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Research Paper

Progress and Challenges of Biological Leaching of Heavy Metal in Coal Ash from a Power Plant

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Abstract

Bioleaching is a technique for reducing the heavy metal content of coal ash by using bacteria, fungi, or yeast. Previous studies in heavy metal bioleaching of coal ash discussed the factors affecting the process, but as yet there is little information on the challenges of using microorganisms. Therefore, this study aimed to obtain comprehensive information regarding the use of microorganisms in heavy metal bioleaching. Heavy metal concentrations in coal ash are low, and the metals are diverse. The components of coal ash are complexes that cannot leach certain heavy metals according to previous studies. These low concentrations and complex components make it difficult to investigate the bioleaching mechanism. The combination of biological and chemical interactions involves various components in this system. The high concentration of iron and heavy metal leached could be toxic for microorganisms. The process is influenced by several factors, such as particle size, pH, and pulp density. Most heavy metal bioleaching studies on coal ash have been conducted on a small scale to control conditions affecting the process. Bioleaching kinetics in coal is a liquid-solid reaction that can be represented by the shrinking core model, which was mainly used in this study.

Keywords: *bioleaching; coal ash; heavy metals; microorganism; power plant.*

Introduction

Coal ash causes environmental pollution in the air, water, and soil due to toxic elements, such as Cr, As, Pb, Co, Ni, Zn, Se, and Cu [1-5]. In India, Fe, Cr, Ni, Mn, and Pb concentrations have been found in groundwater near coal ash dumping sites, with concentrations exceeding the WHO quality standard [1,3]. Furthermore, Cu and Cr have been found in soil and impact soil quality around Serbia's power plants [2]. In Indonesia, The Jakarta Post [6] has reported that fly and bottom ashes were found illegally dumped by companies and caused environmental problems around a landfill area in Kalimantan due to poor waste management.

Metal extraction from fly ash is widely carried out in the industrial sector, primarily through chemical methods [7]. However, a biological extraction method, called bioleaching, separates metals from solid materials. Bioleaching using microorganisms has a leaching efficiency of 6 to 100%, depending on the type of metal and the factors affecting the activity (temperature, pH, nutrition, amount of microorganism biomass, organic acid, etc.) [8-11]. Several factors affect the biological leaching process. For example, microbial biomass and coal ash pulp density influence the bioleaching efficiency of Zn and Cu [7]. Organic acids are essential leaching agents when using fungi and bacteria in the biological process. The most investigated acids are tartaric, citric, gluconic, and oxalic acid [8]. Microorganisms produce gluconic and other organic acids that can increase the effectiveness of leaching rare earth elements (REE) from waste. Meanwhile, the production of these acids is directly proportional to the bioleaching efficiency [12].

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The bioleaching of heavy metals (Cr and Zn) from coal ash by *Acidithiobacillus* sp. obtained 56.38 to 85.01% efficiency. However, this study did not look at the effect of pulp density and kinetics [8]. A study on the bioleaching of heavy metals (Cu and Pb) in coal ash using yeast obtained 40.7 to 42.4% bioleaching efficiency but did not test the factors affecting the mechanism (e.g., pH, temperature, pulp density, microbial biomass, organic acid) [9]. Bioleaching studies using fungi also showed promising results. Reference [10] used *Aspergillus niger* in the bioleaching of metals (Ti, Zr, Ga, Ba, and Sr) from coal fly ash. One-step bioleaching and chemical hydrothermal methods enabled the extraction of 35 to 86.20% of metals. The use of a consortium of indigenous bacteria performed better than the exogenous candidates [13]. Bacteria, fungi, and yeast have been used in the biological leaching of heavy metals from waste and other solid materials [14-16]. However, few studies have been conducted on bioleaching heavy metals in coal ash from power plants [9,11,17-21]. The present study reviewed bioleaching of heavy metals in coal ash from power plants. The progress and challenges in heavy metal bioleaching from coal ash using microorganisms in several aspects are discussed in this paper.

The Coal Ash Generation

Coal remains the primary source of energy in the world. In 2021, the world's coal consumption was 5,362,568,071.08 tons of coal [22, 23]. China, India, and United States are the largest consumers of coal [22]. The production of coal is 7,575 Mt which is equivalent to 7,575,000,000 tons of coal and China was the major producer of coal in 2020 [22]. However, the use of coal creates solid waste (coal ash). The solid waste produced from the combustion of coal is about 5 to 20% of the coal as raw material [24]. Based on that assumption, about 268 million to 1,072 million tons of coal ash was produced in the year 2020.

Challenges of Heavy Metal Bioleaching on Coal Ash

Heavy Metals in Coal Ash

The components of coal ash from some of Asian countries in Table 1 show that bottom and fly ash consist of SiO_2, MgO, Fe_2O_3, Al_2O_3, CaO, K_2O, SO_3, Na_2O, and LOI, with Al_2O_3, SiO_2, and Fe_2O_3 (5.6-61.9 %) as the main constituents. The percentage of each element is different in each region for fly ash and bottom ash. Loss of ignition (LOI) is the main component in the form of metal oxides with varying unburnt carbon content [24]. Studies about coal ash components have shown similarities in the content of coal ash. However, in this result, low concentrations of heavy metals could not be determined.

Table 1 Components of coal ash from Some of Asian Countries.

Component (%)	Fly Ash			
	Indonesia[a]	**Malaysia**[b]	**India**[c]	**China**[d]
SiO_2	17.78-91.78	47.60-58.20	59.85-60.57	47.9-63.49
Al_2O_3	0.30-74.72	23.80-24.67	20.86-26.92	20.54-36.6
Fe_2O_3	0.51-29.33	3.86-7.40	5.01-9.15	5.03-5.26
CaO	0.12-27.90	4.76-10.70	1.43-2.00	3.35-5.44
MgO	0.04-11.74	0.71	0.83-1.40	1.30-1.35
SO_3	0.03-25.32	0.73	0.10-0.22	-
Na_2O	0.01-18.67	0.30-2.10	0.12-0.23	1.01-1.26
K_2O	0.02-10.48	1.40-2.73	1.31-2.26	0.92-1.05
LOI	0.01-39.33	0.43-3.79	1.77-1.82	1.70-4.55
Component (%)	**Bottom Ash**			
	Indonesia[a]	**Malaysia**[b]	**India**[c]	**China**[d]
SiO_2	0.28-89.40	45.3-52.50	56.40-56.44	51.30-57.85
Al_2O_3	0.25-31.20	17.65-18.10	29.13-29.24	17.56-32.10
Fe_2O_3	0.08-36.12	8.30-19.84	8.31-8.44	4.30-7.46
CaO	0.12-60.41	4.72-8.70	0.75-0.78	5.60-10.09
MgO	0.04-8.99	0.58-0.69	0.40-0.42	1.10-1.52
SO_3	0.01-24.11	0.30-0.84	0.21-0.24	0.50
Na_2O	0.01-22.12	0.16	0.07-0.09	0.60
K_2O	0.01-8.76	0.83-2.48	1.27-1.29	1.91
LOI	0.01-89.57	0.1-4.01	0.87-0.89	-

Source: [a][25] [b][26-29] [c][30-32] [d][33-36]

Determination of heavy metals was conducted by measuring the total concentration. In Table 2, fly ash contains metals such as Co, Pb, Cr, Mn, Zn, Cu, and Hg in low concentrations (0.1-9.41 mg/kg). It had more Mn, Zn, and Cr concentrations than the other metals. In comparison, bottom ash contained Fe, Co, Cr, Mn, Ce, and Ti (5.15-94.49 mg/kg), and Fe, Ti, and Cr had higher concentrations than the other metals. Coal ash contains chemical fractions of heavy metals: exchangeable, easily reduced (bounding of Fe-Mn), organic or sulfide-bound, and residue fraction. Metals in the exchangeable fraction are quickly mobilized or can become mobile, depending on environmental conditions (potential redox and acidity) [37].

Table 2 Metals in coal fly ash from some countries.

Metals	United Kingdom's Fly Ash (mg/kg)	India's Fly Ash (mg/kg)	Indonesia's Fly Ash (mg/kg)
Cd	<0.01-4	*(no data found)*	
Co	2-11	128-520	41.96-66.41
Cr	33-19	210-404	62.26-138
Cu	33-47	*(no data found)*	12.35
Hg	<0.01-0.61	9-48	0.10-0.53
Mn	103-1.55	*(no data found)*	624-9,415
Ni	35-583	*(no data found)*	
Pb	<1-976	*(no data found)*	8.95
Zn	49-918	2,029-3,100	58.57-440

Sources: [4], [38-40]

Microorganisms in Heavy Metal Bioleaching

The ability of microorganisms in bioleaching is varied. Table 3 describes the bacteria, fungi, and yeast that participate in the bioleaching process. A comparison of the ability of bacteria in bioleaching is illustrated in Table 4. Bioleaching is a method that refers to using the natural ability of microorganisms to dissolve some rocks or ores. These microorganisms can manifest themselves in extracting or mobilizing metal from minerals.

Two groups are actively involved in bioleaching. The first are chemolithotrophic bacteria, such as *Thiobacillus ferroxidan (Acidithiobacillus ferooxidan)* and other species. The second are heterotrophic microorganisms, which can be bacteria, yeast, and fungi, such as *Pseudomonas sp., Bacillus sp., Aspergillus niger,* and other species [8,10,19,41].

Table 3 Efficiency of metal bioleaching by microorganisms.

Metals	Bioleaching efficiency (%)		
	Fungi[a]	Bacteria[b]	Yeast[c]
V	12.5	15-18	70.2
Cr	*(no data found)*	13.77-85.01	51.8
Fe	1.4-8.6	3.50-70.40	40.9
Ni	11.83-50	30-64	42.6
Zn	62.33-90.84	12.18-100	55.3
Mn	*(no data found)*	6.33	57.5
Mo	100	*(no data found)*	79.5
Cd	*(no data found)*	100	45
Pb	*(no data found)*	<1	40.7
Cu	29.46-33.45	14.61-31.00	42.4

Sources:
[a][10, 19, 42-45]
[b][11, 14, 18, 41, 42, 46, 47]
[c][9]

Table 4 Comparison of heavy metal bioleaching efficiency by heterotrophic and chemolithotrophic bacteria.

Metals	**Heterotrophic**		**Chemolithotrophic**	
	Bioleaching Efficiency	**Bacteria**	**Bioleaching Efficiency**	**Bacteria**
Cu	12.05-<100%	[1,5] *Pseudomonas sp.* [3] Mixed strains and *Thellobacillus sp.* [4]*Alicyclobacillus sp.*	<5%	[11]*Acidithiobacillus thiooxidans*
As	<100%	[1] *Pseudomonas sp.*	-	(No data found)
Pb	31.26-<100%	[1]*Pseudomonas sp.* [3] Mixed strains and *Thellobacillus sp.*	4.12-59 %	[13]*Acidophylic strain* (mix) [16] Mixed strains [17]*Acidithiobacillus ferooxidan*
Cr	8.04-28%	[1,5] *Pseudomonas sp.*	24-88%	[12,14] *A. thiooxidan* [16] *Mixed strains*
Zn	10.63-28.71%	[2] *Pseudomonas aeruginosa* [5] *Pseudomonas sp.*	72.5-74.1%	[16] Mixed strains
Cd	28.98-60.97%	[3] Mixed strains and *Thellobacillus sp.*	80.7-82.1%	[16] Mixed strains
Ni	6-44.67%	[10]*Bacillus megaterium* [8]*Bacillus subtillis*	11.88-<100%	[12,14]*Acidithiobacillus thiooxidan*
Co	38.19%	[10]*Bacillus megaterium*	-	(No data found)
Fe	15-53.11%	[9]*Bacillus cereus*	15-22%	[15]*Thiobacillus thiooxidan*

Sources : [1][48]; [2][49]; [3][50]; [4][14]; [5][18]; [8][51]; [9][52]; [10][53]; [11][54];[12][55]; [13][56]; [14][46]; [15][57]; [16][58]; [17][59]

Heterotrophic microorganisms can also be used in bioleaching. Bosecker [60] stated that in the case of oxide minerals, silicate, and carbonate, the usefulness of *Thiobacillus sp.* is limited due to its poor ability to leach metals. Related to this type of minerals, Bosecker only looked at the use of heterotrophic bacteria and fungi. Heterotrophic bacteria such as *Bacillus* sp., *Pseudomonas* sp., *Acinetobacter sp*., and *Leptospirillum ferriphilum* have been reported to be able to dissolve metal [8,14,61-66]. *Bacillus sp*., part of heterotrophic bacteria family, is the most effective in dissolving metals. Meanwhile, *Aspergillus sp.* and *Penicillium sp.* are the most commonly used [60]. *Bacillus polymyxa* and *Bacillus mucilaginosus* can remove silica from bauxite, while *Aspergillus niger* can solubilize aluminum from silicate aluminum [67]. Heterotrophic microorganisms need organic components as energy source. *Bacillus sp.* culture medium contains sucrose, $CaCO_3$, essential salt, and yeast extract, and it is optimal at pH close to neutral and mesophilic temperature [14,67].

Some heterotrophic microorganisms also contribute to metal leaching by organic acid excretion. For example, *Bacillus megaterium* produces citric acid, while *Pseudomonas putida* yields gluconate and citric acid. *Aspergillus niger* produces citric acid, gluconate, succinate, oxalate, tartrate, and malate [68]. Extraction occurs as a result of a compound containing at least two hydrophilic reactive groups and dissolution of heavy metals through the direct transfer of metal ions as well as the formation of dissolved and chelated complexes [60].

A study comparing aerobic heterotrophic sulfur-oxidizing bacteria to *Thiobacillus* sp. has been conducted [69]. The result showed that those sulfur-oxidizing bacterial stayed longer in the soil than *Thiobacillus* sp. after applying S^0. Therefore, aerobic heterotrophic sulphur oxidizing bacteria had a more important role in S^0 oxidation than *Thiobacillus sp.* in the soil.

Bioleaching Mechanisms of Heavy Metals in Coal Ash

Bioleaching to mobilize, extract, or leach various types of metals from solid materials can be conducted through several mechanisms, including redoxolysis (oxidation reaction), acidolysis (the formation of acids), complexolysis (the excretion of a compression agent), and accumulation (the accumulation of metal ions) [68]. Seidel [57] states that bacteria growth and metal released from coal ash are represented by biological-chemical interactions related to various elements in this system.

Coal fly ash contains a heavy metal band with sulfides and oxides. It is characterized by hematite (Fe_2O_3), mullite ($Al_6Si_2O_{13}$), magnetite (Fe_3O_4), quartz (SiO_2), and several carbons. Besides, some metals also bind to sulfide, such as marcasite (FeS_2), millerite (NiS), sphalerite (ZnS), chalcopyrite ($CuFeS_2$), galena (PbS), cattierite (CoS_2), and siegenite ($(Ni, Co)_3S_4$) [70]. The indirect redoxolysis mechanism allows the bioleaching of heavy metals from coal ash to occur. In the direct mechanisms, the bioleaching of metals occurs through a redox reaction. The dissolution occurs through an enzymatic reaction by contact between microorganisms and solid material, leading to mineral destruction. The direct mechanism involves electron transfer from solid material to an acceptor (mainly oxygen) or inserting electrons into the solid material from a donor such as H_2 (reduction). This mechanism needs contact between bacteria cells and the surface of sulfide minerals. The direct bioleaching mechanism is profitable to autotroph microorganisms because it saves energy during solid oxidation [71].

In the indirect leaching mechanism, ion oxidation, such as Fe^{2+} becoming Fe^{3+}, dissolves metal chemically from solid material. Oxidation of metals occurs by an indirect mechanism mediated by iron ions (Fe^{3+}) from Fe^{2+} ions (from Fe compounds). Fe^{3+} ions can oxidate compounds such as metal sulfide (by microorganisms) [68]. In addition, microorganisms, specifically bacteria, will act as oxidizers in this mechanism. In other mechanisms, metal sulfide will react with Fe^{3+} (the result of oxidation of Fe^{2+}) to become metal ions (such as Cu^{2+}, Zn^{2+}, Pb^{2+}, etc), Fe^{2+}, and sulfide. The metal ions resulting from this reaction will dissolve, and the resulting Fe^{2+} will continue to react with the remaining metal sulfide. S made from this reaction will react with water (H_2O) and oxygen (O_2) to form sulfuric acid (H_2SO_4). This H_2SO_4 will form a complex with metal oxides (complexolysis mechanism) but there is very little information regarding heavy metals in metal oxide form in coal fly ash.

Biologically, bioleaching also occurs due to microorganism biosorption and bioaccumulation of metals. However, this mechanism is more dominant when using fungi or yeast. Su *et al.* in [10,11] have reported the presence of biosorption by bacteria (*Acidithiobacillus thiooxidan*) and fungi. In the liquid phase (leachate solution), metal ions accumulate through metabolism and adsorption reactions due to the balance between solids and dissolved metals.

The acidolysis mechanism involves protons for metal extraction, and metal dissolution occurs through metal compound protonation. It depletes the metal's availability, converts to a cation, and accelerates solid disintegration. Proton protonates anions from metal compounds that are not dissolved, such as O_2 atoms that encircle metal surfaces (metal oxide). Protons and O_2 are joined with H_2O to release metal from the surface. The optimum amount of available protons determines the amount of metal oxide dissolved. This process is usually quick, and it is the most dominant in fungi bioleaching [72].

The third mechanism is bioleaching by organic acids, which leaches metal through complexation to create a dissolved complex. This complexolysis mechanism is relatively slower compared to acidolysis. It dissolves metal ions based on the molecule's complexity capacity. Metals are detached from the compound particle when the ions are stronger than the lattice bond [71]. This mechanism is also essential in bioleaching using fungi [72]. Mycelium is a storage container for the metal ions in the accumulation process. Due to active metabolic reactions and passive adsorption, their accumulation from the leach solution disturbs compounds and dissolved metal balances. The fungal cell wall contains a variety of functional groups (amine, hydroxyl, amine, phosphate, carboxyl, and sulfide groups) that can bind metal ions at higher or smaller levels [73]. *Aspergillus sp.* and *Penicillium sp.* can accumulate metals from the external environment [10,19,74].

The Factors that Influence the Bioleaching of Heavy Metals in Coal Ash

The bioleaching system consists of microbial cells, materials, metabolic, and bioleaching products. Each of these components is interrelated in bioleaching processes [68]. The process is influenced by the physicochemical parameters and microbiological parameters of the bioleaching environment, the properties of the minerals to be leached, and the processing applied. However, the present paper will discuss the influence of particle size of the material, initial pH, and pulp density.

Several studies have been performed to determine how these factors influence the outcomes. Some were conducted on factors that affect the bioleaching of heavy metals from coal ash using indigenous bacteria. Particle size can influence the bioleaching process, and it increases with the contact zone between the microorganism and the material. However, this is not the case in the two-step bioleaching process. Taştan [19] found that particles with a diameter of 250 µm produced better outcomes than particles with a diameter of 120 µm. Rouchalova *et al.* [76] conducted a bioleaching study using bacteria in sediments, which showed that a particle size of 71-100 µm gave better results than a particle size of 100-200 µm.

Chemolithotroph bacteria such as *Acidithiobacillus* sp. can live at low pH [77]. However, some studies [76] used various pH values according to the type of microorganism. For example, *Acidithiobacillus ferrooxidan* was used to investigate metal bioleaching in sediment with a pH range of 1.6 to 2.2. The result showed that the highest metal solubility yield was at pH 1.8 [76]. Pradhan *et al.* [78] conducted a bioleaching study using *Acidithiobacillus ferrooxidan* at pH 2.0. However, unlike other studies, Ref. [79] applied pH 4.0 for *Acidithiobacillus ferrooxidan.* Whereas metals were bioleached from fly ash using *Acidithiobacillus thiooxidans* at a pH range of 3.5 to 5.0 [11,57]. In the heterotroph bacteria procedure, the pH was about 5 to 9 [14].

Pulp density or the amount of material added to the aqueous medium affects bioleaching. Su *et al.* in [11] adapted *Acidithiobacillus thiooxidan* in 1 to 30% pulp density and 10% pulp density used in bioleaching showed the best yield. In another study [14], bioleaching of Cu and Fe with heterotroph bacteria was most effective when 5% pulp density was higher than 20%. The efficiency is also positively correlated to Fe, which dissolves in media. This is caused by proton H^+ and the concentration of Fe (III) influenced by extracellular polymeric substances (EPS).

Type of Bioleaching

Two types of bioleaching are widely used in the field of bioleaching: one-step (direct) and two-step (indirect). One-step bioleaching is conducted using microorganisms on the log phase in liquid form into the medium and the material. The supernatant is employed in the bioleaching process by centrifuging the log phase bacteria, as in two-step bioleaching (indirect bioleaching) [9-11].

Hassanien *et al.* in [75] investigated bioleaching using fungi and bacteria with direct and indirect methods. The results showed that direct bioleaching in monazite was better than indirect bioleaching, i.e., 75.4% and 63.5% rare earth metals were leached, respectively. Only organic acids (complexation mechanisms) and other mechanisms such as biosorption indirectly affect metal bioleaching. Direct bioleaching is more efficient because of several mechanisms. The study also showed that bioleaching is better than chemical leaching. The enhancement of material concentration (pulp density) caused the amount of microorganism biomass to decrease. As a result, the yield of bioleaching increased along with time. In indirect bioleaching, the pH decreases with the bioleaching process. Unlike Hassanien *et al.* in [75], the results of Taştan using fungi in fly ash were shown by two-step or indirect bioleaching rather than one-step bioleaching [19].

Bioleaching Kinetics

Few studies have been done on the bioleaching kinetics of heavy metals in coal ash. Based on the results of the literature review, Pangayao *et al.* [8] conducted a study on the kinetics of bioleaching trace elements in coal fly ash using *Pseudomonas* spp. Other bioleaching kinetics have been studied using different types of microorganisms and materials (not coal ash). The models suggested were shrinking core and stochastic models [74,80]. The bioleaching kinetics of heavy metals from coal ash is a typical liquid-solid reaction [15]. A shrinking core model can mimic bioleaching from coal ash represented by spherical particles. The heavy metal leaching may be restricted by using the subsequent steps of surface chemical reactions (Eq. 1) or internal diffusion through a directed kinetic model (Eq. 2) [81].

$$1 - (1 - x)^{1/3} = kr\, t \tag{1}$$

$$1 - 2/3x - (1 - x)^{2/3} = k_d t \tag{2}$$

where,

x = heavy metal leached (efficiency bioleaching)

kr and kd = the apparent reaction rate constant (h−1).

t = leaching time (h)

According to bioleaching kinetics data covering 30 days using the shrinking core model, metal leaching rates are controlled by diffusion through the ash layer [8]. The kinetic studies of Ni and Cr from electroplating sludge using *Aspergillus niger* by Nikfar *et al.* in [74] showed that the extraction continued with one kinetic stage for Cr and two kinetic stages for Ni. Furthermore, Cr extraction was controlled by interface transfer and diffusion over the item layer.

The primary stage of Ni extraction was controlled by interface transfer and diffusion across the item layer, and the second stage of Ni leaching was a mixed-control mechanism. Diffusion through the product layer and chemical reactions were involved. Inevitably, the results of this study describe that *Aspergillus niger* (filtrate culture) can extract metals from e-waste sludge. Research on the bioleaching kinetics of heavy metals in coal ash using the shrinking core model is still limited. Further studies regarding bioleaching kinetics are needed.

The Progress of Heavy Metal Bioleaching Research in Coal

Some studies have already been carried out on heavy metal bioleaching in coal using microorganisms [8-11, 41, 82]. Pangayao *et al.* in [41] examined the ability of the bacteria from the genus of *Acidithiobacillus* in heavy metal bioleaching from coal. Park and Liang [9] examined the ability of yeast in metal bioleaching from coal fly ash. However, they did not analyze the influence of other factors and the mechanism of heavy metal bioleaching. Some studies investigated the factors influencing metal extraction from coal ash.

Taştan [19] conducted bioremediation by using several fungi to decrease the heavy metal concentration in coal fly ash. The mechanism correlated to organic acid and the availability of Fe (II) has not been analyzed. Besides, Fe (II) has an essential part in heavy metal bioleaching through the indirect redoxylosis mechanism. Su *et al.* [10, 11] examined the ability of *Aspergillus niger* and *Acidithiobacillus thiooxidan* in metal bioleaching using one- and two-step bioleaching.

Su *et al.* in [10,11] showed that one-step bioleaching, combined with chemical hydrothermal treatment, obtained the best result in extracting metals. Park and Liang [9] used two-step bioleaching in their yeast study and obtained the best results. Taştan [19] also compared two kinds of bioleaching using fungi at some initial pH. The result showed that two-step bioleaching obtained a higher value than one-step bioleaching. These two studies showed different results, but Hassanien *et al.* [75] found out that one-step or direct bioleaching is more efficient than two-step bioleaching. In Su *et al.* [10, 11], the importance of oxidizing agents such as Fe^{2+} and S^{+} in the bioleaching process is not highlighted.

Several studies have investigated metal leaching kinetics in fly ash [83-88]. However, bioleaching kinetics of heavy metals in coal ash studies using bacteria has only been performed by Pangayao [8]. Heavy metal bioleaching kinetics in coal ash using yeast or fungi has also been investigated to a limited degree.

Figure 1 shows the number of studies on heavy metal bioleaching in coal ash. The information on heavy metal bioleaching in coal ash is still quite limited, as described in Figure 1. In addition, only a limited number of studies on bioleaching mechanisms and kinetics have been conducted. All mechanisms in bioleaching can occur using bacteria, but which mechanism dominates will be different for each bacterium. Whereas in fungi, yeast, or bacteria that do not play a role in the iron or sulfur redox reactions, the redoxolysis mechanism does not occur in bioleaching. However, it is still possible with other mechanisms. Research on which mechanism predominates in bacteria, fungi, or yeast can be an opportunity for further research. In addition, the study of the bioleaching kinetics of heavy metals, especially in coal ash using fungi and yeast, needs further investigation. This is due to the limited information on this topic.

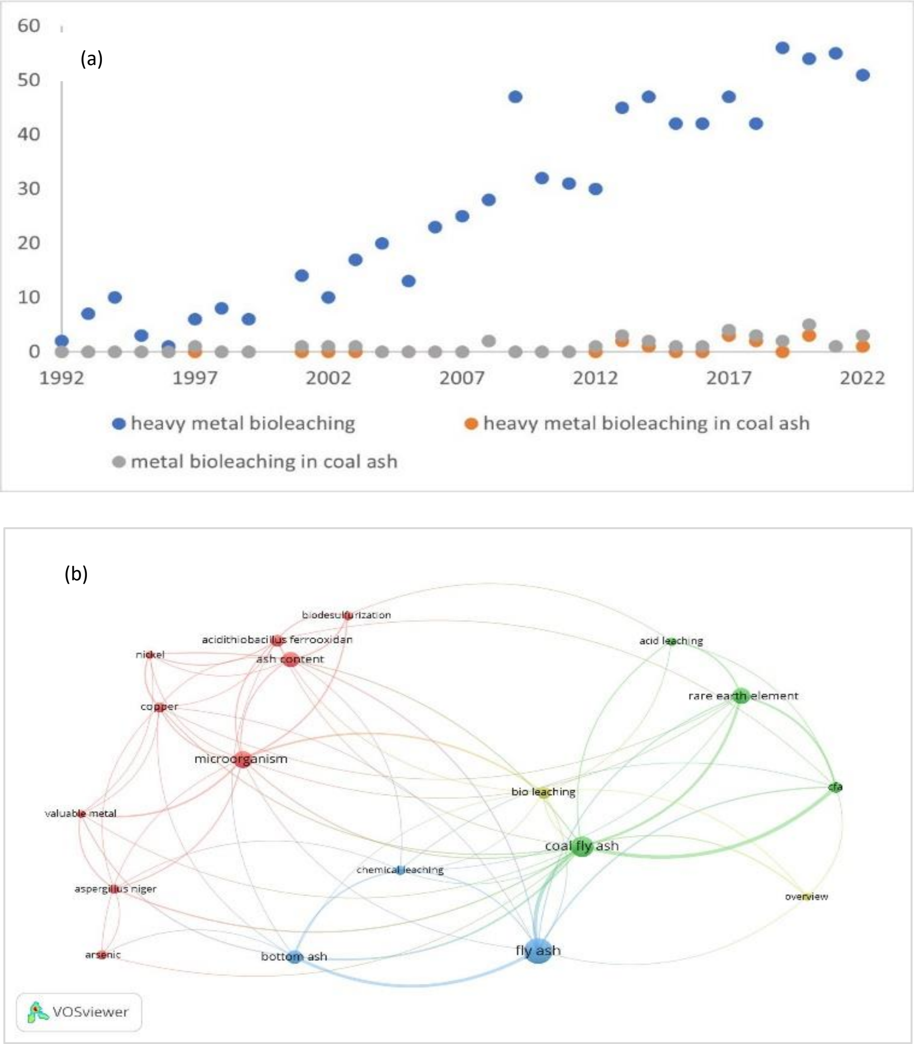

Figure 1 (a) The number of publications related to heavy metal bioleaching, metal bioleaching in coal ash, and heavy metal bioleaching of coal ash. The number of publications was obtained by using the Scopus database. (b) Results of bibliographic analysis by using VOSviewer. The analysis is based on the co-occurrence of keywords related to bioleaching of coal ash.

Conclusions

According to the findings of this study, coal ash contains varying concentrations of heavy metals. Bioleaching of heavy metals in coal ash is combined through biological and chemical reactions that involve numerous constituents in the system. The ability of microorganisms varies and can be influenced by several conditions, such as pH, pulp density, temperature, nutrients, type of material, and others. One or two kinds of metals cannot be leached explicitly because of challenges. All metal in the coal ash will be removed through these methods, and the yield varies according to the bonding and concentration in the coal ash. Furthermore, the high concentration of Fe and heavy metals leached could be toxic for microorganisms. Most of the studies were carried out on a small scale in the laboratory using batch reactors. The bioleaching kinetics of heavy metals in coal ash is a typical liquid-solid reaction as described by the shrinking core model.

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References

[1] Khan, I. & Umar, R., *Environmental Risk Assessment of Coal Fly Ash on Soil and Groundwater Quality, Aligarh*, India, Groundw. Sustain. Dev., **8**, pp. 346-357, 2018.

[2] Savic, D., Nisic, D., Malic, N., Dragosavljevic, Z. & Medenica, D., *Research on Power Plant Ash Impact on the Quality of Soil in Kostolac and Gacko Coal Basins*, Minerals, **8**(2), pp. 1-16, 2018.

[3] Verma, C., Madan, S. & Hussain, A*., Heavy Metal Contamination of Groundwater due to Fly Ash Disposal of Coal-Fired Thermal Power Plant*, Parichha, Jhansi, India, Cogent Eng., **3**(1), pp. 1-8, 2016.

[4] Wiyono, M. & Wahyudi, W., *Analysis of Element Contents in Fly Ash of Coal Power Plant Industry with Neutron Acivation Analysis*, J. Teknol. Lingkung., **19**(2), pp. 221-228, 2018. (Text in Indonesian and Abstract in English).

[5] Shetty, N., Shetty, J.K., Chadaga, D.M. & Shankara, D.U., *Trace Analysis of Heavy Metals in Ground Water and Soil near Coal Based Thermal Power Plant Udupi Karnataka*, J. Univ. Shanghai Sci. Technol., **23**(2), pp. 382-395, 2021.

[6] Ompusunggu, M. & Adri, N., *Kalimantan Coal Firm Fined Rp 2 billion For Dumping Hazardous Waste,* The Jakarta Post, 2018, accessed from https://www.thejakartapost.com/news/2018/03/08/e-kalimantan-coal-firm-fined-rp-2-billion-for-dumping-hazardous-waste.html (April 06, 2022).

[7] Kumari, A., *Novel Approach to Recover Rare Earth Metals (Rems) From Indian Coal Bottom Ash*, Hydrometallurgy, **187**(April), pp. 1-7, 2019.

[8] Pangayao, D., Promentilla, M.A., Gallardo, S. & van Hullebusch, E., *Bioleaching Kinetics of Trace Metals from Coal Ash Using Pseudomonas spp.*, MATEC Web Conf., **268**, 01010, 2019.

[9] Park, S. & Liang, Y., *Bioleaching of Trace Elements and Rare Earth Elements from Coal Fly Ash*, Int. J. Coal Sci. Technol., **6**(1), pp. 74-83, 2019.

[10] Su, H.F., Chen, H. & Lin, J.F., *A Sequential Integration Approach Using Aspergillus Niger to Intensify Coal Fly Ash As A Rare Metal Pool*, Fuel, **270**, November 2019, 117460, 2020.

[11] Su, H.F., Tan, F. & Lin, J.F., *An Integrated Approach Combines Hydrothermal Chemical and Biological Treatment to Enhance Recycle of Rare Metals from Coal Fly Ash*, Chem. Eng. J., **395**(February), 124640, 2020.

[12] Reed, D.W., Fujita, Y., Daubaras, D.L., Jiao, Y. & Thompson, V.S., *Bioleaching of Rare Earth Elements from Waste Phosphors and Cracking Catalysts*, Hydrometallurgy, **166**, pp. 34-40, 2016.

[13] Joulian, C., Fonti, V., Chapron, S., Bryan, C.G. & Guezennec, A.G., *Bioleaching of Pyritic Coal Wastes: Bioprospecting and Efficiency of Selected Consortia*, Res. Microbiol., **171**(7), pp. 260-270, 2020.

[14] Chaerun, S.K., Putri, F.Y., Minwal, W.P., Ichlas, Z.T. & Mubarok, M.Z., *Bacterial Leaching of an Indonesian Complex Copper Sulfide Ore Using an Iron-Oxidizing Indigenous Bacterium*, Microbiol. Indones., **12**(1), pp. 1-6, 2018.

[15] Wang, X., *Effect of Particle Size on Uranium Bioleaching in Column Reactors from a Low-Grade Uranium Ore*, Bioresour. Technol., **281**, December 2018, pp. 66-71, 2019.

[16] Sharada, H.M., Abdel-halim, S.A. & Hafez, M.A., *Bioleaching of Copper from Electronic Waste Using Aspergillus niger*, Biointerface Res. Appl. Chem., **12**(6), pp. 8406-8425, 2022.

[17] Pangayao, D.C., *Bioleaching of Trace Metals from Coal Ash Using Acidithiobacillus Albertensis, Acidithiobacillus Thiooxidans and Local Isolate from Coal Ash Pond*, Gokongwei College of Engineering, 2016, accessed from https://animorepository.dlsu.edu.ph/etd_doctoral/501 (April 06, 2022).

[18] Pangayao, D., Gallardo, S., Promentilla, M.A. & Van Hullebusch, E., *Bioleaching of Trace Metals from Coal Ash Using Local Isolate from Coal Ash Ponds*, MATEC Web Conf., **156**, pp. 2-6, 2018.

[19] Taştan, B.E., *Clean Up Fly Ash from Coal Burning Plants by New Isolated Fungi Fusarium Oxysporum and Penicillium Glabrum*, J. Environ. Manage., **200**, pp. 46–52, 2017.

[20] Das, S.K., *A Novel Approach on Leaching Study for Removal of Toxic Elements from Thermal Power Plant-Based Fly Ash Using Natural Bio-Surfactant*, Case Stud. Chem. Environ. Eng., **4**, October, 100156, 2021.

[21] Chen, Z., *Bioleaching Coal Gangue with a Mixed Culture of Acidithioba-cillus ferrooxidans and Acidithiobacillus thiooxidans*, Minerals, **11**(10), 1043, 2021.

[22] IEA, *Coal Information: Overview*, IEA, Paris, 2021, accessed from https://www.iea.org/reports/coal-information-overview/consumption (Dec. 16, 2022).

[23] BP, *Statistical Review of World Energy*, 2021. Available: bp.com/statisticalreview

[24] Yao., Z.T*., A Comprehensive Review on the Applications of Coal Fly Ash*, Earth-Science Rev., **141**, pp. 105-121, 2015.

[25] Indonesia Power, *Quality Inventory of Fly Ash and Bottom Ash Indonesia Power and Subsidiaries Unit April 2020 Reporting Period (2017-2019*), Indonesia, 2021. (Text in Indonesian).

[26] Rafieizonooz, M., Khankhaje, E. & Rezania, S., *Assessment of Environmental and Chemical Properties of Coal Ashes Including Fly Ash and Bottom Ash, and Coal Ash Concrete*, J. Build. Eng., **49** (November 2021), 104040, 2022.

[27] Mangi, S.A., Wan Ibrahim, M.H., Jamaluddin, N., Arshad, M.F. & Putra Jaya, R., *Short-Term Effects of Sulphate and Chloride on the Concrete Containing Coal Bottom Ash as Supplementary Cementitious Material*, Eng. Sci. Technol. an Int. J., **22**(2), pp. 515-522, 2019.

[28] Hasim, A.M., Shahid, K.A., Ariffin, N.F., Nasrudin, N.N. & Zaimi, M.N.S., *Study on Mechanical Properties of Concrete Inclusion of High-Volume Coal Bottom Ash with the Addition of Fly Ash*, Mater. Today Proc., **51**, pp. 1355-1361, 2021.

[29] Mangi, S.A., Wan Ibrahim, M.H., Jamaluddin, N., Arshad, M.F., Memon, S.A. & Shahidan, S., *Effects of Grinding Process on the Properties of the Coal Bottom Ash and Cement Paste*, J. Eng. Technol. Sci., **51**(1), pp. 1-13, 2019.

[30] Meena, A., Singh, N. & Singh, S.P., *High-Volume Fly Ash Self Consolidating Concrete with Coal Bottom Ash and Recycled Concrete Aggregates: Fresh*, *Mechanical and Microstructural Properties*, J. Build. Eng., **63**(PA), 105447, 2023.

[31] Saikia, B.K., Hower, J.C., Islam, N., Sharma, A. & Das, P.*, Geochemistry and Petrology of Coal and Coal Fly Ash From A Thermal Power Plant In India*, Fuel, **291**, January, 120122, 2021.

[32] Singh, M. & Siddique, R., *Properties of Concrete Containing High Volumes of Coal Bottom Ash as Fine Aggregate*, J. Clean. Prod., **91**, pp. 269-278, 2015.

[33] Zhou, C., *Distribution and Preconcentration of Critical Elements from Coal Fly Ash by Integrated Physical Separations*, Int. J. Coal Geol., **261**, August, 104095, 2022.

[34] Wu, G., Wang, T., Chen, G., Shen, Z. & Pan, W.P., *Coal Fly Ash Activated by Naoh Roasting: Rare Earth Elements Recovery and Harmful Trace Elements Migration*, Fuel, **324**(2022), 124515, 2022.

[35] Cheng, B., *Effects of Fly and Coal Bottom Ash Ratio on Backfill Material Performance*, Constr. Build. Mater., **319**, November 2021, pp. 125831, 2022.

[36] Tan, Y., He, Y., Cui, X. & Liu, L.*, Design and Performance Optimization of Alkali-Activated Waste Coal Bottom Ash / Slag Porous Concrete,* Constr. Build. Mater., **359**, June, 129413, 2022.

[37] Damayanti, R., *Coal Ash and Its Utilization: A Technical Review on Its Chemically Characteristics and Toxicology*, J. Teknol. Miner. dan Batubara, **14**(3), pp. 213-231, 2018. (Text in Indonesian and Abstract in English).

[38] Sear, L.K.A., Weatherley, A.J., CEng, M. & Dawson, A., *The Environmental Impacts of Using Fly Ash – the {UK} Producers' Perspective*, in the 2003 International Utilization Symposium, 14, 2003.

[39] Ahmad, B., Fukelar, M.H. & Pathak, *Impact of Coal Based Thermal Power Plant on Environment and Its Mitigation Measure*, Int Res J Env. Sci, **1**, pp. 60-64, 2012.

[40] Julharmito, Faldi, A. & Dastinawati, Utilization of Coal Fly Ash as a Mixing Material for Geopolymer Concrete, JOM FTEKNIK, **2**(2), pp. 1-7, 2015. (Text in Indonesian and Abstract in English).

[41] Pangayao, D.C., Van Hullebusch, E.D., Gallardo, S.M. & Bacani, F.T., *Bioleaching of Trace Metals from Coal Ash Using Mixed Culture of Acidithiobacillus Albertensis and Acidithiobacillus Thiooxidans*, J. Eng. Sci. Technol., **10**(8), pp. 36-45, 2015.

[42] Groudev, S.N., *Use of Heterotrophic Microorganisms in Mineral Biotechnology*, Acta Biotechnol., **7**(4), pp. 299-306, 1987.

[43] Taştan, B.E., *Effective Removal of Fly Ash by Penicillium Chrysogenum and Determination of Direct Fly Ash Toxicity with Daphnia Magna*, Water Supply, 21(5), pp. 2047-2057, 2021.

[44] Faraji, F., Golmohammadzadeh, R., Rashchi, F. & Alimardani, N.*, Fungal Bioleaching of Wpcbs Using Aspergillus Niger: Observation, Optimization and Kinetics*, J. Environ. Manage., **217**, pp. 775–787, 2018.

[45] Kurniawan, R., Juhanda, S., Gustinah, H.M. & Pratami, A.P., *Application of Bioleaching in the Separation of Nickel Oxide Metal with Aspergillus niger and Penicillium chrysogenum*, Pros. Semin. Nas. Tek. Kim. "Kejuangan," April, pp. A2-(1-7), 2018. (Text in Indonesian and Abstract in English)

[46] Gomes, H.I., Funari, V., Mayes, W.M., Rogerson, M. & Prior, T.J., *Recovery of Al, Cr, and V from Steel Slag By Bioleaching: Batch and Column Experiments*, J. Environ. Manage., **222**, January, pp. 30-36, 2018.

[47] Meer, I. & Nazir, R., *Removal Techniques for Heavy Metals from Fly Ash*, J. Mater. Cycles Waste Manag., **20**(2), pp. 703-722, 2018.

[48] Biswal, B.K., Chen, Z. & Yang, E.H., *Hydrothermal Process Reduced Pseudomonas Aeruginosa PAO1-Driven Bioleaching of Heavy Metals in a Novel Aerated Concrete Synthesized Using Municipal Solid Waste Incineration Bottom Ash*, Chem. Eng. J., **360**, October 2018, pp. 1082-1091, 2019.

[49] Honarjooy Barkusaraey, F., Mafigholami, R., Faezi Ghasemi, M. & Khayati, G., *Zn Bio Extraction From A Zinc Rich Paint Sludge By Indigenous Pseudomonas Aeruginosa*, Chem. Eng. Commun., **209**(10), pp. 1397-1412, 2022.

[50] Liu, C., Ma, Q., Zhou, X., Lai, H. & Li, L*., Bioleaching of Heavy Metals from Sludge by Mixed Strains*, IOP Conf. Ser. Earth Environ. Sci., **208**(1), 2018.

[51] Giese, E.C., Carpen, H.L., Bertolino, L.C. & Schneider, C.L*., Characterization and Bioleaching of Nickel Laterite Ore Using Bacillus Subtilis Strain*, Biotechnol. Prog., **35**(6) 2019.

[52] Jun, Y.H., Nee, Y.S., Qi, C.W., Chieng, S. & How, K.S., *Bioleaching of Kaolin With Bacillus Cereus: Effects of Bacteria Source and Concentration on Iron Removal*, J. Sustain. Sci. Manag., **15**(4), pp. 91–99, 2020.

[53] Cui, X., *Contact-Bioleaching Mechanism of Ni and Co from Sulfide Concentrate at Neutral Ph by Heterotrophic Bacteria*, Miner. Metall. Process., **35**(4), pp. 221-229, 2018.

[54] Ferreira, P.F., Sérvulo, E.F.C., Da Costa, A.C.A., Ferreira, D.M., Godoy, M.L.D.P. & Oliveira, F.J.S., *Bioleaching of Metals from A Spent Diesel Hydrodesulfurization Catalyst Employing Acidithiobacillus Thiooxidans FG-01*, Brazilian J. Chem. Eng., **34**(1), pp. 119-129, 2017.

[55] Fang, D. & Zhou, L.X., *Enhanced Cr Bioleaching Efficiency from Tannery Sludge with Coinoculation of Acidithiobacillus Thiooxidans TS6 And Brettanomyces B65 In An Air-Lift Reactor*, Chemosphere, **69**(2), pp. 303-310, 2007.

[56] Funari, V., Mäkinen, J., Salminen, J., Braga, R., Dinelli, E. & Revitzer, H., *Metal Removal from Municipal Solid Waste Incineration Fly Ash: A Comparison Between Chemical Leaching and Bioleaching*, Waste Manag.,**60**, pp. 397-406, 2017.

[57] Seidel, A., Zimmels, Y. & Armon, R., Mechanism of Bioleaching of Coal Fly Ash By Thiobacillus Thiooxidans, Chem. Eng. J., **83**(2), pp. 123-130, 2001.

[58] Zhang, R., Wei, X., Hao, Q. & Si, R., *Bioleaching of Heavy Metals From Municipal Solid Waste Incineration Fly Ash: Availability of Recoverable Sulfur Prills and Form Transformation of Heavy Metals*, Metals (Basel)., **10**(6), pp. 1-14, 2020.

[59] Ye, M., *Bioleaching Combined Brine Leaching of Heavy Metals from Lead-Zinc Mine Tailings: Transformations During the Leaching Process*, Chemosphere, **168**, pp. 1-11, 2016.

[60] Bosecker, K., *Bioleaching: Metal Solubilization by Microorganisms*, FEMS Microbiol. Rev., **20**(20), pp. 591-604, 1997.

[61] Ghosh, S. & Das, A., *Bioleaching of Manganese from Mining Waste Residues Using Acinetobacter Sp.*, Geol. Ecol. Landscapes, **1**(2), pp. 77-83, 2017.

[62] Thavamani, P., Samkumar, R., Satheesh, S., Subashchandrabos, V., Ramadass, K., Naidu, R., Venkateswarlu, M. & Megharaj, K., *Microbes from Mined Sites: Harnessing Their Potential for Reclamation of Derelict Mine Sites*, Environ. Pollut., **230**, pp. 495-505, 2017.

[63] He, H., Dong, Z., Peng, Q., Wang, X., Fan, C. & Zhang, X., *Impacts of Coal Fly Ash on Plant Growth and Accumulation of Essential Nutrients and Trace Elements by Alfalfa (Medicago Sativa) Grown in a Loessial Soil*, J. Environ. Manage., 197, pp. 428-439, 2017.

[64] Saleh, D., Abdollahi, H., Noaparast, M., Nosratabad, A. & Tuovinen, O., *Dissolution of Al from Metakaolin with Carboxylic Acids Produced by Aspergillus Niger, Penicillium Bilaji, Pseudomonas Putida, and Pseudomonas Koreensis*, Hydrometallurgy, 186, pp. 235-243, 2019.

[65] Wang, X., *Synergistic Effect of Synergetic Effect of Pyrite on Strengthening Bornite Bioleaching by Leptospirillum Ferriphilum*, Hydrometallurgy, **176**, pp. 9-16, 2018.

[66] Guo, Y., Teng, Q., Yang, Z., Sun, B. & Liu, S., *Investigation on Bio-Desilication Process of Fly Ash Based on A Self-Screened Strain of Bacillus Amyloliquefaciens and Its Metabolites*, J. Biotechnol., **341**(August), pp. 146-154, 2021.

[67] Vasan, S.S., Modak, J.M. & Natarajan, K.A., *Some Recent Advances in the Bioprocessing of Bauxite*, Int. J. Miner. Process., **62**, pp. 173-186, 2001.

[68] Brandl, H., *Microbial Leaching of Metals, in Biotechnology*, H.-J. Rehm and G. Reed, Eds. Wiley-Verlag, pp. 191-224, 2001.

[69] Yang, Z.H., Stoven, K., Haneklaus, S., Singh, B.R. & Schnug, E., *Elemental Sulfur Oxidation by Thiobacillus spp. and Aerobic Heterotrophic Sulfur-Oxidizing Bacteria*, Pedosphere, **20**(1), pp. 71-79, 2010.

[70] Vaughan, D.J. & Corkhill, C.L., *Mineralogy of Sulfides*, Elements, **13**(2), pp. 81-87, 2017.Ehrlich, H.L., *Metal Extraction and Ore Discovery, in Encyclopedia of Microbiology, 3rd ed.*, J. Lederberg, Ed. San Diego: Academic Press, pp. 75-80, 1992.

[71] Burgstaller, W. & Schinner, F., *Leaching of Metals with Fungi*, J. Biotechnol., **27**(2), pp. 91-116, 1993.

[72] Kapoor, A. & Viraraghavan, T., *Fungal Biosorption -- An Alternative Treatment Option for Heavy Metal Bearing Wastewaters: A Review*, Bioresour. Technol., **53**(3), pp. 195-206, 1995.

[73] Nikfar, S., Parsa, A., Bahaloo-Horeh, N. & Mousavi, S.M., *Enhanced Bioleaching of Cr and Ni from A Chromium-Rich Electroplating Sludge Using the Filtrated Culture of Aspergillus Niger*, J. Clean. Prod., **264**, 121622, 2020.

[74] Hassanien, W.A.G., Desouky, O.A.N. & Hussien, S.S.E., *Bioleaching of Some Rare Earth Elements from Egyptian Monazite Using Aspergillus Ficuum and Pseudomonas Aeruginosa*, Walailak J. Sci. Technol., **11**(9), pp. 809-823, 2014.

[75] Rouchalova, D., Rouchalova, K., Janakova, I., Cablik, V. & Janstova, S., *Ph and Pulp Density Using Acidithiobacillus Ferrooxidans*, Miner. 2020, **10**(11), pp. 1-26, 2020.

[76] Brenner, D.J., Krieg, N.R. & Staley, J.T, *Bergey's Manual of Systematic Bacteriology, Second edition. Volume Two: The Proteobacteria, Part B: Gammaproteobacteria, 2nd ed*., Springer, 2005.

[77] Pradhan, D., Pal, S., Sukla, L.B., Chaudhury, G.R. & Das, T., *Bioleaching of Low-Grade Copper Ore Using Indigenous Microorganisms*, Indian J. Chem. Technol., **15**(6), pp. 588-592, 2008.

[78] Pandey, A., Srivastava, A. & Pandey, A., *Acidithiobacillus Ferrooxidans: A Bioleaching Bacteria for Better Iron(II) Oxidation Ability*, Int. J. Innov. Biol. Chem. Sci., **1**(January), pp. 32-37, 2011.

[79] Mirwan, A., Susianto, Altway, A. & Handogo, R., *A Modified Shrinking Core Model for Leaching of Aluminum From Sludge Solid Waste of Drinking Water Treatment*, Int. J. Technol., **8**(1), pp. 19-26, 2017.

[80] Sun, Y., Fu, G. & Jiang, L., *Kinetic Study of the Leaching of Low-Grade Manganese Ores by Using Pretreated Sawdust as Reductant*, Minerals, **7**(83), pp. 1-9, 2017.

[81] Singh, P.K., Singh, A.L., Kumar, A. & Singh, M.P., *Mixed Bacterial Consortium as an Emerging Tool to Remove Hazardous Trace Metals from Coal*, Fuel, **102**, pp. 227-230, 2012.

[82] Seidel, A. & Zimmels, Y., *Mechanism and Kinetics of Aluminum and Iron Leaching from Coal Fly Ash by Sulfuric Acid*, Chem. Eng. Sci., **53**(22), pp. 3835-3852, 1998.

[83] Sangita, S. & Panda, C.R., *Kinetics of Aluminium Leaching from Coal Fly Ash by Sulphuric Acid*, Indian J. Chem. Technol., **27**(July), pp. 263-273, 2020.

[84] Shoppert, A., Loginova, I. & Valeev, D., *Kinetics Study of Al Extraction from Desilicated Coal Fly Ash by NaOH at Atmospheric Pressure*, Materials (Basel)., **14**(7700), pp. 1-20, 2021.Wu, Y., Yang, X., Li, L., Wang, Y. & Li, M., Kinetics of Extracting Alumina by Leaching Coal Fly Ash with Ammonium Hydrogen Sulfate Solution, Chem. Pap., **73**(9), pp. 2289-2295, 2019.

[85] Rahaman, M.A., Gafur, M.A. & Kurny, A.S., *Kinetics of Recovery of Alumina from Coal Fly Ash through Fusion with Sodium Hydroxide*, Am. J. Mater. Eng. Technol., **1**(3), pp. 54-58, 2013.

[86] Kim, J., Park, S., Hong, J., Shin, D. & Jeong, J., *Leaching Kinetics of Yttrium Extraction from Coal Fly Ash using Sulfuric Acid*, KEPCO J. Electr. Power Energy, **3**(1), pp. 29-34, 2017.

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