



**CYANIDE DISTRIBUTION IN THE STREAM SEDIMENTS AND
TAILINGS AT THE BONFIM (W-Aa-Bi-Te) MINE,
NORTHEASTERN BRAZIL**

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ABSTRACT

At the Bonfim mine, cyanide processing used to extract gold from abandoned tailings represents an important environmental concern. In order to evaluate cyanide concentrations, twenty samples of sediments of the main streams crossing the mine site, and ten samples of the five mainly tailings were analyzed for total cyanide. Cyanide was found in two samples of tailings (0.83 and 1.44mg/Kg of total cyanide). Cyanide found indicates that its natural degradation could not occur after nine years of exposition of the tailings under surface conditions. Cyanide is probably present as insoluble metallic complexes formed by reactions between solution used in the gold recovery and metals from ore minerals. Cyanide detected at Bonfim shows concentrations below of those reported to contaminated tailings of the U.S. gold mines.

RESUMO

Na mina Bonfim, a cianetação das pilhas de rejeito para extrair ouro trata-se de uma importante problemática ambiental. A fim de se avaliar a ocorrência de cianeto, 20 amostras de sedimento de fundo das principais drenagens atravessando a área da mina e 10 amostras das 5 principais pilhas, foram analisadas para cianeto total. Detectou-se cianeto em 2 amostras das pilhas (0,83 e 1,44mg/Kg de cianeto total). A presença de cianeto indica que sua degradação natural pode não ter ocorrido, após 9 anos de exposição superficial das pilhas. Provavelmente o cianeto está em complexos metálicos insolúveis, formados por reações entre a solução cianetada usada e os metais presentes no minério. As concentrações de cianeto encontradas em Bonfim estão abaixo daquelas de pilhas de rejeito contaminadas de minas de ouro dos Estados Unidos.

INTRODUCTION

Nowadays, it is well known that inactive or abandoned mines can constitute potential sources for environmental hazards in their surrounding areas. These environmental

concerns could be of chemical, physical, or of landscape orders. Such mining areas need to be monitored, and studies of risk evaluation should be carried out. In case of contamination, technologies for mitigation and remediation should be applied in order

to recuperate affected sites (Kesler, 1994, Salomons, 1995). Particularly, gold mines have another associated environmental concern, which is contamination by mercury and cyanide, since these two substances are commonly used for processing ore (Ingles, Scott, 1981, Marsden, House, 1992, White, Markwiese, 1994, Smith, Mudder, 1999).

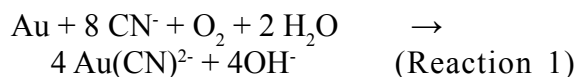
The Bonfim mine presents a particular potential for environmental concerns due to the use of cyanide heap process to extract gold from the abandoned tailings of the ancient activity of scheelite exploitation (Souza Neto, 2001, Pereira, 2003). After cyanide processing, the tailings remained exposed in the surface environment.

This work aims (i) to determine the total cyanide concentration in the tailings and stream sediments from the main drainage crossing the Bonfim mine, (ii) to discuss about the most probable processes taking place during the release of the cyanide to the environment, (iii) to evaluate the environmental risks linked to the cyanide concentrations found, and (iv) to propose preventing and mitigating actions that could be applied at the area of the mining investigated.

CYANIDE HEAP PROCESSES AND THEIR ENVIRONMENTAL CONCERNS

Gold solubility in cyanide-bearing solutions is known since 1783, but research concerning gold dissolution by these solutions and the role of oxygen on the mechanism of reaction were understood by the end of the

XIX century. In 1848, the chemical reaction of gold dissolution by cyanide ion was showed. The reaction produces a complex ion (auricyanide) and is known as Elsner's equation (Reaction 1) - (Marsden, House, 1992):



In the Bonfim mine, the heap leaching was the ore process adopted. This process consists in precipitating a cyanide solution over the tailing that contains powdered ore. When the solution percolates through the tailings, the reaction between ore and solution occurs (Fig. 1). Normally, KCN or NaCN are the solutions used in this process, which is considered very efficient and is largely applied worldwide (for a detailed description of cyanide ore processing see Gliese, 1997). However, other techniques that are cyanide-free were developed in order to improve a minimum environmental hazardous (Kesler, 1994). At Bonfim, the composition of the cyanide solution used is unknown.

During the ore treatment that uses cyanide, the reaction of gold dissolution is conducted with extreme care in relation to the pH. The pH of cyanide solutions must be kept above 11 in order to avoid the reaction between cyanide and hydrogen ion (e.g. from water) that will produce HCN, which is a lethal gas (Kesler, 1994, Smith, Mudder, 1999). The pH is usually controlled by adding CaO (Kesler, 1994). Other special handlings are required because cyanide is

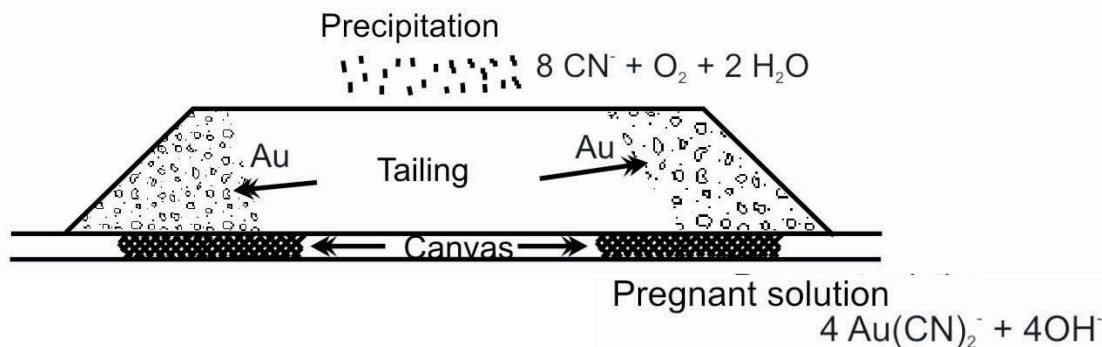


Figure 1: Schematic diagram showing the heap leaching process using cyanide-bearing solution.

very toxic. The concentrations of cyanide in the solutions used are relatively high, around 130mg/L of cyanide ion (Kesler, 1994), and only 50 to 250mg of cyanide ion can cause death in humans (Moore, 1991).

Geoenvironmental aspects and toxicity of the cyanide

In the environment, cyanide can occur under several forms. In water, cyanide is found as the molecular form, either as HCN or as cyanide ion (CN^- ; known as free cyanide). The cyanide ion can be converted to acid under pH 4. In alkaline conditions, free cyanide forms stable metallic complexes. The toxicity of the cyanide compounds depends on their chemical forms and on their constant of stability. Thus, as less stable is the cyanide compound, higher is its toxicity (Linardi, 1998).

As cyanide is highly toxic, it must be manipulated and released to the environment with extreme care. The release of cyanide effluents without appropriate care can promote significant environmental hazard, as degradation of the quality of water bodies and also affecting the biota. The effluent toxicity and its potential for damaging the environment are difficult to predict only by chemical analyses, since they do not reveal synergetic effects (Granato, 1995). Moreover, the difficulty and high costs for a complete chemical analysis of cyanide forms must be considered. Some toxicity tests can be applied for a rapid and practical evaluation in order to determine the damages for the aquatic life in case of release of toxic substances, with no need to characterize them (Trindade, 1994).

Concerning the cyanide toxicity, its effects are not only lethal or acute. Chronic effects of exposition could include reproduction and growing problems, as well as physiological changes. Tests of chronic toxicity are not commonly used due to their time spending and costs (Trindade, 1994). The CN^- ion and the acid form of cyanide (HCN) are toxic to all live forms. They block the oxygen transport in the metabolism (Granato,

1995). In these forms, cyanide is rapidly absorbed through the skin, or by inhalation or ingestion. Posteriorly it is transported by the blood plasma, affecting various constituents that are essentials to vital processes, as the cytochromooxidize enzyme that is responsible for cellular respiration.

The HCN is the most toxic form of cyanide, because is unstable and highly volatile. It forms strong bonds with iron, copper or sulfur, which are key elements of many enzymes and proteins. Cyanide competes with oxygen during its absorption in the organic tissues. The inhalation of high concentrations of HCN can promote asphyxia, paralysis, unconsciousness, convulsion and death by respiratory paralysis. The minimum lethal dose (LD_{50}) for humans is 150mg of NaCN. The TLV (Threshold Limit Value) index for soluble cyanide is $5\text{mg}/\text{m}^3$, and $10\text{mg}/\text{m}^3$ for HCN gas. This index establishes the limit under what workers can be diary exposed without have damages, such as vertigo, vomit, and faint.

Independently on the way of absorption of the cyanide ion (CN^-), the intoxication is always damaging for the organism. Exposure of substances containing small concentrations of cyanide compounds, as 18mg of CN^-/day , are normally well tolerable by the human organism, which transforms them into thiocyanide ion (SCN^-) in the liver, which is eliminated through the urine.

The presence of cyanide in water has significant effect over the biological activity of the aquatic ecosystem. Concentrations of free cyanide below 0.1mg/L can already be toxic for some aquatic organisms (Bretherick, 1981). In a general way, aquatic organisms are more directly exposed to long term intoxication, which results in several alterations, such as diminishing in the average size, egg production, and swimming velocity of these organisms (Ingles, Scott, 1981). Cyanide effects in these organisms are not cumulative, but are highly toxic.

GEOLOGICAL ASPECTS OF THE BONFIM MINE

The Bonfim W-Au (Bi-Te) deposit is situated in the state of Rio Grande do Norte, northeastern Brazil, about 27 km southeast of Lajes (Fig. 2). This deposit has a proven total reserve of 97 tonnes of scheelite and an ore grade average of 4.8 wt% WO_3 (Farias *et al.*, 1973). Scheelite exploitation occurred between 1969 and 1977, but was interrupted by a series of gallery collapses (Lima Neto *et al.*, 1985). In the early 1990's, gold was discovered in the tailings of the abandoned mine, and from this material about 0.1 t of gold has been extracted using the heap leaching process using cyanide-bearing solution. In mineralized rocks, the ore grade varies between 2 and 6 ppm of Au, but can be up to 60 ppm. Gold anomalies of up to 100 ppm have been detected in soil profiles (Docegeo 1996). Significant contents of Ag (up to 7 ppm), Bi (up to 17,000 ppm) and Te (up to 510 ppm) occur in the gold-rich rocks. Native gold contains about 14 % of Ag (Souza Neto, 1999). Gold anomalies of up

to 100 ppm have been detected in soil profile (Docegeo, 1996).

In the Bonfim region, geological units are basement gneisses, meta-sedimentary rocks (Seridó Group that contains three dominant lithologies: marble, quartzite, and schist which belong to the Jucurutu, Equador and Seridó Formations, respectively), granite and pegmatites of the Brasiliano igneous suite, and diabase (Fig. 3). All these rocks are of Neoproterozoic ages, except the basement gneisses that are Paleoproterozoic (Van Schmus *et al.*, 2003) and the diabase that is Mesozoic. This region was affected by a transpressional tectonic event (Brasiliano/Pan-African orogeny at about 600 Ma) with dextral shear in the NE-SW direction. Late in the tectono-metamorphic history of the Bonfim region, a metasomatism event transformed marble and schist into skarns (Souza Neto, 1999).

At Bonfim, two types of skarns have been recognized: skarns replacing marble and



Figure 2: Localization map of the Bonfim mine.

skarns replacing schist, which were developed at the schist-marble contact. The skarns replacing marble are green-colored, medium- to coarse-grained rocks, and show a massive structure and irregular-shaped contacts with pockets of remnant marble. Prograde skarn minerals include diopside, tremolite-magnesio-hornblende, and minor titanite, apatite, and allanite. The main retrograde minerals observed are talc, serpentine, chlorite, recrystallized calcite, and sericite in marble horizons. The skarns replacing schist are medium- to coarse-grained, and commonly show a banded structure, which appears either as alternating (e.g. garnet-amphibole and pyroxene-amphibole) or mixed bands. The typical mineralized skarn in the Bonfim mine exhibits alternating plagioclase and amphibole-pyroxene bands. Almandine, diopside, anorthite, and actinolite-magnesio-hornblende are the prograde minerals observed in these skarns. Accessory prograde minerals are titanite, apatite, allanite, zircon, and monazite. Scheelite, molybdenite, magnetite, and ilmenite are the main ore minerals in the prograde skarn. The retrograde stage of the skarns replacing schist has the following successive mineral assemblages: (i) Alkali feldspar; (ii) Clinozoisite-zoisite-sericite assemblage, probably formed by replacement of prograde anorthite; and (iii) Calcite-rich assemblage composed of recrystallized calcite and remnants of chloritized biotite. At Bonfim gold occurs as free grains disseminated within black, fine-grained minerals that fill thin conjugate brittle-ductile faults and extensional fractures, which cross-cut both the diopside-anorthite prograde, and alkali feldspar and clinozoisite-zoisite-sericite retrograde assemblages. These black minerals also occur as an impregnation in the skarns. Quartz, calcite and quartz-calcite veins (up to 5 cm wide) cross-cut both the prograde skarn and gold mineralization. The gold assemblage exhibits relatively rare minerals, such as bismite (Bi_2O_3) and fluorine-bearing bismite (75-80 vol%), native bismuth (5-10 vol%), bismuthinite (Bi_2S_3) and joseite ($\text{Bi}_4(\text{Te,S})_3$) both 5-10 vol%, and minor (< 5 vol%) chlorite, epidote, prehnite,

chalcocopyrite, sphalerite and gold (Souza Neto, 1999).

MATERIALS AND METHODS

SAMPLING

The sampling of sediment was carried out during dry season (December 2001). The samples were collected at the main streams crossing the site of the Bonfim mine and its surrounding area. At these streams, twenty sampling stations (named LS-01 to LS-20; Fig. 4) were strategically chosen in order to obtain a representative scenario of the area, taking in account both, portions without influence of the mine (upstream stations and stations located at the affluents of the main stream), and portions under direct influence of the mine activity (mine and downstream stations). Duplicate samples were collected from two sampling stations (LS-10 and LS-20). In each sampling station, the upper layer of sediment (0 to 3cm) in the middle of the stream channel was collected. Each sample was composed by several aliquots alleatorily sampled in an area of about 10 m of radius around the station. A plastic hand shovel and plastic bags were used to collect the samples.

The samples from the tailing were collected at five tailings (named BP-01 to BP-05; Fig. 5), which were chosen because of their height (the higher ones, from 2 to 6m height), and also because of their position in relation to the main stream (the closest ones). In each tailing, two samples were collected: one from the top and center, and the other one from the bottom of the tailing.

SAMPLE PREPARATION AND ANALYTICAL PROCEDURES

As the investigated drainages have no water during dry season, all collected samples were naturally dried. In the laboratory, sediment samples were sieved using a set of steel sieves in order to obtain the fine fraction (<63 μm) of the sediments to be used in the cyanide analyses. This particle size is accepted as the fraction that accumulates

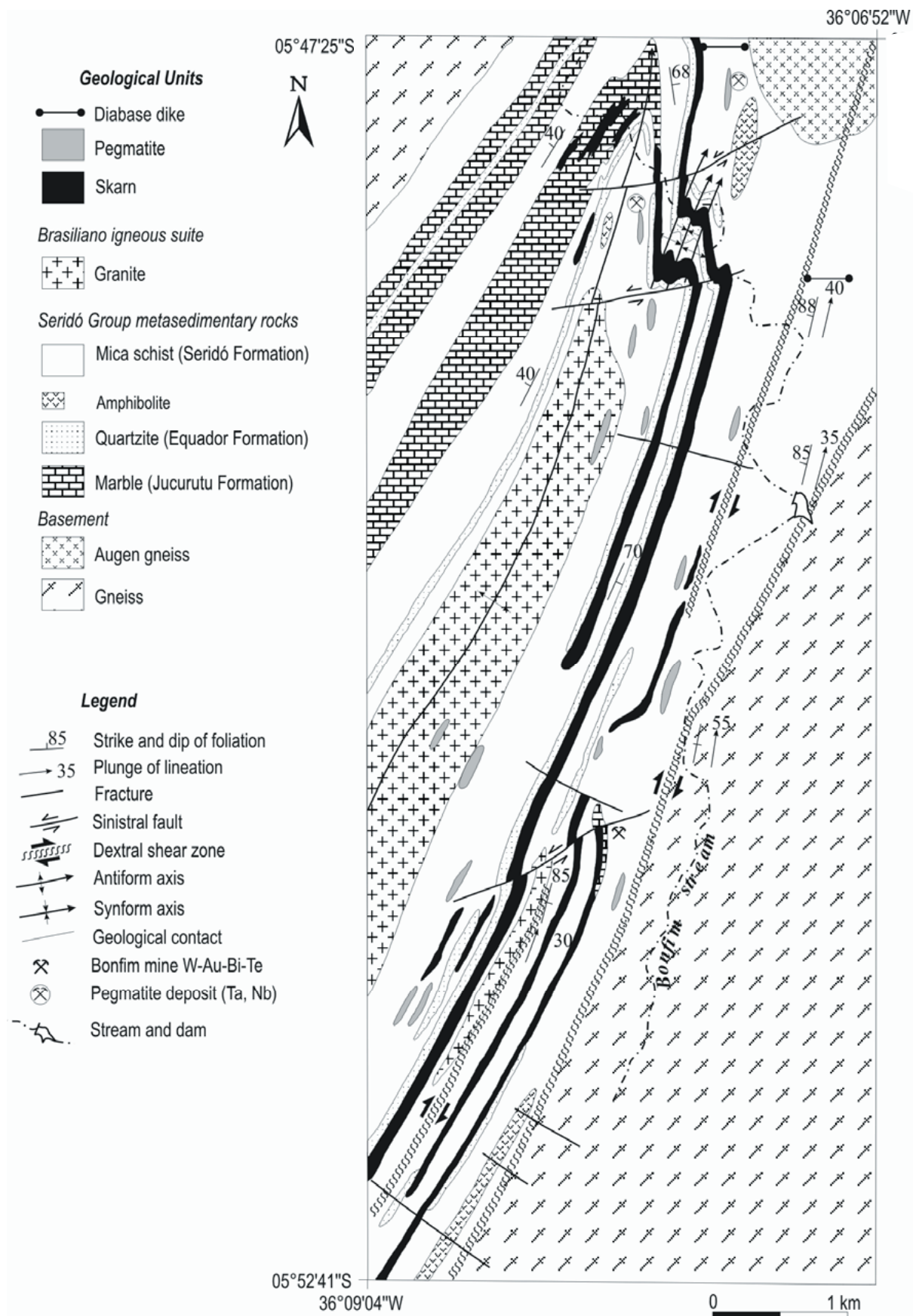


Figure 3: Geological map of the area of the Bonfim mine (simplified from Souza Neto, 1999).

higher concentrations of contaminants (Mantei, Sappington, 1994, Mudroch, Azcue, 1995, Herr, Gray, 1997). Tailing samples were quartered to obtain an aliquot part of about 100g and grinded with pestle in a mortar, both of porcelain. Total cyanide was analyzed at samples of stream sediments and tailings at the Lakefield-Geosol laboratories (Belo Horizonte, Brazil). Samples containing >1.0mg/Kg of total cyanide were analyzed by titulometric method, whereas samples with total cyanide amounts <1.0mg/Kg were analyzed by colorimetric digestion using pyridine and barbituric acid, according to the procedures of the Standards Methods for the Examination of Water and Wastewater (APHA-AWWA-WEF, 2005). Analytical methodology used showed a limit of detection of 0.50mg/Kg, 0.15mg/Kg of precision, and 0.02mg/Kg of accuracy.

It must be noted that analysis of total cyanide determines the total concentration of all cyanide forms present in the sample, excepting some thiocyanide, nitriles (Knowles, Wyatt, 1992) and metallic complexes of cyanide (e.g. auricyanides). Total cyanide is a common parameter used in the investigations of contaminated soil and water (Kjeldsen, 1999).

RESULTS AND DISCUSSIONS

The results revealed that all sediment samples analyzed contain total cyanide below 0.50mg/Kg (limit of detection of the method used). When comparing these data with those reported for other areas worldwide (Table 1), it can be concluded that the concentration of total cyanide found at Bonfim are below of those of non contaminated soils (< 0.005 – 0.5mg/Kg).

Concentrations of total cyanide found in the tailing samples were below 0.50 mg/Kg (limit of detection of the method used), excepting two samples (BP-1A and BP-4A; both from the top of the tailings), which presented 1.44mg/Kg and 0.83mg/Kg of

total cyanide, respectively. Comparing these values with those reported from tailings of gold mines in U.S.A. (1.5-23 mg/Kg; Table 1), it can be observed that the concentrations of total cyanide detected at Bonfim are below those reported in the gold mines in U.S.A.

The low concentration (or total absence) of total cyanide in the samples of stream sediments at Bonfim suggests that: either (i) the cyanide solutions used in the processing of tailings did not reached the streams, or (ii) if cyanide reached the streams, it was completely and naturally degraded or rapidly transported by superficial water during torrential rains, and thus was not fixed in the sediments.

Natural degradation usually occurs with cyanides, by means of oxidation or photodecomposition reactions (Linardi, 1998). It could generate two compounds as by-products, CO₂ and N₂. The oxidation of N₂ can subsequently originate nitrite (NO₂⁻) and nitrate (NO₃⁻) (Linardi, 1998), which is a well known contaminant of water bodies (Clark, Fritz, 1997). The occurrence of natural degradation can be plausible, considering the conditions of arid climate (with insufficient rainfall) at the region, which can catalyze the reactions.

On the other hand, the exposition time of geological material in the surface could corroborate to the low concentrations (or absence) of total cyanide recorded in the sediment and tailing samples in the area of the Bonfim mine. For instance, in the investigated site the tailings have been exposed under superficial conditions for nine years (until sampling), and cyanide present in this material could has been naturally degraded.

Despite the composition of cyanide-bearing solution used in the tailings at Bonfim be unknown, some discussions could be done concerning the path of cyanide degradation that could be occurred. In this case, two main processes could occur, as volatilization/ oxydation and dissociation. Both processes could naturally occur in the superficial and

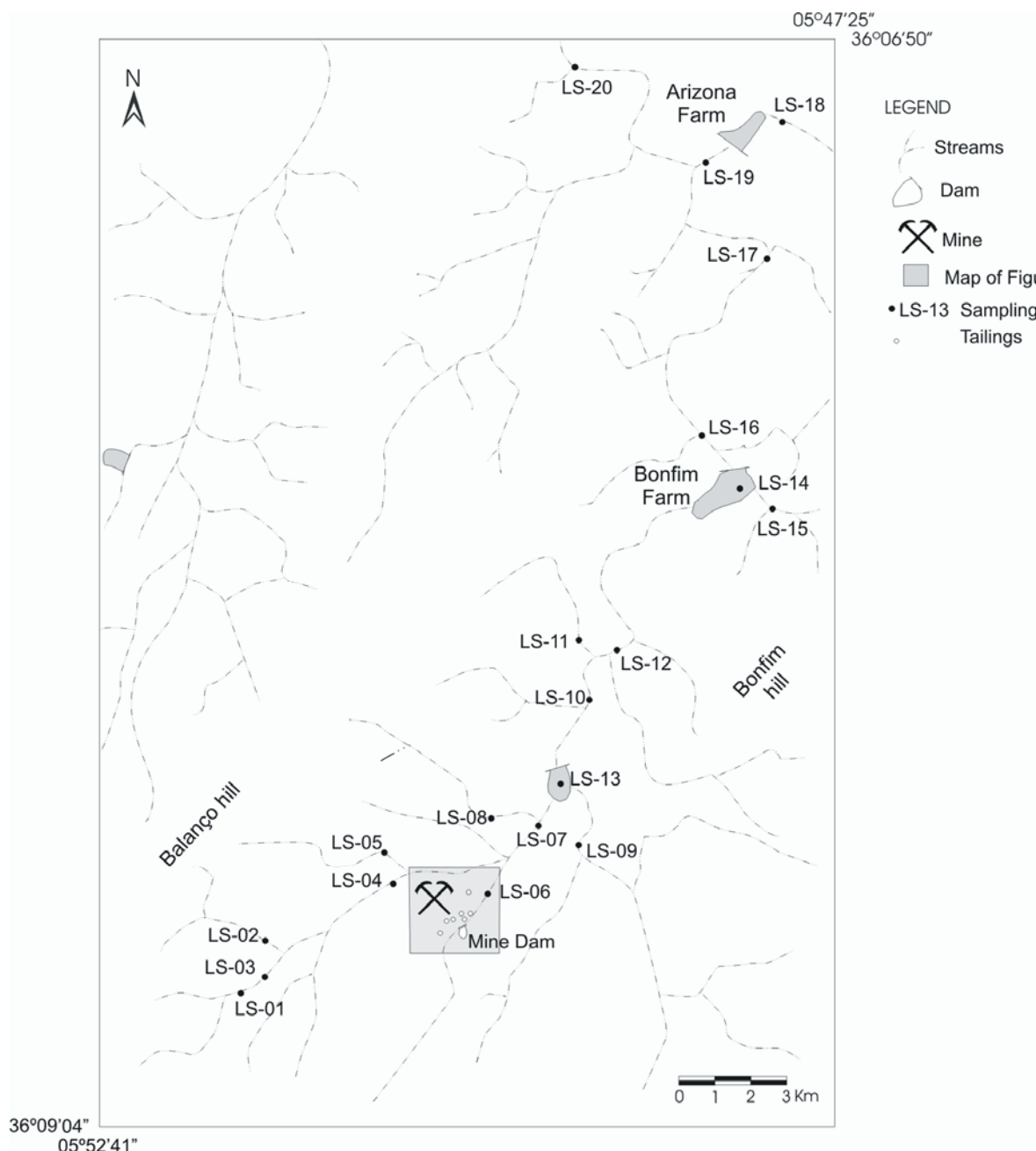


Figure 4: Map of sampling stations of stream sediment in the area of the Bonfim mine. Location of tailings is also shown.

inner zones of the tailings. In the superficial zone, volatilization of cyanide-bearing solution and oxidation of newly formed cyanide-bearing compounds could occur. The volatilization could also occur at the moment of use of cyanide solutions, if their pH value is not about 11. If pH values are below 9.36, under the superficial temperature of the region (26°C in average), the HCN could be formed (Reaction 2). Generally, such a decrease in pH values could be due to the rain or to the atmospheric CO₂ (Smith, Mudder, 1999).



The HCN that eventually forms can be converted to cyanite ion following the reaction below:



Concerning the dissociation as a probable mechanism that took place at Bonfim, it would occur in the inner zone of the tailings, and under oxidizing conditions.

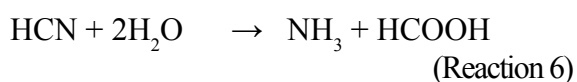
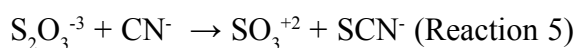


Figure 5: Map of sampled tailings in the area of the Bonfim mine.

The Reaction 4 illustrates this process, if one considers NaCN as the cyanide-bearing solution used.



In the portions of the investigated tailings where reducing conditions predominate, other reactions involving cyanide could take place, such as the formation of thiocyanite (Reaction 5) and the saponification of the HCN that generates ammonia and formic acid (Reaction 6).



Particularly the Reaction 5 could occur at the presence of sulfides, which are found in the mineral assemblage of the Bonfim ore. In order to verify the occurrence of the reactions 5 and 6, the products predicted by them must be further checked, characterizing cyanide species in the tailings of the Bonfim mine.

MEASURES OF REMEDIATION AND PREVENTION FOR THE BONFIM SITE

In order to avoid damage to the aquatic ecosystem, the Brazilian legislation establishes that effluents to be released in the environment must have a maximum concentration of 0.2mg/L of cyanide (CONAMA, 1986). Thus, any industrial effluent with cyanide concentration above this value must be treated before discharged.

Table 1: Concentration of total cyanide in soils and tailings from selected areas worldwide.

Soils / Tailings (location)	Total Cyanide (mg/Kg)	References
Soils (non contaminated) (Michigan, U.S.A.)	<0.005 – 0.5**	O'Hearn & Kesler-Arnold (1990)
Soils (Gas usines, Germany)	10 – 1,000*	Meeussen <i>et al.</i> (1994)
Soils (Gas usines, United Kingdon)	10 – 5,000	Byers <i>et al.</i> (1994)
Tailings (contaminated, gold mine) (New Mexico, U.S.A.)	1.5 – 23	White & Markwiese (1994)
Tailings (gold mine) (Bonfim, Brazil)	0.83 – 1.44	This study

* Average of 12 samples of cultivated soils; ** Variation in 220 samples.

Cyanide can be extracted from solutions by natural methods, involving chemical (e.g. oxidation) or biological (using some bacteria, such as *Pseudomonas fluorescens*, fungi and algae) reactions. The natural degradation is commonly used by large companies working with gold extraction. This treatment consists in keeping cyanide-bearing effluents within dams during a certain period of time. There is no mechanism to accelerate this process, which occurs exclusively in a spontaneous and natural way, depending on the climate (e.g. oxygen availability) and effluent (e.g. pH) conditions. Actually, natural degradation could be the result of interaction between several reactions, as follows (Linardi, 1998):

- i) volatilization of HCN;
- ii) dissociation of complexed cyanides;
- iii) hydrolysis of free and complexed cyanides;
- iv) photodecomposition (ultraviolet radiation of sun);
- v) chemical and bacterial oxidation, and
- vi) precipitation of insoluble complexed cyanides.

Among the reactions above (i) and (ii) are the main ones involved in the natural degradation, which could be used as a pre-treatment in order to reduce the consumption of chemical reagents in the whole process of cyanide extraction.

At Bonfim, none of treatments of cyanide-bearing effluents produced during

gold extraction was reported. Considering the rudimentary way that the cyanide heap process was carried out (iron barrels and plastic tubes, both perforated, and thin plastic canvas were used; all of them are still disposed in the mine site), it is possible to make a conjecture that some spill of the cyanide effluents to the environment probably happened. In this case, cyanide probably underwent natural degradation under the dry climate of the region.

In order to avoid any further release to the environment of the total cyanide detected in the tailings of the Bonfim mine, the complete removal of the tailings to an impermeable site, and the protection of them from the weathering agents are the main measures of prevention proposed.

CONCLUSIONS

This work represents an initial effort to diagnosticate the potential contamination linked to substances (e.g. cyanide and mercury) commonly used in the processes of gold recovery in Northeastern Brazil. Such mining activities are still used at this region, producing abandoned tailings of considerable volume. In recent years, as the environmental care is increasing, such activities without an adequate and careful handling are no more tolerated. The environmental contamination in all old mining sites in this Brazilian region must be evaluated, and in some cases, such as of Bonfim mine, it needs to be monitored.

The detection of total cyanide in two samples of tailings from the Bonfim mine makes necessary particular detailed analysis to investigate the chemical specie(s) that cyanide occurs. Without cyanide speciation data, the results reported in this work allow just the speculation that cyanide found could be present as insoluble metallic complexes (e.g. $\text{Fe}(\text{CN})_2$ and $\text{Cu}(\text{CN})_2$), considering the well known processes of cyanide degradation under similar climate conditions and the geochemical association of the Bonfim ore. Moreover, the investigation of secondary products formed by natural degradation of cyanide, such as nitrate that is a potential contaminant of the superficial water and groundwater bodies, becomes needful. At Bonfim, nitrate concentrations in groundwater must be carefully evaluated and monitored, since this water resource is vitally necessary for the survival of the population in such a region with notorious scarcity of water.

From the results obtained in this work, some other recommendations for further studies could be done:

- Evaluation of new methods of gold extraction, involving non toxic (or with low toxicity) substances, such as thioreia and bromine.
- Evaluation of the physic and chemical phenomena that control the cyanide concentration in different granulometric fractions of the tailings, in order to examine the distribution and speciation of the cyanide

in these fractions. This becomes necessary since the interaction phenomena between water and the tailing materials during a prolonged time, and changes in redox and pH conditions of the environment, as well as microbial destruction of the organic matter deposited in the soil, all of these can remobilize the cyanide available in the surface during percolation of rain water.

- Investigation of the cyanide toxicity in the local population through analysis of this substance in its blood and urine.

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(Footnotes)

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