

Assessment of two kinetic tests to predict the acid mine drainage in waste rock samples of a uranium mine

Avaliação de duas metodologias de ensaios cinéticos para previsão de drenagem ácida de mina em amostras de estéril de mineração de urânio

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Abstract

Acid mine drainage is the result of the oxidation process of sulfide bearing rocks. This process occurs when the sulfide material is exposed to atmospheric conditions. Under these conditions, successive oxidation reactions yield sulfuric acid generating acidic waters. This problem becomes more serious when the surrounding rocks are not able to neutralize the acid. The low pH condition of the drained water accelerates the solubility process of solid materials (rocks, soils and sediments) and favors metal and metalloid leaching making them available to the environment, which impairs the hydric system quality. This is a quite common situation at abandoned mines where the material is exposed, as at the Osamu Utsumi Mine in Caldas, MG. The main goal of this work was to assess and compare two kinetic tests in an attempt to predict the acid generation from the waste rock of the Osamu Utsumi Mine. The tests were carried out using a Soxhlet extractor and a leaching column. The leachate from the two tests was analyzed for the physical-chemical parameters (pH, Eh and electric conductivity) and metals and metalloids.

Keywords: acid mine drainage, leaching columns, Soxhlet extractor.

Resumo

A drenagem ácida de mina é o resultado do processo de oxidação de rochas que contêm minerais sulfetados. Esse processo ocorre quando o material sulfetado é exposto a condições atmosféricas. Nessas condições, sucessivas reações de oxidação dos sulfetos levam à formação de águas ácidas. Isto é agravado quando as rochas existentes no local não têm capacidade de neutralizar o ácido formado. A condição de baixo pH da água drenada acelera o processo de solubilização de materiais sólidos (rochas, solos e sedimentos) e favorece a lixiviação de metais e metalóides, tornando-os disponíveis no ambiente, fato que pode comprometer a qualidade dos cursos d'água adjacentes. Esse quadro é muito comum em minas abandonadas, nas quais o material retirado fica exposto, como é o caso da Mina Osamu Utsumi, em Caldas-MG. O objetivo desse trabalho foi avaliar e comparar dois tipos de ensaios cinéticos, na tentativa de prever a geração ácida do estéril da Mina Osamu Utsumi. Os ensaios foram realizados em extrator Soxhlet e coluna de lixiviação. Os lixiviados obtidos nos ensaios foram analisados para parâmetros físico-químicos (pH, Eh e CE) e metais e metalóides.

Palavras-chave: Drenagem ácida de mina, coluna de lixiviação, extrator Soxhlet, estéril de mineração de urânio.

1. Introduction

In the Caldas municipality (Minas Gerais, Brazil) the first Brazilian uranium mining-industrial complex was installed and named Complexo Mínero-Industrial de Poços de Caldas (CIPC). It was inaugurated in 1982 and is presently being decommissioned under the control of the Brazilian Nuclear Industries (INB - Indústrias Nucleares do Brasil). The Osamu Utsumi Mine was an open pit one, which allowed for better exploitation of the uranium ore. Sulfides are usually associated with uranium deposits such as this and consequently acid mine drainage (AMD) is prone to develop. In the studied mine, the AMD problem was detected when the overburden started to be removed, and reached the mine dumps (eight in total), the pit and access roads. Besides, during mining and ore dressing, liquid and gaseous effluents, aerosols and residues were also produced. These substances must be properly manipulated to avoid dispersion of chemical species that can endanger the environment. The chemical species include heavy metals, toxic anions and natural radionuclides, which can form oxides and salts in suspension, solution or be gaseous such as radon (Fernandes et al., 1995, Cipriani, 2002).

CIPC is located in the Poços de Caldas Alkaline Complex, where the most abundant rocks are nepheline syenites and fine-grained equivalents (tinguaite and phonolites). The hydrothermalized alkaline rocks that host the uranium mineralization are Si- and Al-poor and very K-, Na- and Ca-rich. Rocks with high Ti contents and Mo minerals, zircon, pyrite and fluorite stand out in this alkaline complex (Fraenkel et al., 1985).

Pyrite (FeS₂) is the chief contributor to acid mine drainage (AMD). In nature

pyrite is usually associated with coal deposits and metal ores including Zn, Cu, U, Au, Ag and others. Marcasite is also found but is less common because it is unstable (Evangelou, 1995). Other sulfide minerals are susceptible to oxidation and generation of acid drainage, such as: pyrrhotite (Fe_{1-x}S, where 0 < x < 0.2), chalcopyrite (CuFeS₂), chalcocite (Cu₂S), sphalerite (ZnS), galena (PbS), millerite (NiS), arsenopyrite (FeAsS) and cinnabar (HgS) (US EPA 1994).

The kinetic tests were carried out in waste samples from one of the main dumps of the Osamu Utsumi Mine, dump 4 (BF4). In this dump waste was disposed using the descending construction method and created a 12.4 million m³ waste pile, which occupies an area of 56.9 ha and is 90 m high. The waste mineral composition was determined by INB. The matrix is composed of tinguaite/phonolitic masses, where orthoclase and sericite dominate. It is impregnated by hydrothermal products such as pyrite, fluorite, and U, Mo and Zr minerals (Souza, 1995 and Cipriani, 2002).

The AMD generated in the mine is sent to a treatment station, where pH corrections are made by adding hydrated lime. Before launching in the Cercado stream, flocculation, decantation of heavy metals and calcium sulfate take place. This treatment represents high costs and, annually, an amount between R\$ 800,000.00 and R\$ 900,000.00 are spent considering only the consumption of hydrated lime and flocculant (Flores, 2006).

The AMD control and treatment methods can be divided in three courses of action: prediction, prevention and remediation. Prediction is usually made on the basis of works preliminary to the mining operation project. Different methods are

applied in order to assess the capacity or probability of a certain material to generate acids and bases under certain conditions (Blowes et al., 2003, Mend Report 2009).

Prediction involves laboratory and field methods. When possible, field methods are obviously preferred because they better represent the setting of acid generation in each site. On the other hand, the record of laboratory methods is long, including an ample range of test equipment and methods, besides offering advantages under the technical-economic point of view. Laboratory tests for AMD prediction include static and kinetic methods and are usually associated with geochemical models (Hornberger & Brady, 1998, Evangelou, 1995, Mend Report, 2009, Bernardes de Souza & Mansur, 2011).

The static methods take into consideration the acid-base balance and are widely known in the literature as acid-base accounting or ABA (Sobek et al., 1978). The kinetic methods can supply information on weathering rates and abundance of ions in the leachate, data that are not obtained in the static tests. Thus, besides predicting the acid drainage potential in the long term, they attempt to incorporate the dynamics of the physical, chemical and biologic elements, bringing the prediction closer to the reality of the mine. Among the main kinetic methods, we highlight the following: humidity cell, Soxhlet extractor, leaching column and B.C. (British Columbia) confirmation kinetic tests (US EPA, 1994, Blowes et al., 2003, Machado & Schneider, 2008, Mend Report, 2009).

The present work aimed at assessing two kinetic testing methods, the leaching column and the Soxhlet extractor, to predict AMD in the waste of the Osamu Utsumi Mine dump 4 (BF4).

2. Material and methods

The samples from dump 4 (BF4) were collected by means of pick and shovel ca. 30 cm below the surface, in order to avoid sampling strongly weathered material. In one of the three slopes selected randomly, three increments were collected with ca. 30 kg of waste. The material was disaggregated, homogenized and quartered. After drying at 40°C, a part was used in particle size analysis by sieving. After quartering, aliquots of different fractions obtained by sieving and the crude waste sample were pulverized

to a particle size smaller than 200 mesh. The pulverized samples were analyzed by x-ray diffractometry to determine the mineral composition. This identification was carried out using a Shimadzu diffractometer equipped with a Co ($\lambda = 1.728 \text{ \AA}$) tube. The diagrams were interpreted according to the Powder Diffraction Files (JCPDS 1967). The waste samples were solubilized by acid digestion (HNO₃/HCl/HF) and analyzed by ICP OES for major and trace elements. Uranium contents were obtained by means of a Perkin Elmer

Elan 6000 mass spectrometer (ICP-MS) and SO₄²⁻ contents were determined using a Leco sulfur analyzer. The gravimetric method was also used to determine PPC and SiO₂ contents.

For the leaching column test, a glass column of 70 cm in height and 6.7 cm of internal diameter was used. For the kinetic test a Soxhlet extractor of 1L capacity and internal diameter of 8.0 cm was used. The waste rock was sieved and the fraction less than 19 mm was used in both tests. Such procedure was adopted in order to

maintain inside the column a maximum proportion of four times between the largest particle size and the equipment internal diameter (Souza 1995). A waste quantity of 1,400 g dry weight (<19 mm) and 427 g dry weight (<19 mm) were used in the column and the extractor respectively.

The volume of water added to the column was calculated with the objective of simulating the local yearly average rainfall of 1637.48 mm (Tedeschi, 2005), transformed to two liters of deionized wa-

ter each 10 days until a total of 210 days. In the Soxhlet extractor 37 cycles were carried out with an average duration of 9 hours and 40 minutes using 1 liter of deionized water each cycle. The influent and effluent waters were analyzed for chemical and physico-chemical parameters such as pH, Eh, electric conductivity, metals and metalloids.

The measurements of the physico-chemical parameters were carried out with previously calibrated bench equipment:

Tecnopon/mPA-210 pH meter, Digimed/DM20 Eh meter and Quimis/Q405M conductivitymeter.

The metal and metalloid concentrations in the effluent and influent waters of the kinetic tests and previously solubilized waste were obtained using a Spectro Ciros CCD Inductively Coupled Plasma - Optical Emission Spectrometer (ICP OES). The results were validated using the certified reference material NIST 1643e for the waters and GBW07311 for the waste samples.

3. Results and discussion

The mineralogical determinations by x-ray diffraction revealed the presence of quartz (SiO₂), kaolinite [Al₂Si₂O₅(OH)₄], gibbsite [Al(OH)₃] and muscovite [KAl₂Si₃AlO₁₀(OH,F)₂] as the major components of the waste sample. Peaks of

ankerite [Ca(Fe²⁺,Mg, Mn)CO₃]₂], antigorite [(Mg,Fe²⁺)₃Si₂O₃(OH)₄], vanadinite [Pb₅(VO₄)₃Cl] and microcline (KAlSi₃O₈) were also observed.

The particle size distribution according to Wentworth (1922) is presented in Table

1. The waste rock collected in BF4 contains 26.7% of grains concentrated in the pebble and microparticle (P) fractions. A considerable amount of fine material was observed, once the sum of fine fractions (S/C, SVF and SF) represents 33.2% of the whole sample.

Grain size (µm)	Size terms	Quantity (%)
<63	Silt and Clay (S/C)	13.8
63-125	Sand very fine (SVF)	10.2
125-250	Sand fine (SF)	9.16
250-500	Sand medium (SM)	11.7
500-1000	Sand coarse (SC)	12.8
1000-2000	Sand very coarse (SVC)	15.6
>2000	Pebble e microparticle (P)	26.7

The chemical composition of the waste rock was determined for each particle size fraction with the objective of assessing possible variations in the concentrations of chemical elements as a function of the particle size. Among the elements analyzed, Mn, Ba, Co and Cr revealed a considerable difference as a function of particle size (RSD > 20%). The concentrations of these elements increase as the particle size increases

(Table 2).

The procedure adopted to determine the chemical composition of the BF4 waste samples was also applied to the certified reference material (GBW 07311) and the results were satisfactory for the majority of the elements analyzed. Exceptions are P, Zr and As, because the concentrations obtained for the reference material were below the certified value (CV). This indicates that for these

Table 1

Particle size distribution of the waste rock collected in the Osamu Utsumi Mine BF4 dump.

elements, the results presented for the waste samples may be underestimated (Table 2).

The BF4 waste rock fraction <19 mm is characterized by high SiO₂ contents (47.8 %), followed by Al₂O₃ (29.9%), K₂O (7.81%) and Fe₂O₃ (5.03%). Such result reflects the mineral composition of the material. Among the trace elements, the following are the most abundant: Ba>Sr>S>V>Mo>Pb>Y>U>Th>Y.

Major Elements (%)																
	Al ₂ O ₃	CaO	Fe ₂ O ₃	K ₂ O	MgO	MnO	Na ₂ O	P ₂ O ₅	TiO ₂	ZrO ₂	SiO ₂	PPC				
S/C	27.6	0.0291	5.18	9.01	0.0769	0.0223	0.0914	0.193	0.570	0.312	49.4	7.29				
SVF	27.8	0.0281	4.94	9.31	0.0874	0.0300	0.0967	0.162	0.590	0.252	50.4	6.79				
SF	29.1	0.0298	5.14	8.53	0.0919	0.0442	0.0780	0.162	0.602	0.283	48.0	7.81				
SM	30.3	0.0319	5.69	6.45	0.1080	0.0767	0.0817	0.185	0.685	0.237	46.7	8.45				
SC	32.0	0.0301	5.57	7.40	0.1101	0.1113	0.0783	0.180	0.640	0.266	46.0	8.96				
SVC	32.5	0.0301	5.54	7.06	0.1114	0.1318	0.0755	0.162	0.628	0.265	45.9	9.26				
P	27.3	0.0332	6.58	10.28	0.0944	0.3578	0.1080	0.192	0.531	0.275	49.1	6.42				
Average	29.5	0.0303	5.52	8.29	0.0970	0.111	0.0871	0.177	0.606	0.270	47.9	7.85				
RSD (%)	7.28	5.66	9.77	16.5	13.5	105	13.8	8.11	8.27	8.87	3.67	13.8				
BF4<19mm	29.9	0.0351	5.03	7.81	0.0955	0.111	0.0928	0.172	0.587	0.239	47.8	7.85				
GBW 07311	9.80	0.445	4.37	2.84	0.539	0.279	0.380	0.0429	0.282	0.0106	-	-				
CV	10.6	0.470	4.39	3.28	0.620	0.322	0.460	0.0580	0.350	0.0207	-	-				
Trace Elements (mg.kg ⁻¹)																
	As	Ba	Be	Co	Cr	Li	Mo	Pb	S _{total}	Sr	Th	V	Y	Zn	U	S _{substance}
S/C	67.8	582	5.74	6.13	9.01	54.5	259	255	435	582	98.4	318	130	84.5		
SVF	59.8	599	6.02	10.8	8.47	59.5	230	237	414	519	89.2	334	113	86.1		
SF	63.4	525	6.21	11.2	14.0	58.9	234	229	399	484	91.6	337	117	94.1		
SM	67.6	624	7.98	13.8	19.1	72.9	264	260	437	538	104	376	128	117		
SC	63.7	617	9.01	12.1	20.1	74.7	249	253	396	498	103	374	123	111		
SVC	55.9	649	8.83	11.7	24.2	76.8	227	264	362	457	88.5	365	110	109		
P	60.8	1123	8.59	14.8	35.1	71.7	215	296	519	530	89.2	425	108	110		
Average	62.7	674	7.48	11.5	18.5	67.0	240	256	423	515	94.8	361	118	102		
RSD (%)	6.82	29.9	19.2	24.0	50.3	13.5	7.54	8.44	11.7	7.87	7.13	9.90	7.24	12.9		
BF4<19mm	61.7	668	7.27	13.0	14.3	73.8	270	258	401	502	107	336	128	95.2	119	233
GBW 07311	136	240	24.1	10.0	36.7	69.2	<5.00	648	160	26.8	20.3	54.0	32.7	368		
CV	188	260	26.0	8.50	40.0	70.6	5.90	636	170	29.0	23.3	46.8	42.7	373		

RSD = Relative standard deviation; CV = certified value.

Table 2

Chemical composition of the particle size fractions of the BF4 waste rock sample and the BF4 waste rock fraction <19mm.

Expressing the S content as sulfate (Ssulfate) and total S concentration (Stotal), it was possible to estimate the amount of sulfur present in the sample as sulfide. Thus, S concentration in the sulfide form (Ssulfide) in the BF4 waste rock sample fraction <19 mm, also named pyritic sulfur (Spyritic), is 168 mg/kg. This represents an

acidity potential (PA) of 0.525 kg CaCO₃/t of the BF4 waste. Static tests to obtain the acid-base accounting (ABA) in BF4 waste rock were carried out by Leite (2010) and the results showed that the neutralization potentials (PN) of the material analyzed are significantly different from one method to the other (standard ABA, modified

ABA and the British Columbia Research Initial Test). This shows why the waste rock composition makes the application of traditional methods for the determination of the neutralization potential (PN) and consequently for the evaluation of the acid-base accounting (ABA) impossible (Sobek et al., 1978, US EPA, 1994, Mend report, 2009).

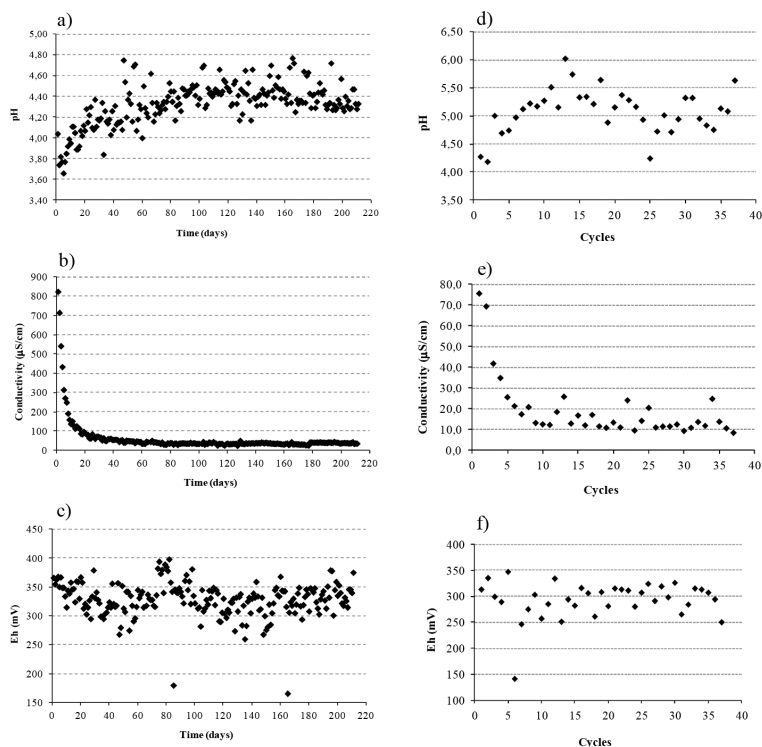


Figure 1
Variations of pH, electric conductivity and Eh during the kinetic tests using leaching columns (A, B, C) and Soxhlet extractors (D, E, F).

The kinetic tests to predict AMD in the BF4 waste rock were carried out in a leaching column and Soxhlet extractor. The objective of the kinetic test using the Soxhlet extractor is to make the material disposed inside the extractor undergo accelerated weathering. This goal is achieved by raising the temperature of the water in direct contact with the material. The waste rock temperature was monitored at the end of each cycle and varied from 43°C to 53°C.

The variations of pH, Eh and electric conductivity in the leaching column (Figures 1A, 1B and 1C) and the Soxhlet extractor (Figures 1D, 1E and 1F) are represented as a function of time. The pH of the leachate varied from 3.74 to 4.77 in the leaching column and from 4.18 to 6.02 in the extractor (Figures 1A and 1D). Conductivity decreased from 824 µS/cm to 27.0 µS/cm in the column and from 75.5 µS/cm to 8.39 µS/cm in the extractor (Figures 1B and 1E). The quantity of material in solution is directly related to different quantities of waste used in both tests, that is, 427g (dry weight) in the

extractor and 1400g (dry weight) in the column. Eh values are characteristic of a predominantly oxidizing environment and varied from 166 mV to 400 mV in the column and from 141 mV to 347 mV in the extractor (Figure 1C and 1E).

The graphs that show the conductivity measurements well represent the behavior of the major elements detected in the leachates in both tests (Figure 2). Thus the concentrations of the majority of the elements dropped significantly in the first 15 cycles. The predominant elements detected and listed in decreasing order of abundance were: S>Ca>Al>K>Mn>Si>Na>Mg. Among the trace elements detected, the predominant were: Zn>Y>Sr>Ba>Zr>Li>Be.

The accelerated decrease during the first 15 days of testing was not observed in the Ba elution. There was an increase in Ba content in the leaching column test from 27.8 µg/L to 151 µg/L. This increase in Ba concentrations in the leached waters is due to a decrease in sulfate, which is the species with which Ba establishes the following equilibrium: $\text{BaSO}_4 \rightleftharpoons \text{Ba}^{2+} + \text{SO}_4^{2-}$ where

K_{ps} is 1.1×10^{-10} . In the extractor leachate Ba concentrations varied from 33.1 to 14.9 µg/L (Figures 2E and 2J). Ankerite retains this element in the waste.

The predominant elements in the leachates were S, Ca and Al (Figure 2A and 2F). Ca concentrations varied from 80.0 to 0.153 mg/L in the column and from 4.01 to 0.178 mg/L in the extractor. Al contents varied from 38.1 to 0.879 mg/L in the column and from 1.41 to 0.030 mg/L in the extractor. Dissolved S concentrations also showed an accelerated decrease in the first days, with concentrations varying from 176 mg/L to 2.57 mg/L in the column and from 7.98 to 0.444 mg/L in the extractor. This reveals the dissolution of secondary sulfate minerals (Figures 2A and 2F). The addition of Ca in the waste rock results mainly from the dissolution of carbonates, fluorite, apatite and plagioclase (Waber et al., 1992).

Considerable Mn, K, Si and Na concentrations were also observed. Sodium varied from 9.49 mg/L to values that reached the detection limit (0.00100 mg/L) in the column and from 2.49 to

0.103 mg/L in the extractor. Potassium concentrations varied from 18.5 to 1.09 mg/L and from 2.62 to 0.234 mg/L in the column and in the extractor respectively. Silicon concentrations varied from 10.1 to 1.04 mg/L in the column and from 1.56 to 0.426 mg/L in the extractor. For Mn concentrations varying from 1.56 to 0.0538 mg/L were observed in the extractor and from 32.1 to 0.0843 mg/L in the column. The presence of Al is related to the predominant minerals in the waste rock, such as gibbsite, aluminosilicates and clay minerals. Si, Na and K are eluted with the dissolution of alkali feldspars present in the waste. Mn detected in the waste rock is due to pyrolusite present in the mine (Waber et al., 1992).

In both tests, Fe and Mg were de-

tected in some samples only. During the tests the concentrations remained below the detection limits (0.0109 mg/L for Fe and 0.00276 mg/L for Mg). The highest Fe concentration observed in the column was 0.0846 mg/L and 0.0403 mg/L in the extractor. The highest Mg concentration was 7.99 mg/L in the column and 0.263 mg/L in the extractor. The lack of Fe in the leachates is due to the precipitation of Fe as $\text{Fe}(\text{OH})_3$, which is highly insoluble, even under low pH conditions (Stumm & Morgan, 1981). An alternative explanation given by Souza (1995) for this fact is the reasonable percentage of fines in BF₄, which contributes to the mechanical and physico-chemical retention of precipitated Fe.

Among the trace elements analyzed,

Zn presented the highest concentrations in the leachate, from 10,600 to 76.2 $\mu\text{g/L}$ in the column and from 470 to 16.7 $\mu\text{g/L}$ in the extractor (Figures 2D and 2I). The presence of Zn in the leachate is due to the occurrence of sphalerite (ZnS) in the region (Waber et al., 1992).

Other trace elements present in considerable concentrations are Y, Sr and Zr. They were detected in all the eluted samples, the concentrations reaching 5,021 $\mu\text{g/L}$, 1,380 $\mu\text{g/L}$ and 68.0 $\mu\text{g/L}$ respectively in the columns. In the extractor the maximum concentrations observed were 165 $\mu\text{g/L}$ for Y, 74.1 $\mu\text{g/L}$ for Sr and 5.30 $\mu\text{g/L}$ for Zr (Figures 2D, 2E, 2I and 2J). In the waste rock, Y occurs as impurity in zircon and also in fluorite and illite (Waber et al., 1992).

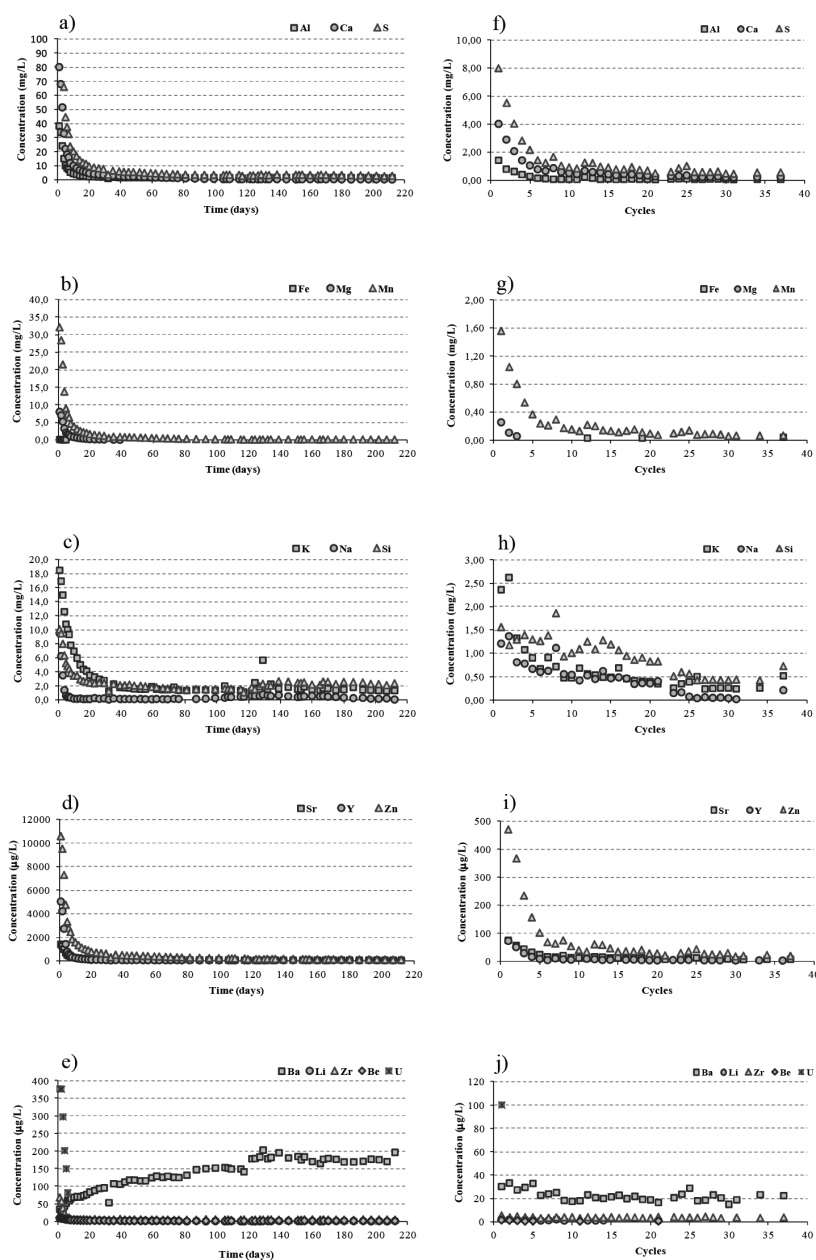


Figure 2
Concentrations of major and trace elements through time in the leaching column (A, B, C, D and E) and in the Soxhlet extractor (F, G, H, I and J).

Elements such as Li and Be were also detected in some samples. In others the concentrations remained below detection limits of 0.832 µg/L for Li and 0.324 µg/L for Be.

Uranium was detected in the first leached sample in the extractor with a concentration of 100 µg/L, and in the first six leached samples in the column (337 to 81.9 µg/L). This shows that the waste rock still liberates significant quantities of the element, despite the deactivation of the

mine in 1995 (Figures 2E and 2J). Campos et al. (2010) observed high concentrations of this element in waters drained from the mine dumps (8,110 µg/L and 8,720 µg/L), from the pit mine (4,020 µg/L) and in the interface between the mine and the surrounding environment (1,660 µg/L).

The presence of As was observed in some samples in the columns, the concentrations reaching 785 µg/L in the first six days of elution. In the test carried out in the extractor, concentrations remained

below the detection limit (70,1 µg/L).

Some trace elements were detected only in the leachates of the column test in the first eight days: Cd (maximum of 47.3 µg/L), Cu (maximum of 17.3 µg/L) and Ni (maximum of 88.3 µg/L). Other elements whose concentrations did not reach the ICP OES detection limits (LQ) in both tests were: Cr (LQ = 9.69 µg/L), Mo (LQ=14.5 µg/L), P (LQ = 107 µg/L), Pb (LQ = 86.6 µg/L), Ti (LQ = 6.74 µg/L), V (LQ = 6.95 µg/L) and Th (LQ = 19.5 µg/L).

4. Conclusions

In both tests there was a decrease in the concentrations of the majority of the major and trace elements, excepting Ba. The progressive decrease of these metals is due to the depletion of sulfides in the waste rock, leading to progressive increase in pH. The limited quantity of material used in the extractor (427 g dry weight) made the assessment of some trace

elements impossible, since their concentrations did not reach the detection limits of the technique used. On the other hand, only in the extractor was there observed an increase in pH with time. The column test illustrated the high capacity of acid generation in the BF₄ waste rock under environmental conditions, since the pH remained around 4.00 along the 212 days

of experiment. This experimental time period is equivalent to five years of rainfall, considering the local annual rainfall. The column test also showed that there is still significant uranium added to the leachate, reaching up to 337 µg/L, when compared to the maximum value established by CONAMA (2005) for class I waters of 20 µg/L.

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