



Fate of the naturally occurring radioactive materials during treatment of acid mine drainage with coal fly ash and aluminium hydroxide



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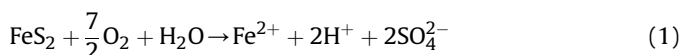
ABSTRACT

Mining of coal is very extensive and coal is mainly used to produce electricity. Coal power stations generate huge amounts of coal fly ash of which a small amount is used in the construction industry. Mining exposes pyrite containing rocks to H₂O and O₂. This results in the oxidation of FeS₂ to form H₂SO₄. The acidic water, often termed acid mine drainage (AMD), causes dissolution of potentially toxic elements such as Fe, Al, Mn and naturally occurring radioactive materials such as U and Th from the associated bedrock. This results in an outflow of AMD with high concentrations of sulphate ions, Fe, Al, Mn and naturally occurring radioactive materials. Treatment of AMD with coal fly ash has shown that good quality water can be produced which is suitable for irrigation purposes. Most of the potentially toxic elements (Fe, Al, Mn, etc) and substantial amounts of sulphate ions are removed during treatment with coal fly ash. This research endeavours to establish the fate of the radioactive materials in mine water with coal fly ash containing radioactive materials. It was established that coal fly ash treatment method was capable of removing radioactive materials from mine water to within the target water quality range for drinking water standards. The alpha and beta radioactivity of the mine water was reduced by 88% and 75% respectively. The reduced radioactivity in the mine water was due to greater than 90% removal of U and Th radioactive materials from the mine water after treatment with coal fly ash as ThO₂ and UO₂. No radioisotopes were found to leach from the coal fly ash into the mine water.

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1. Introduction

Acid mine drainage (AMD) is the second major threat to sustainability after global warming. Acid mine drainage is a product of FeS₂ oxidation in the presence of O₂ and H₂O to produce H₂SO₄ followed by enhanced chemical weathering of the resultant bedrock (Lottermoser, 2007).



The resultant water is laden with potentially toxic elements such as Fe, Al, Mn, etc. If the surrounding bedrock contains

radioactive elements such as U and Th, the resultant AMD will contain elevated concentration of radioactive elements (Scott, 1995; Durand, 2012).

Mine water treatment is complex and very expensive. High costs associated with mine water treatment are due to the complexity and diversity of mine water composition, which means there is no “one-fits-all” treatment option for mine water treatment. Mine water treatment options can be broadly classified as passive or active methods (Lottermoser, 2007; Wieder and Lang, 1982; Neculita et al., 2007; Steed et al., 2000; Hedin et al., 1994; Johnson and Hallberg, 2005; Pulles et al., 1992; Bosman et al., 1990; Hlabela et al., 2007; Bologo et al., 2012; Bosman, 1983; Smit and Sibilski, 2003). The disadvantages of passive treatment of mine water are; the recovery of treated water is very limited, requires extensive land area to accommodate high flow and/or highly contaminated mine water and lastly the quality of process

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water is not guaranteed since the process is not monitored frequently. The major disadvantage of active treatment method is that it requires continuous input of energy, reagents and the need of skilled manpower to run and maintain the treatment plant makes these techniques expensive.

Treatment of AMD with fly ash (FA) could reduce the costs of mine water treatment. This is because FA is waste material from coal fired power stations. Fly ash is capable of neutralising acidity and precipitating out most of the potential elements to acceptable limits for irrigation or even potable water quality (Gitari et al., 2008; Madzivire et al., 2011; Surender, 2009). One drawback that might limit the coal FA treatment technology is the fact that FA contains elevated concentrations of radioactive materials compared to the parent coal (Baykal and Saygili, 2011; Papastefanou, 2010; Peppas et al., 2010; Turhan et al., 2010; USGS, 1997). In addition, some AMDs have been found to contain radioactive materials in proportions much higher than the acceptable limits for irrigation and drinking purposes.

This study investigates the activity levels of radioactive materials present in the FA and AMD samples. The radioactivity of solid residues and product water after treatment of mine water with coal FA were also evaluated. The chemistry of the removal of the radioactive nuclides during treatment of mine water was also elucidated using Act2 program of the Geochemist's workbench software (GWB).

2. Materials and methods

2.1. Sampling and characterization of mine water

Mine water was collected from a gold mine in the Witwatersrand goldfields of South Africa. The water was filtered through a 0.45 μm pore filter membrane. The cation samples were preserved with 2 drops of concentrated HNO_3 for every 100 mL of sample. Both anion and cation samples were preserved at 4 °C analysed using ion chromatography (IC) and inductively coupled plasma–optical emission spectroscopy (ICP–OES) respectively. Mine water samples to be analysed for radioactivity were filtered through 8 μm pore filter membrane and 0.45 μm pore filter membrane to remove coarse materials and suspended solids. The samples were then acidified to ensure radioactive materials were not adsorbed on the container walls and then analysed using alpha and gamma-ray spectrometry to determine alpha and beta radioactivity and the radioactive materials.

2.2. Sampling and characterization of fly ash

Coal FA was collected directly from the hoppers of a coal power station in Mpumalanga Province in South Africa. Samples were sealed in plastic bags devoid of air to avoid the reaction of CaO in the FA with CO_2 which would cause the formation of CaCO_3 therefore reducing the CaO content. The FA samples were analysed using quantitative X-ray diffraction (QXRD) spectroscopy to determine the percentage mineral composition. Fly ash samples were dried overnight in an oven at 105 °C. The samples were then milled to obtain a homogeneous powder so that representative portions could be sampled for the various analyses. The homogenized sample (500 g) was placed in Marinelli beakers and analysed for gross alpha and beta to obtain a first order estimate of the total activity of the sample. After determination of the gross alpha and beta the samples were analysed for various radioisotopes using low energy gamma analysis and high energy gamma analysis using a method adopted from Newman et al. (2008) and ^{226}Ra was determined by measuring its decay products. A three week waiting period was allowed to establish secular equilibrium of ^{226}Ra and its decay products considering the 3.82 day half-life of radon (^{222}Rn) gas in the series.

2.3. Treatment of mine water with coal fly ash

Gold mine water (80 L) and coal FA (13 kg) were mixed together using a jet loop reactor with jet nozzle sizes set at 8 mm. After 30 min, 86.58 g of $\text{Al}(\text{OH})_3$ was added to the mixture. The pH, EC and temperature were measured after 15 min and aliquot samples were collected after every 30 min for 150 min. This procedure was chosen based on the optimization of the treatment of neutral mine drainage with coal FA and $\text{Al}(\text{OH})_3$ by Madzivire et al. (2012). Samples were filtered using a 0.45 μm pore filter membranes and analysed using ICP–OES, IC, alpha and gamma spectroscopy. The radioactivity characteristics of the mine water FA and the solid residues were analysed to determine the fate of the radioactive materials. The chemistry involved in the removal of different radioactive materials was determined using the Act2 program of the GWB in the presence of elements were chosen with concentration greater than 10 mg/L. The independent variable was chosen as $\log_a\text{Ca}^{2+}$ and the dependent variable was the pH. These values were chosen based on the fact that treatment of mine water with coal FA was based on the neutralization of pH due to the dissolution of the CaO fraction in coal FA.

3. Results and discussion

3.1. Characterization of mine water

Major precious minerals that were mined in the Witwatersrand goldfields were Au and U (Cole, 1998). These minerals exist in combination with FeS_2 and $\text{MgCa}(\text{CO}_3)_2$. The characteristics of the gold mine water collected are shown in Table 1.

The pH of the mine water was 2.65 which was out of the target water quality range (TWQR) for potable and irrigation water. According to Lottermoser (2007), the mine water can be classified as

Table 1
The physicochemical parameters of the mine water.

Parameter ^a	Raw mine water	Irrigation limit (DWAF, 1996)	Potable limit (WHO, 2011)
pH	2.65 ± 0.6	6.5–8.4	6–9
EC	2292 ± 36	NA ^b	0–700
Sulphate	2562.41 ± 6.85	NA	0–500
Fe	201.05 ± 0.55	0–5	0–0.1
Al	26.63 ± 0.29	0–5	0–00.15
Ca	360.15 ± 4.25	NA	0–32
Mg	153 ± 0.7	NA	0–30
Mn	60.16 ± 0.17	0–0.02	0–0.05
Ni	2.11 ± 0.0043	0–0.2	NA
Zn	1.93 ± 0.013	0–1	0–0.5
Sr	0.45 ± 0.0034	NA	NA
Cu	0.28 ± 0.0033	0–0.2	0–1
U	0.28 ± 0.001	0–0.01	0–0.03
Li	0.069 ± 0.00029	0–2.5	NA
Se	0.061 ± 0.0023	0–0.02	0–0.02
Ba	0.026 ± 0.00043	NA	0–0.7
Cr	0.023 ± 0.00029	0–0.1	0–0.05
Th	0.018 ± 0.00031	NA	NA
Pb	0.0075 ± 0.000017	0–0.2	0–0.01
Cd	0.0068 ± 0.000012	0–0.01	0–0.003
As	0.0056 ± 0.000025	0–0.1	0–0.001
Be	0.0039 ± 0.000042	0–0.1	0–0.012
V	0.0012 ± 0.000076	0–0.1	0–0.01
Mo	0.00053 ± 0.000024	0–0.01	0–0.07
B	0.00023 ± 0.000004	0–0.5	0–2.4
Hg	0.0000039 ± 0.0000012	NA	0–0.001

^a All units are mg/L except that of EC ($\mu\text{S}/\text{cm}$) and pH.

^b NA means not applicable.

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