# Radioactivity of mine water from a gold mine in South Africa

G. Madzivire[1], P. Maleka[2], R. Lindsay[3] & L. F. Petrik[1]
[1]*ENS Group, Chemistry Department,*
*University of the Western Cape, South Africa*
[2]*Department of Environmental Radioactivity,*
*iThemba Labs, South Africa*
[3]*Physics Chemistry Department,*
*University of the Western Cape, South Africa*

## Abstract

Gold mining in South Africa has occurred for over 100 years in the Witwatersrand goldfields. The associated geology contains 3% pyrite ($FeS_2$) together with radioactive elements such as uranium and thorium. Mining of gold in the Witwatersrand basin leaves $FeS_2$ exposed to $H_2O$ and $O_2$ in mine tailing and mine voids. This resulted in the formation acid mine drainage (AMD) due to the oxidation of $FeS_2$ in the presence of $H_2O$ and $O_2$. The acidity generated from $FeS_2$ caused chemical weathering of the surrounding tailings or bedrock. The mine water from the mine voids and mine tailings in the basin is acidic and contains elevated concentration of radioactive elements such as U, Th, Ra and Pb in addition to heavy metals such as Fe, Al, Mn, Ca and Mg. The radioactivity of the mine water determination using alpha and beta spectrometry indicated that mine water from the gold mine had alpha and beta radioactivity 6 and 12 times greater than the target quality water range (TQWR) for potable water respectively.

*Keywords: mine water, radioactive elements, alpha and beta radioactivity and target water quality range, pyrite.*

## 1 Introduction

The geology that contains pyrite ($FeS_2$) together with radioactive containing elements such as U and Th could form mine drainage with radioactive materials.

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This is because when $FeS_2$ is oxidized in the presence of $H_2O$ and $O_2$, it forms acidic water that in turn dissolves the radioactive containing minerals. Radioactivity is the disintegration of the nucleus of an unstable atom by emitting particles containing ionization energy. Three type of radiation particles are alpha, beta and gamma.

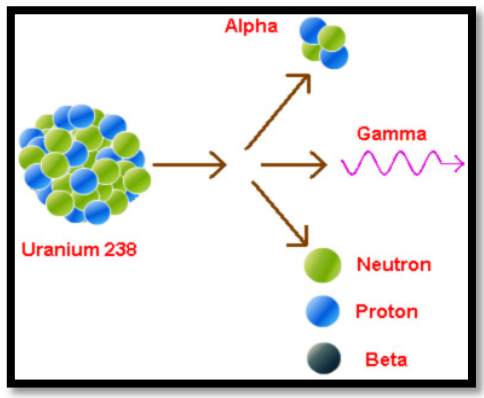


Figure 1: Schematic representation of different types of radioactive decay of an unstable atom [1].

Alpha particles are produced when the nucleus of an unstable atom loses two protons and two neutrons (He nucleus), while beta radiation occurs when a nucleus of an unstable atom loses either a positron or an electron. Gamma radiation is produced when the nucleus that was left in an excited state after alpha or beta decay loses excess energy to attain a stable state. An alpha particle is positively charged, the beta particle is neutral while the gamma particle can be negative (electron) or positive (positron). Different radioactive particle have different penetrating potential. An alpha particle can be stopped by a sheet of paper, while a beta particle can penetrate a sheet of paper but cannot pass through an aluminium foil. The gamma particle can pass through an aluminium foil but can be reduced significantly by a thick block of lead.

Mine water from Au and U mines is usually contaminated with radioactive elements such as U and its decay products such as Ra and Th. Naturally, the most abundant isotope is $^{238}U$ (99.27%) with a half-life of $4.5 \times 10^9$ years. Other isotopes that exist in nature are $^{235}U$ (0.72%) and $^{234}U$ (0.006%). The half-life of $^{235}U$ and $^{234}U$ are $7.04 \times 10^8$ and $2.46 \times 10^5$ years respectively [2]. Uranium exists in various oxidation states of +2, +3, +4, +5 or +6). The most stable oxidation states are +4 and +6. The uranium (IV) state mainly exists as species which are highly insoluble in the natural environment and therefore are generally far less mobile than U (VI). In nature Th exists mainly as $^{232}Th$ with half-lives of $1.4 \times 10^{10}$ years [3]. The radioactive decay series of U and Th form various nuclides and the end product is a stable $^{206}Pb$ isotope shown in fig. 2.

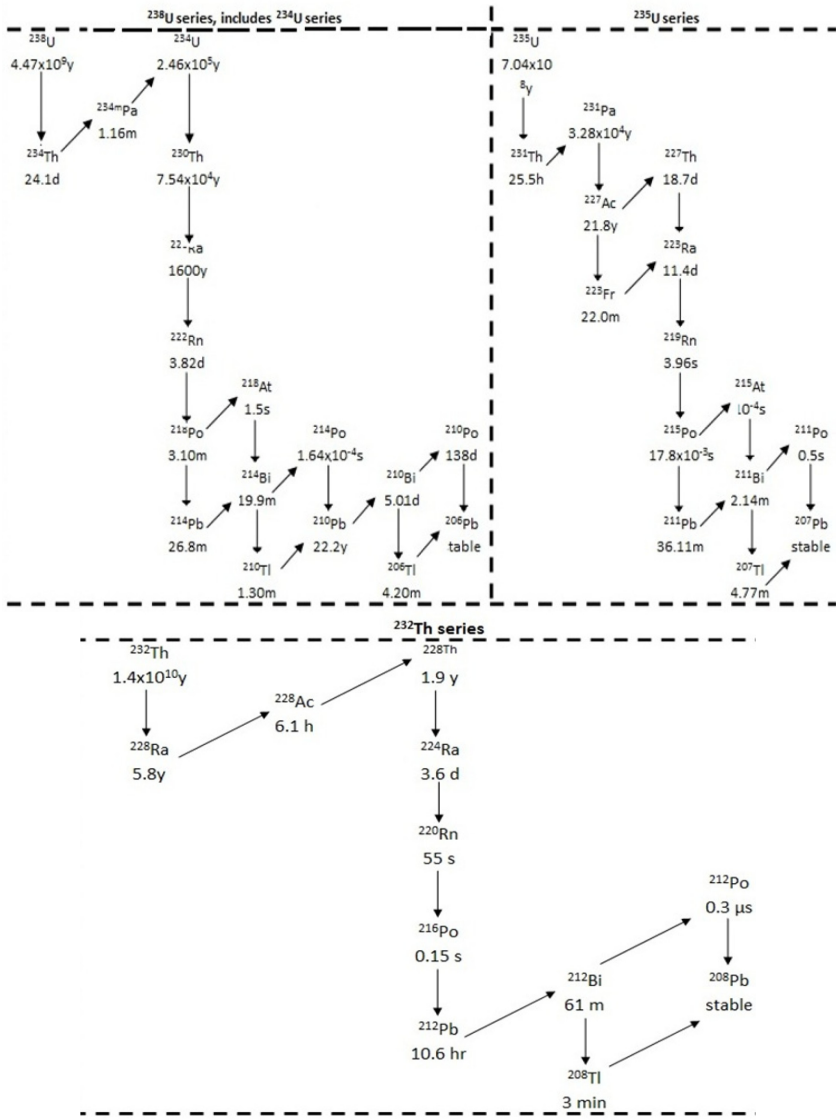


Figure 2: The radioactive decay series of $^{238}U$, $^{235}U$ and $^{232}Th$ (Arrows pointing downwards represent an alpha decay and the arrows pointing upwards represent beta decay) [4].

The varied decay intermediates have different geochemical properties and therefore are fractionated into different geological environments. In acidic aqueous media, the chemistry of U and Th generally exist as free cations. Each isotope in the $^{238}U$, $^{235}U$ and $^{232}Th$ decay series has a unique fingerprint of alpha and gamma decay energies that can be used to identify and quantify each

radioisotope [2]. The height of a peak with particular energy can be used to quantify the radionuclides in the samples based on the calibration with known concentration of specific radionuclides.

### 1.1 Guidance levels for radioactive nuclides in drinking water

Radioactivity species identification in drinking water is a very expensive and sophisticated process. The screening steps used to identify if the water is suitable for drinking purposes in terms of radioactivity is as shown in fig. 3 below.

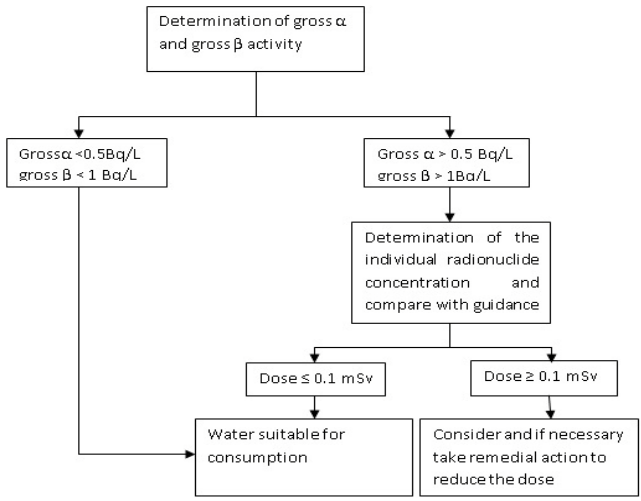


Figure 3: The outline of the screening process for the suitability of drinking water in terms of radioactivity [5].

The first step before determining the concentration of individual radionuclides in water is to determine the gross alpha and gross beta activity of the water. If the gross alpha and beta are less than 0.5Bq/L and 1Bq/L respectively, the water will be suitable for drinking in terms of radioactivity [5]. On the other hand if the gross alpha and beta activities are greater than 0.5 and 1Bq/L respectively then the concentration of the individual radionuclide should be measured and compared to the World Health Organization (WHO) guidelines. If the dose is at most 0.1mSv, the water is suitable for drinking. If the dose is greater than 0.1mSv then remedial action should be undertaken. A dose of at most 0.1mSv is achieved if the following equation is satisfied:

$$\sum_i \frac{C_i}{GL_i} \leq 1 \tag{1}$$

where $C_i$ = measured activity of radionuclide i, and $GL_i$ is the guideline concentration of radionuclide at an intake of 2 liters per day for one year. This will result in an effective dose of 0.1mSv per year [5].

# 2 Methodology

## 2.1 Sampling and characterization on the gold mine water

The mine water used in this study was collected from a gold mine in the Western Basin in Gauteng Province (fig. 4). The water was filtered through a 0.45µm pore membrane filter paper using a manual pumping device. The filtered samples were divided into two portions of 100mL each for cation and anion analysis. The cation samples were preserved with 2 drops of concentrated $HNO_3$ (55%) for approximately 100mL of sample. Both cation and anion samples were preserved at 4°C until analysis for anions using ion chromatography (IC) and cations using inductively coupled plasma–optical emission spectroscopy (ICP-OES).


Figure 4: The Google earth image of the area where the acids mine drainage was sampled.

The mine water was also filtered through a 0.45µm filter paper and the acidity was determined using a Metrohm Autotitrator. Mine water samples that were supposed to be analysed for radioactivity were first filtered through 8µm and 0.45µm to remove coarse materials and suspended solids. The samples were then acidified to ensure radionuclides are not adsorbed on the container walls.

## 2.2 Determination of alkalinity

The alkalinity of mine water used in the experiments was determined to gain an understanding of the acid neutralizing potential. This parameter is very important for cation/anion balance in Geochemist's workbench geochemical modeling.

Acidity was determined by titrating AMD (20mL) sample with 0.1M NaOH to an end point of 8.3. The acidity was calculated as follows:

$$mg \cdot L^{-1}(CaCO_3) = \frac{V(NaOH) \times [NaOH] \times 1000}{V(sample)} \quad (2)$$

where V = mL and [ ] = mol/L.

### 2.3 Radioactivity analysis of the gold mine water mine water

The gold mine water samples were filtered, acidified and then stored at 4°C before analysis. Before the samples were analysed using alpha spectrometry, they were first pre-treated and prepared specifically for analysis of a specific element. The following steps cover how the samples were prepared and analysed using alpha spectrometry.

#### 2.3.1 Sample preparation for alpha spectrometry determination of U

The method was based on solid phase extraction of uranium from water samples, with detection of the uranium isotopes by alpha spectrometry. An aliquot (200mL) of the sample was measured into a beaker and uranium-232 internal tracer was added for recovery determination. The sample was evaporated to a volume of 10mL, and acidified using 2 drops of 2M $HNO_3$. The sample was loaded on a TruSpec (Eichrom resins) column to absorb uranium. A mixture of HCl and HF acid was added to the column to elute uranium. The separated uranium was loaded on a cation exchange column to further purify uranium. Uranium was eluted with concentrated 2M HCl and collected on a filter paper by lanthanum fluoride micro co precipitation.

#### 2.3.2 Sample preparation for alpha spectrometry determination of Th

The method was based on solid phase extraction of thorium from water samples with detection of the different thorium isotopes by alpha spectrometry. Thorium-229 was added as an internal tracer to a 200mL aliquot of the sample. Sample volume was reduced to a volume of 10mL by evaporation and acidified using 2M $HNO_3$. The sample was loaded on a TruSpec column (Eichrom resins) to absorb thorium and other nuclides. Thorium is eluted with a mixture of HCl and HF. The collected eluent was loaded on a cation column for further purification. Thorium was finally eluted with 2M $H_2SO_4$ solution and collected on a filter paper by lanthanum fluoride micro co precipitation.

#### 2.3.3 Sample preparation for alpha spectrometry determination of Ra

Barium-131 was added as an internal tracer to the sample. Radium was separated from the bulk of the sample material by adding Pb and Ba carriers to the sample followed by precipitating these elements and radium present as sulphate ions. The precipitate is purified from other ions by washing, and then dissolved and by adding a complexing agent. Barium (and radium) was again precipitated and filtered as sulphate while Pb was kept in solution by careful adjustment of the pH. Barium-131 in the separated fraction is measured for on a radiation detector for yield determination.

### 2.3.4 Sample preparation for alpha spectrometry determination of Pb and Po

Polonium-209 was added as internal tracer for recovery determination. The water sample was acidified with 2M HCl and heated to a temperature of 90°C. A silver disk was added to the water sample and the solution was stirred for several hours to induce spontaneous deposition of polonium-210. A reducing agent was added at the start of the process to prevent iron from plating on the silver disk. The disk was removed, washed and air-dried to prepare it for measurement on the alpha spectrometer.

### 2.3.5 Alpha spectrometry analysis of the prepared samples

Prepared samples were counted on Canberra Alpha Analyst or Alpha Apex systems. These systems consist of 12 vacuum chambers each hosting an alpha PIPS detector. Samples were counted for a period of 24 hours to reach the required detection limits. The acquired spectra were analysed for counts collected in the respective alpha peak positions shown in table 1.

The peaks positions were chosen based on the highest intensities of the alpha energy of the respective decay of the nuclide [2]. These counts were entered into data reduction programs to calculate nuclide activities, associated uncertainties and minimum detectable activity concentrations. A control sample containing known amounts of the analyte nuclides were used to calibrate the machine for each batch of samples.

Table 1: Peaks position used for identification of nuclides using alpha spectrometry.

| Radioisotope | Peaks position (MeV) |
|---|---|
| $^{238}$U | 4.72 |
| $^{235}$U | 4.40 |
| $^{234}$U | 4.77 |
| $^{232}$Th | 4.01 |
| $^{230}$Th | 4.69 |
| $^{227}$Th | 5.76, 5.93 and 6.04 |
| $^{228}$Th | 5.42 |
| $^{226}$Ra | 4.78 |
| $^{224}$Ra | 5.68 |
| $^{210}$Po | 5.30 |

Measured activities must lie within prescribed limits for the results of the batch to be accepted. The yield for a sample must also be within a range. Background, energy, peak width and efficiency checks were performed on a weekly basis to ensure correct operation of detectors.

## 2.4 Geochemist's workbench modeling

The Geochemist's Workbench (GWB) is a software that is comprised of different programs to manipulate chemical reactions, calculate stability diagrams and the equilibrium states of natural waters, trace reaction processes, model

reactive transport, plot the results of these calculations, and store the related data [6]. The software is constantly upgraded. In this study SpecE8 program of the GWB 8.0 essential software were used. SpecE8 is capable of calculating the species distributions in aqueous solutions, mineral saturation indices and gas fugacities. SpecE8 can also account for sorption of species onto mineral surfaces according to a variety of methods, including surface complexation and ion exchange [6].

The Spec8 program of the GWB software was used to calculate the species distribution and the saturation indices of the different minerals in the mine water. This was done using the pH, electrical conductivity (EC), alkalinity and acidity of the mine waters and the concentration of individual ions. The pH and electrical conductivity (EC) of the mine water was measured in the field. Alkalinity was determined using a Metrohm Autotitrator. The elemental composition of the mine water was obtained using ICP-OES and IC.

## 3 Results and discussion

The gold mine water was collected from the Western Rand basin in Witwatersrand Goldfields, South Africa. The mine is a semi abandoned mine since the mine tailings are being reworked and no underground mining is taking place anymore. The pH, EC and total dissolved solids (TDS) were measured onsite. The chemical composition of the samples were analysed using IC and ICP OES and the results obtained are shown in table 2.

The pH of the gold mine water was 3.48, which was not within the target water quality range (TWQR) as shown in table 2. According to Morin and Hutt [8], the gold mine water can be classified as acid mine drainage (AMD) because the pH was below 6. The gold mine water had a high concentration of Fe and Al. Typically AMD contains more of the combined concentration of Fe, Al and Mn than the combined concentration of Ca, Mg and Na [9, 10, and 11]. In the Western Rand basin Au occurs in association with pyrite ($FeS_2$). Pyrite makes up about 3% of the Au bearing minerals [12]. The gold mine water was formed by oxidation of the acid producing mineral $FeS_2$ by exposure to $O_2$ and $H_2O$ as shown in eqn. (3).

$$FeS_2 + \tfrac{7}{2}O_2 + H_2O \rightarrow Fe^{2+} + 2H^+ + 2SO_4^{2-} \quad (3)$$

The other associated minerals such as dolomite and limestone occurred in insufficient proportions to neutralize the acidity generated by the oxidation of acid producing minerals resulting in acidic water. The acidity generated from $FeS_2$ caused the chemical weathering of surrounding rocks, therefore leaching potentially toxic elements into the water such as Ca, Mg, Na, Cl, Al, Mn, B, Cr, Pb, Th, Ba, Hg, As and Se.

Out of these elements, the gold mine water contained elevated concentration of Fe, Ca, Mn, Mg, B, Al, Cr, Pb, U and sulphate above the TQWR for domestic use as shown in table 4 [5, 7]. The pH of the mine water makes it unsuitable for domestic, agricultural and industrial use [5, 7].

Table 2: The physicochemical parameters of mine water.

| Parameter | Mine water | Potable water limit |
|---|---|---|
| pH | 3.48 ± 0.58 | 6-9 |
| EC | 3292 ± 36 | 0-700 |
| acidity | 752 ± 3 | NA |
| TDS | 1685 ± 52 | 0-600 (0-450) |
| hardness | 1549 ± 33 | 0-200 (0-100) |
| Sulphate | 4126 ± 44 | 0-500 |
| Fe | 895.62 ± 0.45 | 0-0.3 (0-0.1) |
| Ca | 376.33 ± 0.78 | 0-32 |
| Mn | 282.21 ± 38 | 0-0.1 (0-0.05) |
| Mg | 155.46 ± 0.34 | 0-30 |
| Na | 81.08 ± 0.55 | 0-200 (0-100) |
| Cl | 10.24 ± 1.04 | 250 (0-100) |
| B | 5.43 ± 0.22 | 0-2.4 |
| Al | 4.06 ± 0.89 | 0-0.2 (0-0.15) |
| Cr | $3.15 \pm 4.16 \times 10^{-3}$ | 0-0.05 |
| Sr | 0.56 ± 0.15 | NA |
| Pb | 0.51 ± 0.013 | 0-0.01 |
| K | 0.46 ± 0.02 | 0-50 |
| Cu | 0.21 ± 0.052 | 0-2 (0-0-1) |
| U | 0.29 ± 0.083 | 0.07 (0-0.03) |
| Zn | 0.25 ± 0.19 | 0-3 (0-0.5) |
| Th | 0.013 ± 0.18 | NA |
| P | $0.11 \pm 1.72 \times 10^{-3}$ | NA |
| Se | 0.058 ± 0.42 | 0-0.02 (0-0.04) |
| Ba | $0.06 \pm 6.46 \times 10^{-3}$ | 0-0.7 |
| Li | 0.003 ± 0.01 | NA |
| Be | $5.7 \times 10^{-3} \pm 8.6 \times 10^{-4}$ | 0-0.012 |
| Cd | $7.1 \times 10^{-3} \pm 1.27 \times 10^{-3}$ | 0-0.003 (0-0.005) |
| As | $4.1 \times 10^{-3} \pm 3.46 \times 10^{-3}$ | 0-0.001 |
| V | $1.7 \times 10^{-3} \pm 1.41 \times 10^{-3}$ | (0-0.01) |
| Ni | $7.1 \times 10^{-4} \pm 7.25 \times 10^{-6}$ | NA |
| Mo | $4.81 \times 10^{-4} \pm 2.08 \times 10^{-4}$ | 0-0.07 |
| Hg | $1.2 \times 10^{-6} \pm 4.3 \times 10^{-7}$ | 0-0.006 (0-0.001) |

*Note: values in brackets obtained from Department of Water Affairs of South Africa 1996 if the values are different from those indicated by the World Health Organization [5, 7]). NA stands for not applicable. TWQR stands for target water quality range. The units for parameters are mg/L except pH which has no units, EC (mS/cm) and TDS and hardness (mg/L of $CaCO_3$)*

### 3.1 Radioactivity characterization of mine water

The geology of the Western basin is made of more U minerals than Au bearing minerals [13]. Analysis of the gold mine water for radioactivity was carried out using alpha and gamma spectrometry as outlined in section 2.3 and the results obtained are shown in table 3. The results obtained indicated that the gross alpha and beta radioactivity of the mine water was 12 and 6 times more than the required limit for potable water respectively, as shown in table 3.

Table 3: Alpha, beta and isotope activities of gold mine water.

| Isotope | Activity ($Bq.L^{-1}$) | Concentration (µg/L) | WHO, 2011 ($Bq.L^{-1}$) |
|---|---|---|---|
| $^{238}U$ | 3.16 ± 0.04 | 253.99 ± 3.22 | 10 |
| $^{234}U$ | **4.71 ± 0.05** | $0.020 \pm 2.17 \times 10^{-4}$ | 1 |
| $^{230Th}$ | 0.69 ± 0.11 | $9.59 \times 10^{-4} \pm 1.47 \times 10^{-4}$ | 1 |
| $^{226}Ra$ | 0.36 ± 0.01 | $9.87 \times 10^{-6} \pm 2.74 \times 10^{-7}$ | 1 |
| $^{210}Po$ | 0.02 ± 0.0037 | $4.47 \times 10^{-8} \pm 8.10 \times 10^{-9}$ | 0.1 |
| $^{235}U$ | **0.145 ± 0.002** | 1.81 ± 0.025 | 1 |
| $^{227Th}$ | 0.202 ± 0.016 | $1.77 \times 10^{-10} \pm 1.41 \times 10^{-11}$ | 10 |
| $^{229}Ra$ | 0.101 ± 0.01 | $1.33 \times 10^{-14} \pm 1.32 \times 10^{-15}$ | - |
| $^{232}Th$ | 0.0619 ± 0.071 | 15.24 ± 1.75 | 1 |
| $^{228Th}$ | **0.124 ± 0.01** | $4.23 \times 10^{-9} \pm 3.41 \times 10^{-10}$ | 1 |
| $^{224}Ra$ | 0.0306 ± 0.045 | $5.11 \times 10^{-12} \pm 7.51 \times 10^{-13}$ | 1 |
| gross alpha | **6.01 ± 0.93** | | 0.5 |
| gross beta | **6.05 ± 0.41** | | 1 |

The maximum gross alpha and beta radioactivity for potable water are 0.5Bq/L and 1Bq/L respectively [5]. Radioisotopes that contributed to the radioactivity of the mine water were; $^{238}U$, $^{234}U$, $^{230}Th$, $^{226}Ra$, $^{235}U$, $^{227}Th$, $^{229}Ra$, $^{232}Th$, $^{228}Th$, and $^{224}Ra$. The activities of radioisotopes that were greater than the allowed limit for potable water were $^{234}U$, $^{235}U$ and $^{228}Th$ as shown in table 3. If U is allowed to accumulate in the kidneys it causes kidney failure [5]. Total U concentration that was determined for U in the mine water was about 256µg/L. This was well above the allowed limit for total U concentration set by [5], which is 30µg/L. Analysis of the mine water with ICP-OES showed that the water contained 290µg/L of U and 18µg/L of Th as shown in table 2. These results were close to those obtained using alpha and gamma spectrometry which found that the concentration of U and Th were about 256µg/L and 15µg/L respectively. This showed that ICP-OES can be used to analyse radioactive elements such as Th and U (in aqueous solutions) instead of the sophisticated, rigorous and expensive analytical techniques such as alpha and gamma spectrometry. There were no radioactive isotopes for K and Pb detected in the mine water using alpha and gamma spectrometry.

The radioactivity detected in the mine water is attributed to the fact that U is mined in addition to Au mining at the gold mine. The exposure of $FeS_2$ to oxidizing conditions results in formation of AMD. The low acidity of the water enhances the dissolution of the associated U containing minerals, resulting in AMD which is radioactive. Since the radioactivity in the gold mine water was much greater than the required limit for potable water, the treated water should be evaluated for radioactivity as well.

### 3.2 Aqueous distribution of NORMs

Two of the natural occurring radionuclide materials (NORMs) identified in mine water was U and Th. The distributions of these NORMs were modeled using SpecE8 program of the GWB as outlined in section 2.4 and the results are shown in fig 5.

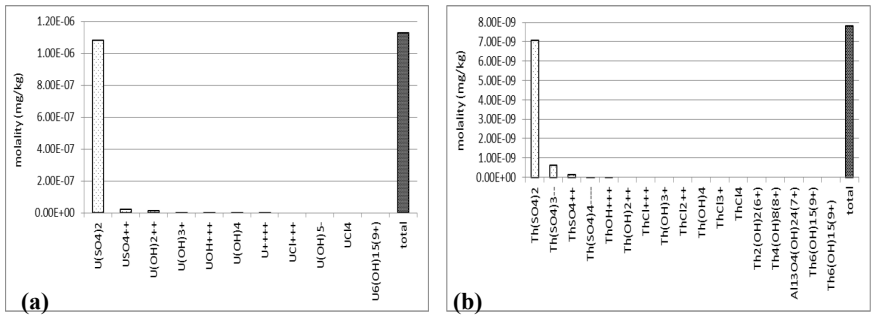


Figure 5: Uranium (a) and thorium (b) aqueous distribution in gold mine water.

The modeling results show that U species in the gold mine water were predicted to be mainly U species associated with sulphate ions, that is 96% of $U(SO_4)_2$ and 2% of $USO_4^{2+}$ as shown in fig. 5(a). The modeling results produced by GWB software of Th species in the mine water are shown in fig. 5(b). According to SpecE8 program the main Th species comprised of 90% of $Th(SO_4)$, 8% of $Th(SO_4)_3^{2-}$ and 2% of $ThSO_4^{2+}$ as shown in fig. 5(a).

## 4 Conclusion

The gold mine water can be classified as acid mine drainage because the pH was less than 5 and contained elevated concentration of Fe, Al, Mn and sulphate ions. The mine water was unsuitable for any purpose (drinking, irrigation or industrial) because of the lower pH and the elevated concentration of Fe, Al, Mn, Pb and sulphate ions in the water. The radioactivity of the gold mine water was found to be well above the required limit for potable water. The gross alpha and beta radioactivity of the water were 12 and 6 times above the potable limit respectively. The radioactivity was mainly due to U, Th, K and Ra radioisotopes. Treatment of the mine water therefore requires evaluation of the radioactivity of the product water. The SpecE8 program of the GWB software has shown that the NORMs such as Th and U were found to exist in association with sulphate ions in the gold mine water. This meant that these ions were less bioavailable as their mobility was reduced because of their nature.

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