing 2.5 mM potassium nitrate and with or without an As source (Na₂HAsO₄ 7H₂O) at a concentration of 50 mg l⁻¹. The As-contaminated layer was poured to a depth of 10 cm (bottom layer), allowed to set and then the As-free agar (non-contaminated top layer) was poured on top to a depth of 10 cm from the contaminated layer into either a circular container of 35 × 22 cm or a rectangular container of 39 × 46 × 27 cm. In each case, the anode was placed in the middle and six cathodes were placed around the anode in a hexagon shape as shown in Fig. 1 (a) and (b). The electric field was applied at 0, 1, and 2 V cm⁻¹ for 72 h continuously, and then samples for analysis of the As level were collected from around each electrode.

Table 1
Physical properties and basic chemistry of the mine tailings used in the experiment

Property of the mine tailings	Method	Value
Sand: Silt: Clay	Hydrometer	83.8:15.2:1.0
pH	1:2 (v/v) soil: water mixture	7.68
Electrical conductivity ($\mu\text{S cm}^{-1}$)	1:2 (v/v) soil: water mixture	1,654
Oxidation reduction potential (mV)	1:2 (v/v) soil: water mixture	271.0
Cation exchange capacity ($\text{cmol}_c \text{ kg}^{-1}$)	Ammonium acetate saturation	3.0
Electrical resistance ($\text{k}\Omega$)	Wenner soil resistivity test 4-point	27
As (mg kg^{-1})	USEPA 3052	68.70

For collecting the agar samples, the sample collection area was divided into nine (one per electrode) as shown in Fig. 1 (c) and (d), and five samples were removed, each as a core, using a 2 cm diameter and length 25 cm HDPE pipe. Each sample core was divided into the two layers (top and bottom layer) for separate analysis and then the five top layers were pooled to form one, as were the five bottom layers. After that, the agar samples were extracted according to the USEPA method 3015A (USEPA, 2007a) by microwave digestion and then the total As content was analyzed by Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES).

Movement and accumulation of As from mine tailings into *P. purpureum* cv. Mott with EK co-treatment

The experimental setup was prepared by planting *P. purpureum* cv. Mott in an ex-situ greenhouse in cylindrical pots of 30-cm height and 25-cm diameter with one plant per pot and As-contaminated mine tailings (68.70 mg kg^{-1}) at 11 kg per pot. In each experiment, seven stainless steel (316L) electrodes were used per one pot with a central anode and six cathodes inserted around the edge of the pot in a hexagonal shape at appropriate distances, as shown in Fig. 2. Thereafter, 200 ml of water was added to each pot every 2 d to control the humidity—the mine tailings had an initial moisture content of 15% (w/w)—before applying the electric field for 3 h d^{-1} (09.00 to 12.00 h). Note that no fertilizer was administered for the duration of the experiment. Providing 3 hours of electricity per day does not effect on biomass and appropriate for photosynthesis. The experiment was performed in four sets of an applied electric field of 0 (control), 1, 2, and 4 V cm^{-1} , respectively. The plant specimens and mine tailings were collected after 15, 30, 45, 60, 75, 90, 105, and 120 d for analysis of the As levels.

For sampling the mine tailings, they were divided into two parts. The first part was dried at room temperature in order to analyze the properties of mine tailings, while the second part was baked at 105°C for 24–48 h and then extracted according to the USEPA method 3051A (USEPA, 2007b) by microwave digestion and then analyzed for total As using ICP-OES.

Plant sampling was performed by separating the mine tailings from the plant roots by rinsing with tap water three to four times and then with de-ionized water. The plants were then separated into aboveground (stem and leaves) and underground (root) parts. The fresh weight of each plant part was measured and then baked at 105°C for 24–48 h. The dried specimens were ground separately and extracted according to the USEPA method 3052 (USEPA, 1996) using microwave digestion and then analyzed for total As using ICP-OES.

Data analysis

Analysis of the variance of the data was performed using ANOVA to find the F-value at the 95% confidence level using the SPSS (Statistical Package for the Social Sciences) program. The significance of differences between means were evaluated using Duncan's New Multiple Range Test (DMRT).

Results And Discussion

Effect of EK under different conditions on the As mobility in agar

From the results of the As mobility in the agar medium with EK, the As concentration level in each sampling area was significantly different ($P \leq 0.05$). The As tended to move towards the anode (area 5) away from the cathodes (areas 1–4 and 6–9), resulting in the highest As concentration in the anode area (Fig. 3). Thus, the EK energy caused the As to move from the cathode to the anode horizontally, which reflects the negative charge of the arsenate ion (AsO_4^{3-}). This is consistent with previous studies that reported the electric field stimulated As compounds to break down into negatively charged ions and to move towards the anode by electromigration (Virkutyte et al., 2002; Putra et al., 2013). Moreover, As accumulation was also observed in the top 10 cm, which is the part of the agar that was not contaminated with As, and so the EK caused As to move from the cathode to the anode vertically as well as horizontally.

With respect to the effect of the type of container on the As mobility, at an applied electric field of 1 V cm^{-1} the spherical cylinders with graphite or stainless steel electrodes had the highest As concentration at the anode, at 84.14 ± 1.02 and $68.39 \pm 0.11 \text{ mg L}^{-1}$, respectively, (Fig. 3a and 3b) compared to the hexagonal setup (rectangular container) with graphite or stainless steel electrodes at 38.84 ± 0.23 and $35.38 \pm 0.29 \text{ mg L}^{-1}$, respectively, (Fig. 3c and 3d). This is because the spherical containers had a greater area influenced by the electric field, since the position of the electrodes covers a larger area than in the rectangular container. This is consistent with previous studies that reported the motion of electric fields from the hexagonal electrodes resulted in a higher amount of As movement from each area to the anode when it is positioned at the center of the container and the cathodes are placed in a shape similar to the spherical container (Kim et al., 2012; Isosaari and Sillanpaa, 2012; Jeon et al., 2015). For the rectangular container, some areas were not influenced by the electric field. As a result, the As showed almost no movement.

When the applied electric field was increased to 2 V cm^{-1} with stainless steel electrodes, the rectangular container had a numerically, but not significantly, higher As concentration at the anode ($96.99 \pm 0.89 \text{ mg L}^{-1}$; Fig. 3e) than in the spherical container ($96.10 \pm 1.20 \text{ mg L}^{-1}$; Fig. 3f). However, with graphite electrodes the rectangular container had the highest As concentration at the anode ($114.49 \pm 0.91 \text{ mg L}^{-1}$; Fig. 3g) although this was not statistically different from that for the spherical container with graphite electrodes ($114.06 \pm 2.13 \text{ mg L}^{-1}$; Fig. 3h). This is because increasing the applied electric field from 1 V cm^{-1} to 2 V cm^{-1} resulted in better As movement, but the agar, which has an electrical resistance of $0 \text{ k}\Omega$, undergoes a faster rate of water hydrolysis at the anode, causing the agar medium at the graphite electrodes to begin to break down from 50 h onwards and resulting in less to no As movement. That the As movement to the anode varied directly with the applied electric field is in accord with Tang et al. (2018). However, there were resistance factors in the medium. The electrode also affected the As movement, where with an applied electric field of 1 or 2 V cm^{-1} using graphite electrodes in the square or circular container had a significantly different As concentration at the anode to that with stainless steel electrodes at the corresponding applied electrical field and container shape with anodal As concentrations of 38.84 ± 0.23 , 84.14 ± 1.02 , 114.49 ± 0.91 and $114.06 \pm 2.13 \text{ mg L}^{-1}$ respectively.

Effect of the applied EK on the As mobility in mine tailings under different conditions

The As movement in mine tailings with an applied electric field of 1 and 2 V cm^{-1} in a spherical container model with hexagonal-sided stainless steel electrodes was examined after 3, 5, and 7 d of treatment at the cathodes, anode, and in-between areas (Table 2). With an applied electric field of 1 V cm^{-1} , the concentration of As in all three areas were not statistically different at each time point and between time points. Thus, an applied electric field of 1 V cm^{-1} was insufficient to cause significant As movement. The mine tailings had a resistance value of $21 \text{ k}\Omega$, and so the electric field had a low influence on As motion. This is consistent with the study of Kim et al. (2012), who reported that a low electric field or short duration did not cause any significant As movement in soils with a high sand particle composition.

Table 2

The As concentration in mine tailings after an applied electric field of 1 and 2 V cm⁻¹ for 3, 5, or 7 d using stainless steel 316 L electrodes in a spherical container.

Electric field	Trial period (d)	As concentration (mg kg ⁻¹)		
		Cathode	Anode	In-between
1 V cm ⁻¹	3	61.71 ± 0.91 ^a	62.33 ± 1.66 ^a	62.17 ± 1.05 ^a
	5	60.73 ± 1.40 ^a	58.51 ± 0.78 ^a	61.76 ± 1.07 ^a
	7	60.85 ± 2.19 ^a	57.77 ± 0.68 ^a	61.02 ± 2.74 ^a
2 V cm ⁻¹	3	64.15 ± 1.63 ^a	60.92 ± 2.13 ^a	60.92 ± 2.13 ^a
	5	59.04 ± 0.61 ^a	57.52 ± 1.16 ^a	63.07 ± 1.60 ^b
	7	61.33 ± 1.85 ^b	53.60 ± 0.53 ^a	62.43 ± 0.74 ^b

Data are shown as the mean ± 1 SD, derived from 3 repeats. Means with a different letter are significantly different (p < 0.05; DMRT).

For an applied electric field of 2 V cm⁻¹ (Table 2), the As concentration in the anode and cathode areas were not statistically different after 3 and 5 d respectively, but the area in-between the cathode and anode had a significantly higher As level after 5 and 7 d of treatment, indicating that by 5 d the As had moved from the anode and cathode areas to the area in-between them, which may be because the As is then in a water insoluble form.

When considering the various mechanisms occurring at the same time that cause As to move in opposite directions, electro-osmosis will lead the water to the cathode and electromigration will carry As, in the form of negative ions, to the anode which is in the opposite direction and so resulting in the higher As accumulation in the area in-between the anode and cathodes. This is consistent with Isosaari and Sillanpaa (2012), who reported that the use of EK to treat As-contaminated soil can also cause As deposition between the polar regions, and gives a different effect to that for other heavy metals that are mostly in the form of positive ions. In addition, hydrogen ions (H⁺) and hydroxide ions (OH⁻) formed by hydrolysis reactions can also cause a marked change in the pH of the solution in each mine tailing.

Preliminary characteristics and factors affecting the absorption and accumulation of As in *P. purpureum* cv. Mott

The relation of electric current, humidity, and the relative growth rate (RGR).

With an applied electric field of 1, 2, or 4 V cm⁻¹, the electrical current over time in the experimental set up ranged from of 1.10–1.30, 1.63–1.83, and 2.14–2.61 A, respectively, (Fig. 4). The treatment of As-contaminated mine tailings with electricity can be used as a measure of the resistance of the mine tailings, where the initial resistance value of these mine tailings (after drying) was 27 kΩ. However, in general environmental conditions, there are often many factors that affect resistance, such as the humidity, porosity, temperature, and the concentration of other metal ions in solution and not in solution. However, the factor that has a significant effect on the current is the humidity, as can be seen that the current will always vary directly with the moisture content in the mine tailings (Fig. 4), particularly at a low applied electric field (1 and 2 V cm⁻¹). On the other hand, at a high applied electric field (4 V cm⁻¹), the electric current and moisture content in the mine tailings tends to decrease due to the intense hydrolysis reaction. Furthermore, the plant roots can also increase the porosity of the mine tailings, which will create gaps between the mine tailing particles and so increase the water retention and ion mobility (Sanchez et al., 2018).

In addition, plants also increase the ionic concentration of mine tailings around their roots due to the release of various charged compounds to help in the process of nutrient absorption for increased plant growth (LeFevre et al., 2013; Vassilev et al., 2004). The reduction in the current in an applied electric field of 4 V cm⁻¹ over time was correlated with the reduced amount of ions in the plant roots. This is because under this condition the plants have a very low to negative RGR after 120 d (Fig. 5), causing the plant's nutrient

absorption to stop from the root area. Moreover, there may also be other processes that help increase the electrical resistance of mine tailings (Cang et al., 2011; Chirakkara et al., 2015; Risco et al., 2015; Risco et al., 2016).

Electrical conductivity (EC) of mine tailings.

Tables 3 and 4 summarize the changes in the EC of the mine tailing samples at the anode and cathode areas at different applied electric fields [0 (C), 1, 2, and 4 V cm⁻¹]. The trend in the EC of the mine tailings at the anode and the cathode decreased with increasing time of applied electrical fields. The conductivity in the anode area after 15 d at an applied electrical field of 0, 1, 2, or 4 V cm⁻¹ increased from 578.42 ± 61.10 µS cm⁻¹ at 0 V cm⁻¹ to 2384.73 ± 110.40 µS cm⁻¹ at 4 V cm⁻¹ (Table 3), while at the cathode it increased from 1635.45 ± 34.78 to 854.46 ± 94.31 µS cm⁻¹, respectively, (Table 4). After 120 d, the EC of the mine tailings had decreased from 1543.21 ± 61.26 µS cm⁻¹ at 0 V cm⁻¹ to 933.07 ± 73.85 µS cm⁻¹ at 4 V cm⁻¹ in the anode area (Table 3), and from 1635.45 ± 34.78 to 209.00 ± 26.27 µS cm⁻¹, respectively, in the cathode area (Table 4).

Thus, a high applied electric field (4 V cm⁻¹) had a faster rate of the hydrolysis reaction at the electrode than at a low applied electric field (1 and 2 V cm⁻¹) and the anode had a higher value than the cathode. In addition, the conductivity of H⁺ produced by the reaction at anode area is slightly higher than that of OH⁻ from the cathode (35.0 compared to 19.8 mSm² mol⁻¹) and the other anions, such as chloride (Cl⁻), sulfate (SO₄²⁻), and nitrate (NO₃⁻) (Cameselle et al., 2019). However, spatial changes in the water content between mine tailings particles can result in different conductivity values. Here, the ion movement of the various elements in the mine tailings in solution have a good mobility when the water content in the medium or mine tailings is high (high humidity). The EC always varied directly with the amount of H⁺ in an electric field. In addition, the EC was very high at the anode during the first month of the experiment. This may be due to the influence of organic compounds released from the roots (root exudates) while the plant is alive and has a high RGR together with the acidification at the anode region caused by electrolyte-osmosis and electric flux, which results in various elements, both those that the plants can and cannot solvate and use as food, to accumulate. When the duration of the experiment was increased to 120 d, changes in the pH and some precipitation of ions around the electrode resulted in a reduced EC over time (Lopez-Vizcaino et al., 2017).

Table 3
The EC of mine tailings in the anode area

Trial period (d)	EC (µS cm ⁻¹)			
	0 V cm ⁻¹	1 V cm ⁻¹	2 V cm ⁻¹	4 V cm ⁻¹
15	1578.43 ± 61.10 ^{ef}	1868.69 ± 55.27 ^c	2040.02 ± 62.95 ^b	2384.74 ± 110.40 ^a
30	1569.53 ± 66.13 ^{ef}	1665.37 ± 64.99 ^{de}	1793.73 ± 84.53 ^c	2059.50 ± 55.53 ^b
45	1567.60 ± 63.31 ^{ef}	1513.85 ± 50.28 ^{fg}	1532.45 ± 75.97 ^{fg}	1767.13 ± 43.62 ^{cd}
60	1561.77 ± 63.71 ^{ef}	1398.42 ± 84.96 ^h	1235.73 ± 54.48 ⁱ	1437.77 ± 23.59 ^{gh}
75	1558.48 ± 63.18 ^{efg}	1198.69 ± 69.06 ^{ij}	1122.29 ± 160.17 ^{jk}	1089.50 ± 15.90 ^{kl}
90	1542.21 ± 67.90 ^{efg}	1053.74 ± 34.60 ^{klm}	1023.61 ± 34.94 ^{klm}	1016.43 ± 66.61 ^{klm}
105	1546.91 ± 65.69 ^{efg}	1048.15 ± 42.48 ^{klm}	1018.32 ± 64.39 ^{klm}	972.68 ± 7.22 ^{lm}
120	1543.21 ± 61.27 ^{efg}	1004.95 ± 34.21 ^{klm}	972.39 ± 7.46 ^{lm}	933.07 ± 73.86 ^m
Data are shown as the mean ± 1 SD, derived from 3 repeats. Means with a different letter are significantly different (p < 0.05; DMRT).				

Changes in the pH of mine tailings

The pH of the mine tailings after 15, 30, 45, 60, 75, 90, 105, and 120 d of treatment at the anode and cathode are summarized in Tables 5 and 6. At an applied electric field of 1, 2, and 4 V cm⁻¹, the mine tailings had a significantly different pH value at the anode compared to that at the cathode, where the pH at the anode decreased with increasing duration of the experiment from 7.55 ± 0.09, 7.35 ± 0.05, and 7.15 ± 0.03 after 15 d at 1, 2, and 4 V cm⁻¹, respectively, to 5.21 ± 0.03, 4.42 ± 0.04, and 2.93 ± 0.05, respectively, after

120 d. In contrast, the pH at the cathodes increased over treatment time, rising from 7.88 ± 0.01 , 7.86 ± 0.09 , and 7.95 ± 0.03 , respectively, after 15 d to 9.32 ± 0.07 , 9.63 ± 0.05 , and 10.15 ± 0.22 , respectively, after 120 d.

Table 4
The EC of mine tailings in the cathode area

Trial period (d)	EC ($\mu\text{S cm}^{-1}$)			
	0 V cm^{-1}	1 V cm^{-1}	2 V cm^{-1}	4 V cm^{-1}
15	1635.45 ± 34.79^a	1498.63 ± 64.33^b	1187.92 ± 68.93^c	854.46 ± 94.31^{fg}
30	1632.11 ± 32.21^a	1212.32 ± 97.70^c	983.38 ± 14.52^e	672.18 ± 103.49^{hi}
45	1628.34 ± 30.30^a	1086.84 ± 22.89^d	868.64 ± 44.31^{fg}	573.16 ± 37.10^{ij}
60	1624.26 ± 31.66^a	923.58 ± 20.69^{ef}	775.89 ± 21.79^{gh}	468.90 ± 77.35^{kl}
75	1620.85 ± 29.51^a	826.54 ± 21.03^{fg}	672.04 ± 41.28^{hi}	361.48 ± 32.60^{lm}
90	1620.06 ± 28.63^a	654.81 ± 28.16^i	636.51 ± 20.29^{ij}	316.11 ± 10.29^{mn}
105	1618.31 ± 28.21^a	582.75 ± 70.31^{ij}	534.54 ± 38.87^{jk}	237.16 ± 23.50^{no}
120	1614.21 ± 31.50^a	455.07 ± 36.13^{kl}	418.17 ± 34.28^{lm}	209.00 ± 26.27^o
Data are shown as the mean $\pm$ 1 SD, derived from 3 repeats. Means with a different letter are significantly different.				

Thus, increasing the applied electric field or time decreased the pH at the anode and increased it at the cathode area. This is due to the hydrolysis of water at both electrodes, where at the anode the oxidation reaction ($2\text{H}_2\text{O} \rightarrow \text{O}_{2(\text{gas})} + 4\text{H}^+ + 4\text{e}^-$) resulted in more H^+ (lower pH) as the electric field increased, whereas at the cathode the reduction reaction ($2\text{H}_2\text{O} + 2\text{e}^- \rightarrow 2\text{OH}^- + \text{H}_{2(\text{gas})}$) resulted in a more OH^- (higher pH). Since H^+ are able to move faster than the larger OH^- , it resulted in the expansion of acidic areas compared to the alkaline areas.

In addition, OH^- can precipitate as various metal hydroxides at the negative pole (Cameselle et al., 2013; Cang et al., 2012; Paz-Garcia et al., 2012; Peng et al., 2012). Finally, the reactions and processes of plant roots may also increase the acidity of mine tailings around the roots, due to the organic compounds that are released through the roots of plants, such as sugars, amino acids, and organic acids, including citric, oxalic, malic, and acetic acids (Bais et al., 2006; Carvalhais et al., 2011). As mentioned, it is a common mechanism that plants use for solubilizing, sequestering and obtaining nutrients (Wang et al., 2006).

Table 5
The pH of mine tailings in the anode area

Trial period (d)	pH			
	0 V cm ⁻¹	1 V cm ⁻¹	2 V cm ⁻¹	4 V cm ⁻¹
15	7.68 ± 0.02 ^a	7.56 ± 0.09 ^a	7.35 ± 0.05 ^b	7.16 ± 0.04 ^c
30	7.67 ± 0.02 ^a	6.73 ± 0.04 ^d	6.15 ± 0.05 ^{fg}	5.71 ± 0.07 ^h
45	7.68 ± 0.02 ^a	6.41 ± 0.14 ^e	5.73 ± 0.35 ^h	5.33 ± 0.04 ^j
60	7.66 ± 0.02 ^a	6.21 ± 0.03 ^f	5.52 ± 0.05 ⁱ	4.92 ± 0.04 ^l
75	7.67 ± 0.01 ^a	6.07 ± 0.03 ^g	5.20 ± 0.04 ^k	4.49 ± 0.04 ⁿ
90	7.65 ± 0.01 ^a	5.83 ± 0.03 ^h	4.96 ± 0.08 ^l	3.90 ± 0.01 ^o
105	7.67 ± 0.02 ^a	5.55 ± 0.05 ⁱ	4.71 ± 0.03 ^m	3.45 ± 0.04 ^p
120	7.67 ± 0.01 ^a	5.21 ± 0.04 ^k	4.42 ± 0.04 ⁿ	2.94 ± 0.05 ^q
Data are shown as the mean ± 1 SD, derived from 3 repeats. Means with a different letter are significantly different (p < 0.05; DMRT).				

Effect of EK on the absorption and accumulation of As in *P. purpureum* cv. Mott

The results of the As-contaminated mine tailing treatment with *P. purpureum* cv. Mott together with the use of EK are summarized in Fig. 6. The As accumulation in the underground plant parts (roots) in all experiment sets was not significantly different over the first 30 d. However, the As levels became significantly different after 60 d, especially in the experimental set ups with the highest As concentration and accumulation in the roots. The highest As concentration and accumulation was found at an applied electric field of 1 V cm⁻¹ after 90 d (7.69 ± 0.16 mg kg⁻¹), followed by that after 105 d (6.91 ± 0.09 mg kg⁻¹), 120 d (6.69 ± 0.13 mg kg⁻¹), and 2 V cm⁻¹ at 120 d (6.48 ± 0.21 mg kg⁻¹).

Table 6
The pH of mine tailings in the cathode area

Trial period (d)	pH			
	0 V cm ⁻¹	1 V cm ⁻¹	2 V cm ⁻¹	4 V cm ⁻¹
15	7.65 ± 0.10 ^l	7.89 ± 0.02 ^{kl}	7.86 ± 0.09 ^{kl}	7.95 ± 0.03 ^k
30	7.65 ± 0.09 ^l	8.00 ± 0.06 ^k	8.00 ± 0.06 ^k	8.03 ± 0.27 ^k
45	7.64 ± 0.07 ^l	8.75 ± 0.26 ^j	9.00 ± 0.12 ⁱ	9.03 ± 0.04 ^{hi}
60	7.65 ± 0.04 ^l	9.13 ± 0.17 ^{ghi}	9.16 ± 0.21 ^{fghi}	9.38 ± 0.31 ^{efg}
75	7.65 ± 0.02 ^l	9.21 ± 0.09 ^{fghi}	9.23 ± 0.07 ^{fghi}	9.50 ± 0.07 ^{de}
90	7.66 ± 0.03 ^l	9.20 ± 0.13 ^{fghi}	9.22 ± 0.10 ^{fghi}	9.77 ± 0.31 ^{bc}
105	7.68 ± 0.01 ^l	9.28 ± 0.14 ^{efgh}	9.40 ± 0.07 ^{def}	9.92 ± 0.04 ^b
120	7.67 ± 0.02 ^l	9.32 ± 0.08 ^{efg}	9.63 ± 0.05 ^{cd}	10.15 ± 0.23 ^a
Data are shown as the mean ± 1 SD, derived from 3 repeats. Means with a different letter are significantly different (p < 0.05; DMRT).				

For the aboveground plant parts (stem and leaves), there was very little As absorption and accumulation over the first 15 to 60 d, and were mostly not significantly different between treatments and times. However, after 75 d, the As absorption and accumulation in the

stem and leaves was ranked (lowest to highest) as 2 V cm^{-1} after 75, 90, and 105 d, 4 V cm^{-1} after 60, 75, and 120 d, and then 1 V cm^{-1} after 105 and 90 d, at 3.12 ± 0.16 , 3.10 ± 0.06 , 2.91 ± 0.51 , 2.85 ± 0.14 , 2.79 ± 0.28 , 2.73 ± 0.14 , 2.63 ± 0.73 , and $2.59 \pm 0.04 \text{ mg kg}^{-1}$, respectively. Thus, the As accumulation level in the stem and leaves of the plants were significantly different between different applied electric fields and durations (Fig. 6).

Considering the concentration of As in mine tailings at different applied electric fields and durations (Fig. 7), the amount of As accumulated in the mine tailings with an applied electric field of 1 and 2 V cm^{-1} after 120 d were 60.42 ± 1.06 and $60.41 \pm 0.48 \text{ mg kg}^{-1}$, respectively, which are similar but significantly lower than in the other conditions (applied electric field and duration). The second largest reduction in the As concentration was with an applied electric field of 1 or 2 cm^{-1} after 90 d. The As levels found in the shoot and leaves were consistent with a previous study that co-treated As-contaminated (20 mg kg^{-1}) soil from paddy field areas with plants and EK (Mao et al., 2016). In that study, a higher level of As accumulation was found in the plants with an applied electric field than without, and the As accumulation was greater in the roots than in the stem and leaves.

The relationship between the pH of the mine tailings at the anode (Fig. 7) and the amount of As accumulated in the plants revealed that an applied electric field of 1 and 2 V cm^{-1} for 90–120 d gave noticeably better As accumulation, especially after 90 d, that had the highest As concentration. A pH range of 5.20–5.80 in the mine tailings provided the best As absorption by the plants since the As in mine tailings is often surrounded by iron and manganese oxides that reduce the pH of mine tailings and may increase the solubility of As (Han et al., 2004). In addition, the pH level does not reach a level that will inactivate the microbial system in the rhizosphere of plant roots. The organic acids produced from this group of microbes increase the absorption of metals catalyzing redox reactions and organic ligands reaction (Thangavel and Subbhuraam, 2004). In accordance, the acidic conditions caused by microbes around the plant roots were reported to increase the efficacy of As absorption and accumulation in plants (Gonzaga et al., 2012).

However, the As accumulation level in the plant tissues is still very small compared to the As content in the mine tailings. The mechanism of induction of As accumulation and transportation in plants depends on many factors, such as the type of As, As concentration, the pH of the mine tailings, and plant species. For example, many plants absorb As in the form of arsenate by the phosphate transporter protein group on the membrane cells of the roots. However, in low oxygen conditions, such as in flooded areas, As is in the form of arsenite, which is easier for plants to absorb than arsenate via the nodulin-26-like intrinsic membrane proteins, which are proteins in the aquaporin group (Zhao et al., 2009).

Conclusion

The factors that affect the movement of As in agar medium using EK were concluded as a small but similar As accumulation at the cathode areas, but a higher level at the anode, resulting from EK moving the As from the cathode to the anode horizontally and vertically. The movement of As in the agar medium was greater in a spherical container and optimal with a graphite electrode at an applied electrical field of 2 V cm^{-1} . For the effect of EK on As movement in mine tailings (without plants), an applied electric field of 2 V cm^{-1} for 5 d induced the highest As movement from the cathode to the anode. However, graphite has a high cost per rod and it is fragile and is easily broken, and so it is not suitable for use in actual mine tailing areas. Thus, alternative materials that can provide a good electric field and are resistant to corrosion from the acid conditions at the anode should be selected, such as stainless steel, nickel alloys, aluminum alloy, and platinum.

For the co-treatment of the As-contaminated mine tailings with *P. purpureum* cv. Mott phytoremediation and EK, all the evaluated applied electric fields gave better As treatment results than without an applied electric field, and *P. purpureum* cv. Mott had higher As accumulation levels in the roots than aboveground parts (stem and leaves). The highest As content in the roots ($7.69 \pm 0.16 \text{ mg kg}^{-1}$) was obtained with 1 V cm^{-1} for 90 d. When considering the As accumulated per biomass, the co-treatment with *P. purpureum* cv. Mott and an applied electric field of 1 or 2 V cm^{-1} for 120 d gave the highest value. With respect to other related factors, such as the RGR, EC, anode erosion, and the pH, a 120-d duration was not suitable because of the drastic change in the conditions of the mine tailings, especially the pH, and its EC was too low. Therefore, if this is applied in agricultural soil, it will result in a loss of soil fertility. In addition, suitable plants for use in the EK treatment should be biennial plants with a high biomass, such as plants in the same family as the turnips (*Brassica juncea*), or plants with a high absorption capability and resistance to As, such as plants in the same family as ferns. Fern evolution has happened in parallel to angiosperm evolution. These plants were exposed to extreme environmental conditions including nutrient-poor environments, contaminated soil with toxic material, extremes of temperature. Moreover, ferns could be sources of stress-tolerance genes for phytotechnology. This has caused to biodiversity of fern for natural adaptations on the environmental

condition. From this study, the suitable electric field for As treatment and plant growth was 1 V cm^{-1} . However, the electric field should not be applied continuously or for too long a period per day, but should be reduced in order not adversely affect the plant growth and prevent the destruction of plant root cells.

Declarations

Ethics approval and consent to participate

Not applicable in this section.

Consent for publication

Not applicable in this section.

Availability of data and materials

Not applicable in this section.

Competing interests

The authors declare that they have no competing interests.

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Authors' contributions

This article is an original research paper. All authors participated in all experiments; PS designed the experiments; KW undertook the data analysis; PS and KW drafted the manuscript; PS advised on data analysis; interpretation and provided the original idea for the paper, commented on the data analysis, and edited the manuscript. All authors reviewed the manuscript and helped with data interpretation and gave final approval of the version to be submitted to this journal and agree to be held accountable for the work performed therein.

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Conflict of Interest

The authors declare that no conflicts of interest and confirm that the manuscript has been submitted solely to this journal and is not published, in press, or submitted elsewhere.

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Figures

Figure 1

Characteristics of the experimental container and agar sampling points. Schematic diagram showing the (a) rectangular and (b) circular container, and the sampling points for the (c) rectangular and (d) spherical container.

Figure 2

Schematic diagram showing the experimental setup of *P. purpureum* cv. Mott cultivation in mine tailings with the use of EK showing the (a) side and (b) top view of the experimental setup.

Figure 3

The As distribution in the upper (Top) and lower (Bottom) a gar layers at the nine areas around the electrodes (see Fig. 2) with an applied electric field of (a – d) 1 V cm⁻¹ or (e – h) 2 V cm⁻¹ in a (a, b, f, h) circular or (c – e, g) square container with (a, c, g, h) graphite or (b, d, e, f) stainless steel electrodes. Data are shown as the mean $\pm$ 1 SD, derived from 3 repeats. Means with a different letter are significantly different ($P < 0.05$; DMRT).

Figure 4

The relation of electric current (A) to the moisture content (%) in mine tailings over time at different applied electric fields and durations. Data are shown as the mean $\pm$ 1SD, derived from 1 independent repeats.

Figure 5

The RGR of *P. purpureum* cv. Mott at different applied electric fields and durations. Data are shown as the mean $\pm$ 1SD, derived from 3 independent repeats. Show the SD as error bars.

Figure 6

The As concentration accumulated in the roots (belowground) and stem and leaves (aboveground) of *P. purpureum* cv. Mott after different treatments (0 [C], 1, 2 and 4 V cm⁻¹) and times (15 – 120 d). Data are shown as the mean $\pm$ 1 SD, derived from 3 repeats. Means with a different letter are significantly different (p < 0.05; DMRT).

Figure 7

The As concentration in mine tailings at different applied electric fields and durations. Data are shown as the mean $\pm$ 1 SD, derived from 3 repeats. Means with a different letter are significantly different (p < 0.05; DMRT).