

Article

Assessment of Characteristics of Acid Mine Drainage Treated with Fly Ash

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Abstract: Acid mine drainage (AMD) occurs naturally in abandoned coal mines, and it contains hazardous toxic elements in varying concentrations. In the present research, AMD samples collected from an abandoned mine were treated with fly ash samples from four thermal power plants in Singrauli Coalfield in the proximate area, at optimized concentrations. The AMD samples were analyzed for physicochemical parameters and metal content before and after fly ash treatment. Morphological, geochemical and mineralogical characterization of the fly ash was performed using SEM, XRF and XRD. This laboratory-scale investigation indicated that fly ash had appreciable neutralization potential, increasing AMD pH and decreasing elemental and sulfate concentrations. Therefore, fly ash may be effectively used for AMD neutralization, and its suitability for the management of coalfield AMD pits should be assessed further.

Keywords: abandoned mine; fly ash; acid mine drainage; SEM



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1. Introduction

Mining activities exert an inherently high impact on the environment, and as such a common framework for responsible mining is urgently needed [1]. Among the various pollution sources related to mining activities, acid mine drainage (AMD) is created in areas that have previously been mined and that contain pyritic materials in spoil piles or mine shafts; iron pyrite in the tailings chemically reacts with oxygen and water, facilitated by the *Thiobacillus* sp. bacteria, resulting in the creation of AMD. Therefore, AMD is a dangerous industrial byproduct, highly acidic and rich in sulfates [2]. The recycling of this secondary waste may reduce environmental risks, as well as increase the economic profitability of the mining industry [3]. In any case, while mine waste piles accumulate, AMD is continuously formed, and this affects industrial environments well after closure of mining activities. In and around coal mines, the pollution of surface water by AMD and other mine effluents may have serious environmental implications [4–6]. In many industrial settings, the consequences of AMD are considered moderate to severe [7,8]. There are various methods for treating AMD, such as using calcium carbonate, sodium hydroxide, sodium bicarbonate or anhydrous ammonia, as well as zeolite-adsorption processes. The fresh zeolite, for example, presents a high ammonia removal efficiency from mine effluents [9–11]. These chemicals raise the pH to acceptable levels and decrease the solubility of dissolved metals, but the cost of the treatment of AMD is usually very high since the latter is generated up to 300 m³/day for 2000 tons/day or less of mine production capacity [12]. On another note, fly ash is another deleterious byproduct generated by coal burning. Its toxicity potential (metal leaching) may be higher in low pH batches [13]. In the literature, neutralization of AMD has been attempted through various treatment systems, a simple one being the addition of lime. This method efficiently increases AMD

pH value, but it necessitates large lime quantities and produces unstable secondary wastes. Other chemical [14,15], biological and microbiological [16–18] methods have also been applied, but clogging due to metal hydroxide precipitation may interfere with the biological and microbiological activity [19]. As such, a satisfactory and economic solution for the long-term treatment of AMD is still needed. Ideally, the material applied should be an undesirable byproduct not used for any other application [20].

Especially in India, which mostly depends on thermal power plants to meet its energy requirements, fly ash production is significant. India's total installed power-generation capacity was 350,162 MW in March 2019, out of which the thermal power plants contributed to 222,927 MW; i.e., 63.7% of the total power-generation capacity of India [21]. Fly ash is indeed an undesirable byproduct of coal combustion, disposed of in specially designed landfills in an environmentally safe manner. This waste has been proven to be appropriate for AMD treatment due to its alkaline properties [22–25]. As an extravagant example, water quality from a stream heavily affected by AMD, with low macroinvertebrate productivity and diversity, significantly improved after impoundment works that directed waters from a fly ash pond to the stream. Species diversity and benthic production increased, and a few opportunistic fish feeding on insects appeared [26]. Focusing on these two kinds of toxic industrial waste, fly ash produced from a thermal power plant was characterized by its geochemical and mineralogical profile. Its suitability to successfully neutralize AMD pits was then assessed. These preliminary results may be utilized to manipulate fly ash to aid the neutralization of AMD, reducing its potential for metal mobilization.

2. Materials and Methods

2.1. Field Investigation and Sample Collection

The Singrauli Coalfield is located between latitudes 24°12' N and 23°47' N in India, and four combustion plants are present (Figure 1). The fly ash used in this study was collected from these plants in a dry state from the plants' electrostatic precipitators. The chute hoppers were carefully opened, and gunny sacks made of strong poly-coated cotton with a 30 kg capacity were used to collect the dry fly ash. Each bag's mouth was sealed immediately after collection, inserted into another poly-coated bag and brought to the laboratory (Banaras Hindu University).

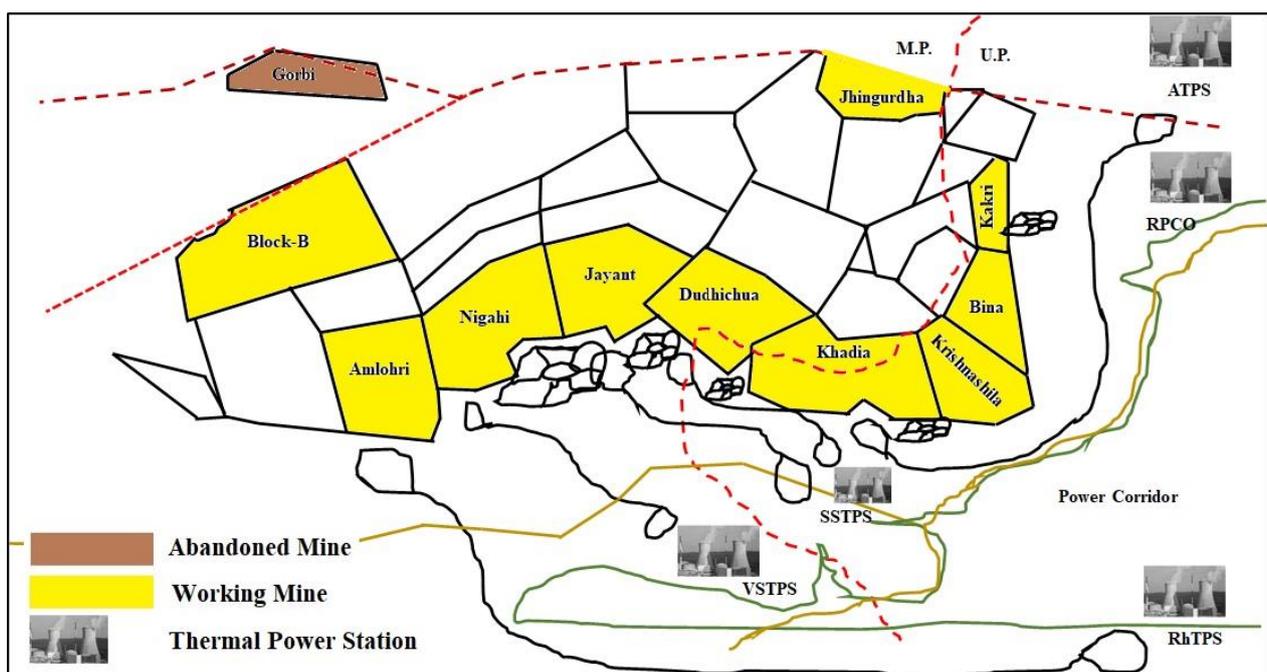


Figure 1. The study area.

The Gorbi mine (Figure 2) is an abandoned mine at the north of the Singrauli field that has been shown to produce AMD (pH 2.54, in-house data). Pit I, Pit II and Pit III of the mine were used for the collection of AMD for the present experiment. Further information on the AMD pits is given in Table 1. The sample collection and the physical and species chemical analysis of AMD followed Indian standard guidelines [27–30]. All samples were brought to the laboratory (BHU) in sealed gunny sacks and stored until further analysis.

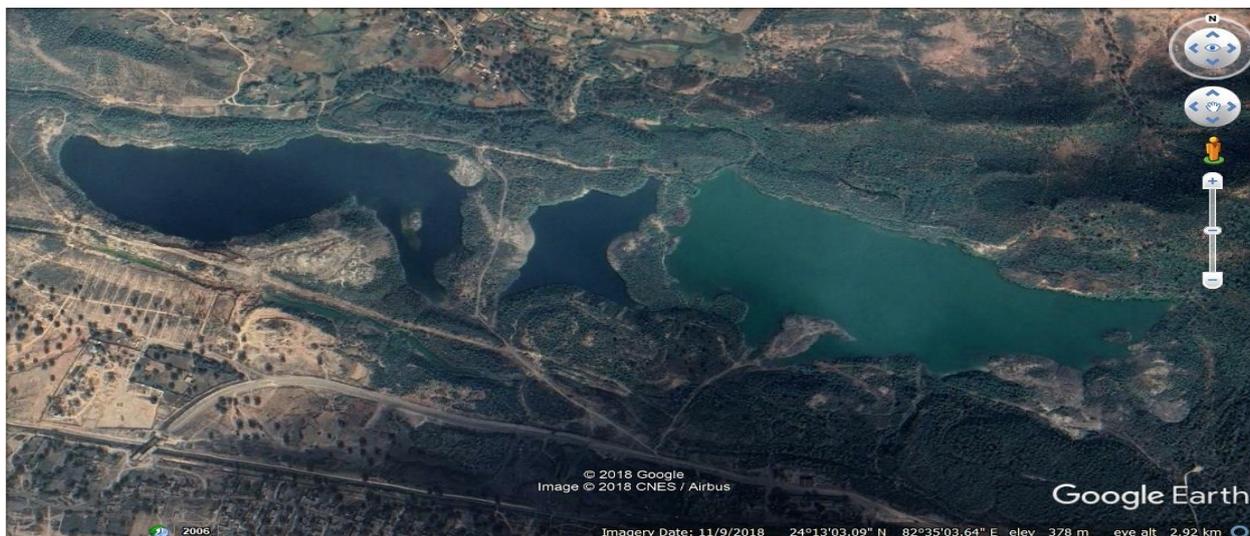


Figure 2. The Gorbi abandoned mine, showing Pits I, II and III (via Google Earth).

Table 1. Details of examined pits.

Sl. No.	Pit No.	Pit Area in Ha	Depth	Water Volume
1	I	40.00	50 m *	20.00 million m ³
2	II	8.00		4.00 million m ³
3	III	26.00		13.00 million m ³
		74.00		37.00 million m ³

* Approximate depth was considered.

2.2. Fly Ash Characterization

The fly ash samples were air-dried for 2 to 3 days in a drying oven (Thermo Scientific Heraeus VT6060P) and mixed by the coning and quartering method, obtaining a representative sample for analysis [31]. Mineral identification in fly ash was characterized using X-ray fluorescence (XRF), X-ray diffraction (XRD) and scanning electron microscopy (SEM).

X-ray fluorescence (XRF) was used for the nondestructive chemical analyses of the fly ash. Mineral phase recognition by X-ray diffraction method depends on the diffraction peaks at different 2θ values relating to the mineral's d-spacing value. The X-ray plot of a diffraction angle (2θ) versus intensity of radiation reveals the interplanar spacing and, in turn, the type of mineral present.

X-ray diffraction (XRD) of oven-dried fly-ash samples of less than $75\ \mu$ was carried out to detect the mineral composition of its various mineral phases. Mineral phase recognition by XRD depends on the diffraction peaks at different 2θ values relating to the mineral's d-spacing value. According to Bragg's law, as each mineral has a highly ordered crystalline structure, its d-spacing values are characteristics of a set of diffraction peaks. AN Ultima IV (Austin, TX 78717, USA) with a Cu K-alpha central instrumentation facility was used in the current work. The scattering angle 2θ ranged from 5° to 85° at a scanning rate of $5^\circ/\text{min}$ with a step size of 0.05° , and the target used was a Cu-Ka ($\lambda = 1.5418\text{\AA}$) target.

Furthermore, the fly ash samples were examined under high-resolution scanning electron microscopy (JOEL model JSM-6480 LV, Tokyo, Japan) to understand their surface

micro-morphology. Since fly ash is nonconductive, it was coated with a platinum layer for 180 s as needed for a current of 50 mA at vacuum before acquiring the SEM micrograph.

2.3. Neutralization Experimental Setup

For the experiment, 1000 mL of AMD (pH 2.54) were collected into containers. Four sets of AMD were prepared in separate beakers. The AMD sample's physicochemical characteristics were measured according to Indian standard guidelines, as mentioned [27–30]. In brief, fly ash samples (100 g; one from each plant) were added to the beakers. Each sample was subjected to mine water leaching for 35 days. Hence, four different types of leachate samples were produced. Finally, the leachate samples were collected and filtered (Whatman Grade 42 filter paper) into sampling bottles and assessed for the aforementioned physicochemical parameters.

3. Results and Discussion

3.1. XRF Analysis of Fly Ash

The major minerals phases in the fly ash samples were found to be SiO_2 , Al_2O_3 , CaO , MgO , MnO , Na_2O , K_2O and FeO , as shown in Table 2. Different percentages of the minerals were found in each of the four fly ash samples. The Anpara TPP sample showed the highest concentration of Al_2O_3 and CaO (and K_2O), and Shaktinagar TPP showed the highest concentration of SiO_2 (and MnO). Na_2O , K_2O , CaO and MgO can be considered as acid-neutralizing groups, and Al_2O_3 and F_2O_3 as amphoteric groups.

Table 2. Characteristics of the fly ash samples.

Sampling Location	Parameters							
	SiO_2	Al_2O_3	CaO	FeO	MgO	MnO	Na_2O	K_2O
Concentration in %								
Anpara TPP	45.9	27.7	17.4	3.2	2.9	1.6	0.6	0.7
Renusagar TPP	40.5	23.0	16.1	8.1	8.0	2.2	1.4	0.7
Shaktinagar TPP	44.7	26.3	15.6	7.9	1.7	2.1	1.3	0.4
Vindhyachal TPP	44.0	20.7	14.9	8.4	5.8	4.8	1.2	0.2

Note: TPP = thermal power plant.

The following chemical reactions were expected after mixing with AMD:

- (i) Silica (SiO_2) is stable below 870 °C. It will not react with any base or acid except hydrofluoric acid. Hence, SiO_2 will not have an impact on AMD.
- (ii) Alumina (Al_2O_3) will also not have any impact on AMD, at least at room temperature.
- (iii) Hematite (Fe_2O_3) will have negligible impact on AMD, up to the most extreme temperature and pressure found in the field.
- (iv) Lime (CaO) present in fly ash will react with H_2O to give $\text{Ca}(\text{OH})_2$.
- (v) Magnesia (MgO) present in fly ash will react with H_2O to give $\text{Mg}(\text{OH})_2$, which will also neutralize acidic solutions
- (vi) Na_2O will react with H_2O to give NaOH .
- (vii) K_2O will react with H_2O to give KOH .

Thus, considering the ability of CaO , MgO , Na_2O and K_2O to provide hydroxyl radicals in the solution, the acid-neutralization potential of fly ash may be calculated. The amount of SO_3 in the fly ash was only 0.18%, and for the sake of simplicity, its effect may be disregarded [32].

3.2. XRD Analysis of Fly Ash

The mineral phase resulting peaks (Figure 3) were compared to the Joint Committee on Powder Diffraction Standards (JCPDS) data. The crystalline phases present in the fly ash consisted of quartz (SiO_2), mullite (Al_6SiO_2), hematite (Fe_2O_3) and calcium oxide (CaO). Similar results (material rich in quartz, mullite, hematite and quicklime) were noted in

the XRD analysis of a fly ash sample from a coal-fired boiler's electrostatic precipitators, generally in bituminous fly ashes [22].

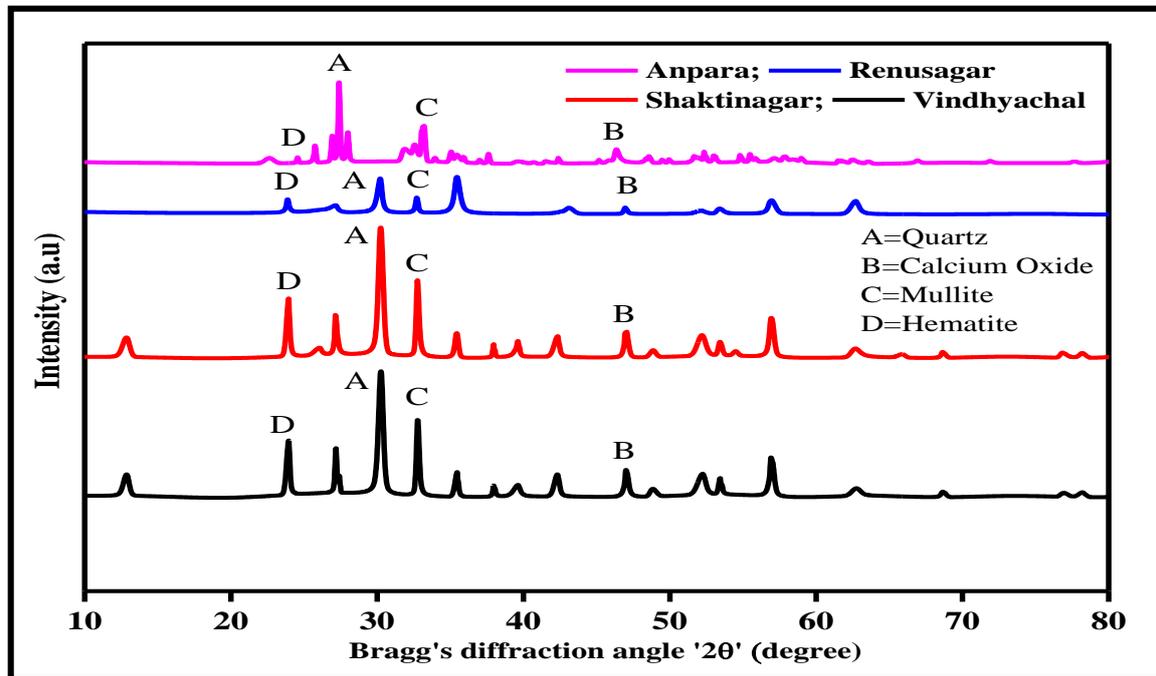


Figure 3. X-ray diffractograms of selective fly ash samples from various thermal power plants using the abbreviations for minerals followed by Kretz [33].

3.3. SEM Analysis of Fly Ash

SEM images of 2 mm size fly-ash particles are shown in Figure 4. Large particle agglomerations were frequently seen, and several spherical particles, together with irregularly shaped particles, were also observed. The rounded particles were mostly glassy, while the angular particles were typically made of crystalline solids such as quartz, mullite, magnetite, hematite, etc. A similar composition was noted for Canadian [25] and British coal fly ash samples [34], with their crystalline portion comprising mainly quartz and mullite. Both ashes mainly consisted of an amorphous phase; here, the amorphous phase was mostly dominant in the Shaktinagar and Renusagar fly ashes (Figure 4).

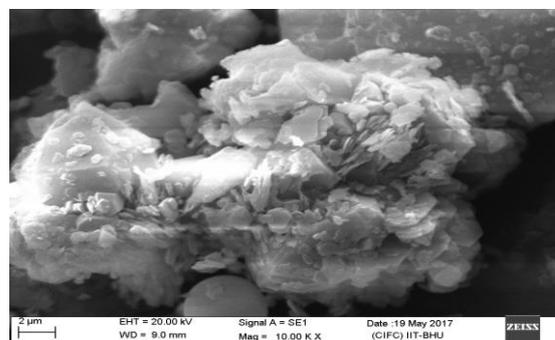


Figure 4. Cont.

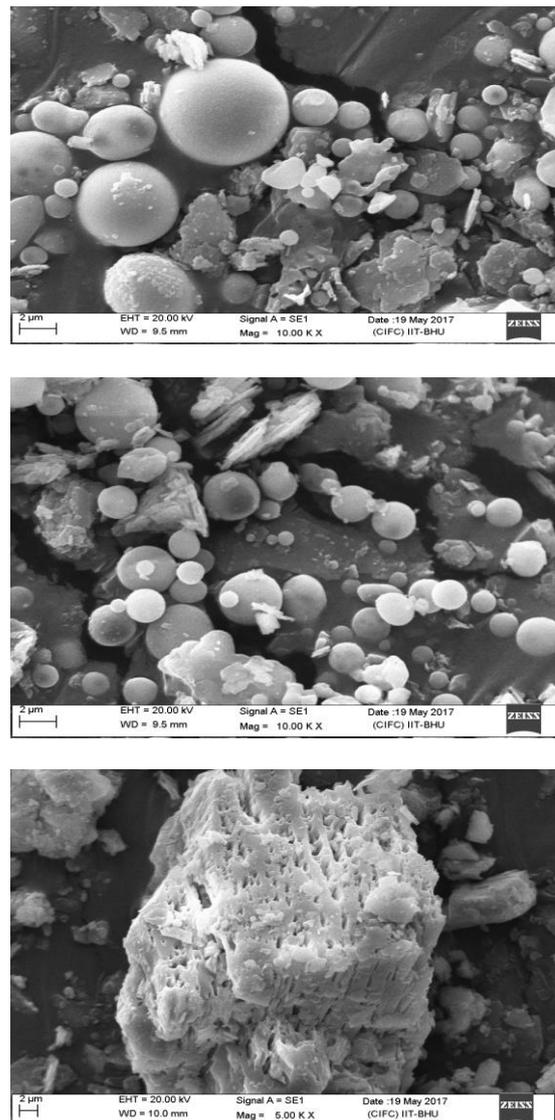


Figure 4. SEM images (2 µm) of fly ashes from Anpara, Shaktinagar, Renusagar and Vindhyachal TPPs.

3.4. AMD Quality Analysis and Neutralization Potential

In the present research, AMD analysis revealed low pH, high TDS concentrations, electrical conductivity and various elemental ions (Table 3). Considerable neutralization of AMD was achieved with the addition of fly ash, as shown in Table 4. Fly ash showed a pH of 12, and as such the pH of AMD was elevated from 2.5 to 6.0 within 35 days (Figure 5). There was a decreasing trend in all chemical species of the leachate. The pH remained relatively constant after 35 days of the experiment. These results may be attributed to the fly ash Ca-bearing minerals [23]. It has also been postulated that CaO reacts with the other siliceous and aluminous materials of the fly ash and forms cementitious complexes that stabilize AMD's leaching elements [25]. Comparison of the four samples showed that the highest CaO content was found in the Anpara TPP fly ash. However, silicates and aluminum and ferrous oxides also contribute to acid buffering at low pH [23].

Electrical conductivity and TDS of the treated AMD were reduced. All of the leachates showed an initial decrease in the sulfate concentrations. This was due to the formation of CaSO_4 upon reaction with the fly ash. Al concentration decreased from 33.7 mg L^{-1} to 21.0 mg L^{-1} . It was believed that the aluminum reacted with the silica in the ash and formed more mullite. In general, the heavy-metal concentrations decreased. B, Ba, Be, Cd, Co, Cr, Cu, Mn, Ni, Pb, Sr and Zn concentrations were all low and decreased to undetectable

levels. In brief, after the described laboratory experiment, the physicochemical parameters of the AMD from the abandoned mine were under the permissible limits for release in water catchments.

Table 3. Physicochemical characteristics of the AMD water.

Parameter	Unit	Concentration
pH (AMD water)	–	2.5
pH (Fly ash)	–	12.0
EC	μS/m	2990
TDS	ppm	2270
Acidity (CaCO ₃)		3097
Na		200
Mg		45
Al		33.7
K		43.5
Ca		158
Fe		166
Fe ²⁺		145
Fe ³⁺		156
SO ₄ ²⁻		190
NO ₃ ⁻		45
Al	mg/L	37.5
B		0.07
Ba		0.02
Be		<0.005
Cd		0.05
Co		0.58
Cr		<0.005
Cu		0.06
Mn		45
Ni		0.70
Pb		0.05
Sr		0.74
Zn		3.0

Table 4. Physicochemical characteristics of the AMD after addition of fly ash.

Parameter *	Unit	Thermal Power Plant			
		Anpara	Renusagar	Shaktinagar	Vindhyachal
Concentration					
pH	–	6.0	6.0	6.0	6.0
EC	μS/m	765.0	780.0	735.0	752.0
TDS	ppm	682.0	675.0	695.0	672.0
Na		20.0	15.0	21.0	19.0
Mg		53.0	54.0	51.0	49.0
Al		29.0	21.0	27.0	25.0
K		12.0	10.0	9.0	13.0
Ca	mg/L	55.0	49.0	53.0	52.0
Fe		3.1	4.5	2.1	4.7
Fe ²⁺		32.0	32.0	30.0	31.0
Fe ³⁺		21.0	20.0	19.0	20.0
SO ₄ ²⁻		32.0	29.0	24.0	50.0
NO ₃ ⁻		15.0	12.0	24.0	33.0

* All other species below limit of detection.

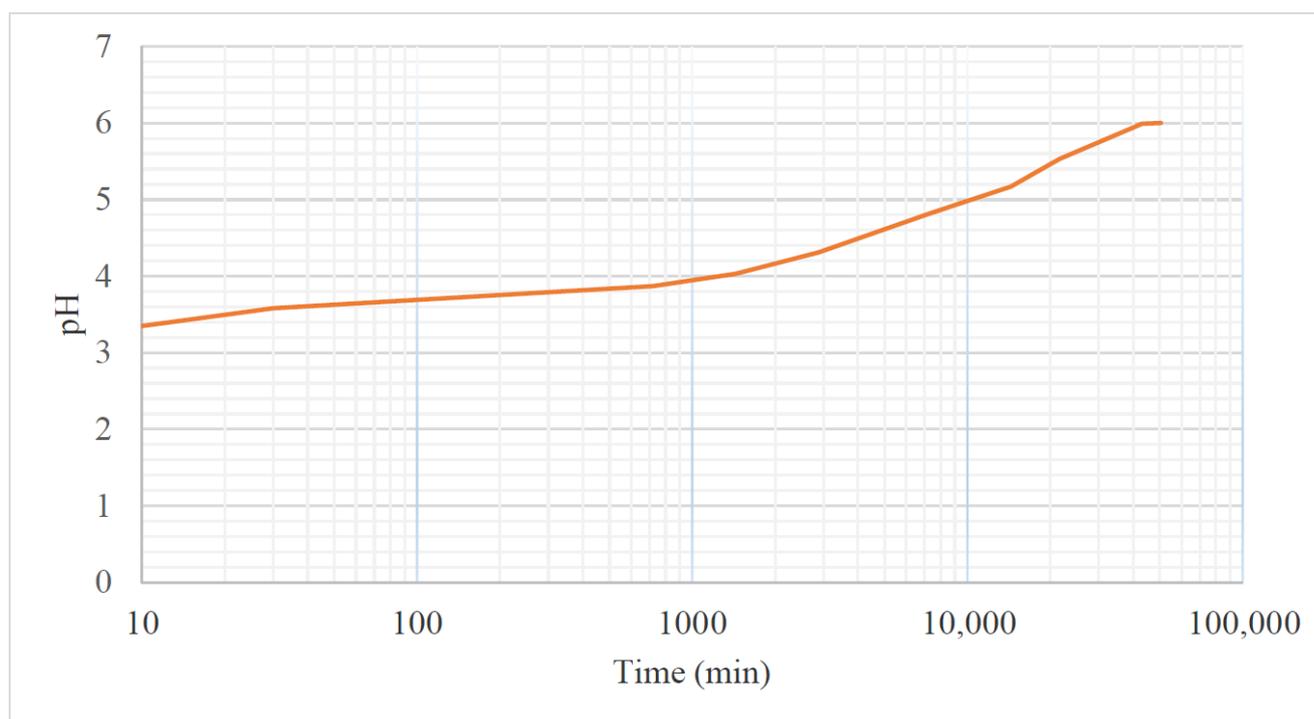


Figure 5. Variation of pH in relation to duration treatment of the AMD with fly ash.

The amount of fly ash required to reach acceptable limits is approximately 4–5 times higher than the lime requirement; therefore, high amounts of fly ash can be disposed of. It should be noted that fly ash may contain metals of toxicological concern, such as Cd, Cr, Co, Ni and Pb [35]; these metals may leach into the environment during the AMD neutralization process, creating additional problems. If high amounts of fly ash are used, more metals are released into the environment [23]. So, only 30% of the produced fly ash was used in this experiment. Nevertheless, these elements' leaching can be limited by maintaining near-neutral pH conditions and adding materials that can absorb and adsorb them. It is known that the mine waste material itself can achieve the latter [22]; furthermore, low AMD pH is counterbalanced by high fly ash pH, and vice versa [34], so near-neutral conditions can be achieved. The remaining materials (treated AMD) may be processed further (for example, with zeolite), and the solid residue may be used as a mine backfill material for a true zero-waste environmental management scheme [24].

4. Conclusions

AMD neutralization by optimized amounts of fly ash may become very useful in terms of the safe and economic disposal of waste, especially when thermal power stations and coal mines are near each other. As shown in the present research, AMD pH was raised considerably after the addition of fly ash. Metal concentrations and sulfate ions were reduced, and AMD physicochemical characteristics were improved. This study concludes that fly ash may be carefully placed in abandoned mine pits containing AMD and neutralize this acidic liquid waste. Nevertheless, additional research should focus on the new waste created, its stability, its leaching potential and its general environmental impact.

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