

Cyanides in the Environment and Their Long-Term Fate

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ABSTRACT: A number of recent cyanide spills and tailings-dam failures have made world headlines. While hand cyanides are the primary chemicals used in the extraction of precious metals from low-grade resources, contributing substantially to the world economy, an appreciable anti-cyanide activism is in progress and search for alternative lixiviants is accelerating. Health effects of cyanide compounds other than free-cyanide are often insufficiently addressed or remain not required for reporting to regulatory agencies. This paper addresses cyanidation-extraction byproducts and their long-term fates in the ecosystem. Data show that a number of currently accepted cyanide abatement approaches can create long-lasting products that degrade at very low rates.

1 INTRODUCTION

The estimated amount of gold production in the world in 1998 was some 2,460,000 kg, more than 90 % of which was by cyanidation. The remaining 10% or so can be accounted for by gravity separations and such technologies (USGS, 1999; von

Michaelis, 1984). Based on reaction (1) the molecular ratio of cyanide-to-gold consumption is 2:1 which works out as 0.498 kg of NaCN for each kilogram of gold recovered.



Table 1. World gold production using cyanide; *based on actual consumption is 0.4 kg NaCN/kg Au

<u>Gold production</u>	<u>kg, NaCN equivalent needed</u>		
	kg	Stoichiometric	Actual*
World production (including US production)	2,460,000	1225080	984,000
US production	366,000	182268	146400
US production by cyanidation	341,000	169818	136400
US cyanidation, in tanks and closed containers	238,000	119000	95200
<u>US cyanidation in heaps and dumps</u>	<u>103,000</u>	<u>51294</u>	<u>41200</u>
	<u>NaCN Usage (kg /kg Au)</u>		
Stoichiometric kg NaCN/kg gold	0.498		
Usage in Canada (average kg NaCN/kg Au)	0.450		
Usage in S-Africa (average kg NaCN/kg Au)	0.280		
<u>Usage in Free world (average kg NaCN/kg Au)</u>	<u>0.400</u>		

Considering that some of the cyanide is recycled and some of it is lost to cyanicides, that is, cyanide-consuming-non-gold entities, the amount of sodium cyanide handled in the world for gold recovery comes out to be some 1 million kilograms per year. These numbers are summarized in Table 1 which excludes unrecorded numbers, likely, utilized in some developing countries.

Cyanide usage is not confined to the gold recovery industries but also includes metal recycling and metal plating and finishing industries. Notably, cyanide as NaCN and KCN, is a ubiquitous chemical used in the metallurgical industries since 1887. Flotation-concentration of Cu-Zn ores are also common users of cyanide as a modifying agent. Cyanides are also components of pesticide

formulations and their degradation products and are generated by numerous plants and organisms as well (Towill, 1978; Huiatt, 1983). Yet, their high toxicity is firmly under control during usage. Control of pH and safe handling practices appear to be the sufficient requirements for safety in the workplace. Figure 1 is the speciation-diagram for dissolved HCN gas where it is seen that at pH >10 essentially all of the cyanide is in the ionic (CN⁻) form. Under these conditions, in the plant, cyanide is not the "ogre" that it has been made to be, in recent years.

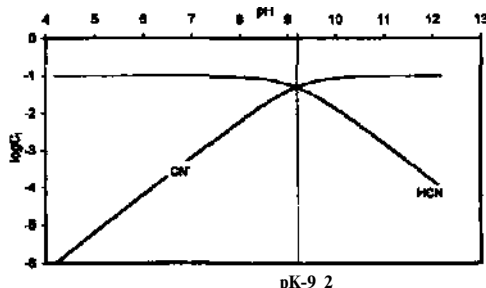


Figure 1. Speciation diagram for HCN in water (NaCN equivalent : 5 g /L)

Table 2. Some examples of recent high-profile cyanide accidents in the world.

Date	Accident
1992	Summitville, Colorado, USA; leaking mine abandoned, now a superfund site
1995	Omi gold mine, Guyana dam ruptures about 3.5 million tons of toxic waters are spilled with tailings.
1997	Nevada, gold quarry mine teach pad collapse releases 980 million tons of cyanide bearing slurry spills
1998	Kyrgyzstan-Issyk-Kul lake receives 1.7 tons of solid sodium cyanide from a truck overturn accident
1998	Homestake mining operation; Black-hills, S.D , USA 6 tons of cyanide-bearing tailings are lost to the environment.
2000	Baia Mare gold Mine, Romania, 100,000 tons of slurry is released into the rivers Lapus, and followed to Somes, Tizsa, and the Danube, from dam failure
2000	Papua, New Guinea, Dome Resources Freight helicopter spills some 100 kg of sodium cyanide during transportation

In this paper, the primary focus is on the use of cyanides as lixivants in the recovery of gold from natural resources. In this context, primary problems associated with it arise, mostly, after usage as a leaching-agent, in tailings ponds, active and

abandoned heaps and *at* disposal alter usage as a laboratory chemical and pilot plant material.

Numerous high profile and some secondary spills in the last decade have brought cyanide and its apparent perils to the limelight. A few of these are collected in Table 2. In all of the cases cited here, extensive efforts have been made for the remediation of the immediate damage to the environment with results considered satisfactory, while in some others such as the Summitville Mine In Colorado large dollar numbers have been dedicated towards remediation efforts.

2 ENVIRONMENTAL CONTAMINATION

Potential mechanisms of contamination in mining and metallurgy related areas include the following:

- 1) During transportation of sodium cyanide to site of use
- 2) Tailings dam failures
- 3) Heap and dump failures
- 4) Seepage from heaps dumps and impoundments
- 5) Clandestine usage

Especially in the cases of dam failures and seepage processes, cyanides take a secondary seat to metals such as Pb, Hg, Zn, Cu etc. which gain special notoriety and exacerbate abatement and control costs. The Summitville Colorado, Superfund site is a case in point where the current effort includes extensive attention to heavy metals. The cost of cleanup for this site has been estimated to be in excess of \$100 million.

The primary concern, in addition to the elimination of the immediate effects of such spills is the need to provide answers to a number of questions: i.e.: " what happens to the toxic material?"; " is it really rendered harmless and gone for ever?"; and "where does it all go?"

3 ANALYTICAL REPORTING METHODS

Cyanide compounds associated with mining and metallurgy can be broadly grouped, (Scott, 1981) as shown in Table 3. The strength of the metal-cyanide complexes on the other hand is better expressed in terms of their stability constants, that is, in essence their resistance to dissociation under ambient conditions. These values (recalculated after Scott, 1981 and Beck, 1987) are compiled in Table 4, where it is clearly observed that gold, cobalt silver and iron complexes are thermodynamically highly stable against dissociation.

Table 3 Examples of cyanide species in groups as used in assay and reporting procedures.

Ü E L	Examples
Free cyanide	CN ⁻ , HCN
Simple cyanide compounds (some partly soluble)	NaCN, KCN, Ca(CN) ₂ , Zn(CN) ₂ , Cu(CN) ₂ , Ni(CN) ₂ , Ag(CN)
Cyanide complexes	Weak complexes: Zn(CN) ₄ ²⁻ , Cd(CN) ₄ ²⁻ Moderately strong complexes: Cu(CN) ₂ , Ni(CN) ₄ ²⁻ Strong complexes: Fe(CN) ₆ ³⁻ , Co(CN) ₆ ³⁻ , Au(CN) ₂ ⁻

Table 4. Stability constants of some metal-cyanide complexes.

Species	logio Ks
Au(CN) ₄ ⁻	56
Au(CN) ₂ ⁻	37.65
Co(CN) ₆ ⁴⁻	50
Ag(CN) ₂ ⁻	39.1
Fe(CN) ₆ ³⁻	35.4
Cu(CN) ₃ ⁻	29.2
Ni(CN) ₄ ²⁻	26
Cu(CN) ₂ ⁻	19.95
Zn(CN) ₄ ²⁻	19.01
Cd(CN) ₄ ²⁻	18.93
AgCN _w	13.80

For compliance with regulatory agency requirements (In the USA) cyanides are reported in three forms:

- Free cyanide, which includes HCN and CN⁻
- WAD (weak-acid-dissociable) cyanide,
- Total cyanide

It is also customary to determine and report metals such as Zn, Cu, Ag, Hg, Mn, Fe etc. together with pH and total dissolved solids.

As can be seen from Table 5, these techniques do not permit the reporting of all species among them cyanate (CNO⁻) and thiocyanate (SCN⁻) which are evidently not-negligible sources of toxic materials. Species missed by these techniques are considered innocuous although there is ample evidence indicating that microorganisms, certain plants, crustaceans and other aquatic organisms such as fish and frogs are highly sensitive to their toxic effects.

Accumulation of cyanides is also known to occur in some plants and also fish. These include complexes and SCN⁻ . Furthermore, not only fatal toxicity, but toxicity that reduces the agility of these organisms as well as their reproductive cycles should not be overlooked. Sometimes, low levels of ingested toxic concentrations, become lethal when temperature or oxygen content or heavy metals as well as nitrates and ammonia concentration in the aqueous environment change. Beside heavy metals such as Pb, Hg, Cu, Au, Co etc., sulfide species arising from the presence of sulfide minerals (e.g.; sulfide, poly-thionates, thio-sulfate and sulfate) can

create thiocyanate (SCN⁻) which is a reactive and persistent species of cyanide.

Persistent cyanide compounds present two types of hazard:

- 1: They are constant sources of cyanide emission and
- 2: They act as sources of heavy metals which would otherwise have been dilute as they travel away from the source.

The stabilities of metal cyanides and complexes constitute a "hazard" because in effect, they act as repositories of cyanide until an opportunity arises for its release. Taking for example Zn(CN)₄²⁻ with a dissociation constant of 1.02 x 10⁻¹⁹. On its own in equilibrium with alkaline water it is an emitter of small concentrations of CN⁻. In contact with a sulfide containing solution, in the same environment owever, it is liable to the formation of ZnS since the solubility product of ZnS equals 1.2 x 10⁻²⁸ (Weast, 1979). The equilibrium thermodynamics of this system is clearly in favor of the displacement of CN⁻ by S²⁻. Ion-sensitive electrode measurements conducted at the author's laboratory show that such reactions occur at high rates.

Table 5. Cyanide types determined for reporting and species they may miss.

Assay type	Reports	Test conditions	Misses
Free Cyanide	OT, HCN	alkaline pH	Most compounds and complexes of Ni, Co, Au, and PGM
WAD-Cyanide	freeCN, HCN, compounds and complexes that break up at mild pH	pH ~ 4-4.5	Cyanates, SCN ⁻ , complexes of Co, Ni, Au and Fe
Total Cyanide	Compounds and complexes that break up at pH £ 1 in hot water	pH < 1 in hot water	Cyanates, SCN ⁻ , complexes of Co, Ni or PGM

4 WHAT HAPPENS TO CYANIDE

Cyanides are disseminated and modified by two major routes: (1) Natural attenuation and (2) planned treatment procedures.

4.1 Natural attenuation and breakup

Processes such as evaporation, wind action, oxidation by oxygen from air, acidulation by CO_2 / H_2CO_3 ; bacterial action, uv-radiation; dilution and attenuation by rocks and minerals, including adsorptions, precipitations and catalytic phenomena are operative natural processes. CO_2 which results in HCN generation at shallow depths of disposal lagoons is the primary operator, accounting for about 90 of all cyanide loss.

Yet, the sum of these mechanisms has only limited effect on the total rate of natural decyanidation of process waters. In tailings ponds. Even then, the final products beside HCN, CNO^- , SCN^- or NH_3 are not environmentally-friendly. Furthermore, many of those processes that involve rocks minerals and dissolved ionic species create precipitates and adsorbed species that are longer-lasting than the original free-cyanide if it were to be treated.

On the optimistic side, It appears that natural degradations in abandoned heaps can be utilized as a preliminary de-cyanidation stage followed by chemical destruction rinses of the residual cyanide.

This is a promising prospect in the exploitation of natural processes to this end. Indeed, some test data indicate that an 84000-ton heap, after 3 months of abandonment retains about 11.5% of the applied cyanide. The calculated amount of total cyanide retained in the pores of this heap is almost 4.4 tons. What is more interesting is that after another 18 months, some 85% of the cyanide present had been naturally-degraded (Huiatt 1983).

Table 6. Natural cyanide attenuation processes and products.

ATMOSPHERE

Operators and reactants : Air currents, HCN, O_2 , CO_2 , H_2O , uv-radiation, air-borne-micro-organisms

Processes and Products : Dilution and dispersion processes and decompositions giving NH_4^+ , HCOO^- and HCN

WATER ENVIRONMENTS

Operators and reactants : HCN, CN^- , O_2 , Metals (e.g.: Ni^{2+} , Cu^{2+} , Fe^{3+} , etc.), S^{2-} , microorganisms, mineral catalysts.

Processes and Products: Hydrolysis " HCOO^- ", NH_3 /
Precipitations and complexations, e.g. $\text{Ni}(\text{CN})_4^{2-}$, $\text{Cu}(\text{CN})_2$, $\text{Fe}(\text{CN})_6^{4-}$, SCN^- and degradations by uv-radiation e.g. CN^- , CNO^- , NH_3 , HCOO^- , NO , etc.

SOIL ENVIRONMENTS

Operators and reactants; HCN, CN^- , H_2O , sulfur, micro-organisms and enzymes, metals, mineral catalysts

Processes and Products: Products, as in water-environments also numerous biodegradation products including NH_3 , CO_2 , CR , SCN^- and $\text{Me}(\text{CN})_x$ species

Table 7. Some methods used for cyanide abatement.

	Effluent treatment method(s)	
<i>Cyanide destruction by oxidation</i>	<i>Cyanide destruction by photolytic methods</i>	<i>HCN vaporization reduced solution pH</i>
Direct radiation	Electrical potential	Hydrolysis, distillation
With Ozone	Electrical potential with chlorine in medium	AVR
With hydrogen peroxide	Chlorine/hypochlorite	SO ₂ /air
With catalysts	Oxygen	Caro's acid
	Ozone	
	Hydrogen peroxide	
	Hydrogen peroxide plus Cu^{2+}	
	Hydrogen peroxide plus Castone (proprietary reagent)	

These natural processes and their products are compiled in Table 6. Thiocyanate, cyanate and metal cyanides in this table are the persistent ones which act, essentially, as constant reservoirs and emitters of cyanides and metal-ions.

4.2 Planned treatment procedures

The printed literature contains numerous cyanide removal and/or destruction methods (Davuyt, 1991; Botz, 1998; Robbins, 1996). These include :

Physical separations, biological procedures and chemical destruction methods.

4.2.1 Physical separations

Membrane filtration, electro and reverse osmosis, use of zeolites and ion exchange resins, or indeed adsorption on active carbon are typical methods of physical separation.

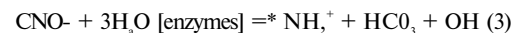
Ion exchange or reverse osmosis may be used as "polishing steps" for treated water. The problem is: they are complex and costly to operate and generate their own saline solutions which need special arrangements for disposal. Presently active carbon is a potential alternative when all else fails. Its additional advantage is that it removes tons beside free cyanide and by-products such as Pb, Cu, Hg and also WAD cyanides.

Admixture of metal salts or metal oxides, hydroxides or composite ores such as the iron-rich bauxite with contaminated solutions can remove cyanide by the formation of $K_3Fe(CN)_6$, $Cu_2Fe(CN)_6$ or $Zn_2Fe(CN)_6$ followed by filtration. Such substances during impoundment however, can decompose by the influence of solar radiation, bacterial action or catalytic effects and emit HCN.

These hybrid processes have not been demonstrated at industrial scale.

4.2.2 Biological oxidation

Micro-organism based oxidations can be represented by the following simplified equations:



Use of bacteria such as "Pseudomonas Pseudoalkaligenes" or "Bacillus Pumilus" isolated from mine-waters are known to degrade cyanide (Arps, 1994). One plant in the USA uses bacteria to remove cyanide and some of the heavy metals from the effluents of a metallurgical plant. In this practice a biological treatment by the "attached growth method" uses rotating biological contactors to facilitate the removal of cyanide, thiocyanate and some toxic metals from effluent water. The primary problems with this approach relate to the necessity of controlling temperature, bacterial nutrients such as phosphate and difficulty to maintain reproducible strains of bacteria.

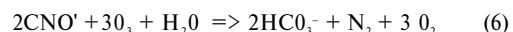
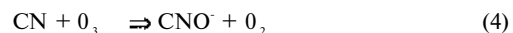
4.2.3 Chemical breakup methods

Cyanide can be converted to other chemical compounds by numerous methods. A number of these are given in Table 7. Four widely-used industrial methods include the following: (i) Alkali chlorination, (ii) Hydrogen peroxide treatment,

(iii) SO_2 / air oxidation and (iv) Volatilization from acidified effluent solutions or slurries. Brief discussions of these methods are given below.

4.2.3.1 Ozone treatment

Ozone gas, is a strong oxidant and functions as shown in equations 4-6. It is used to a limited extent.



One property of ozone is that after the initial formation of SCN in sulfur containing media, It generates HCN from thiocyanate; it needs to be used at $pH < 11$ and plus, it is a costly substance to utilize at the industrial quantities needed.

4.2.3.2 Alkali chlorination

This is a well-established method that uses chlorine gas (Cl_2), sodium hypochlorite ($NaOCl$) or calcium hypochlorite $Ca(OCl)_2$].

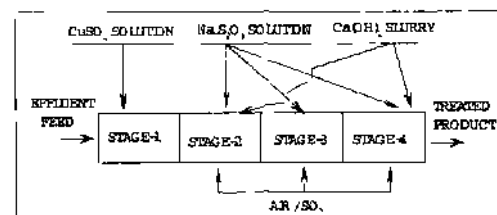


Figure 2. The INCO- SO_2 /Air process (schematic). $pH=10$, CaO 2-4 g/g cyanide, (SO_2/Air) 2-10/100, SO_2 3-6 g/g cyanide, $T=5-60^\circ C$, Cu^{2+} = 0-50 mg/L

The basic reactions in this approach consist of a number of steps summarized in equations (7) and (8).

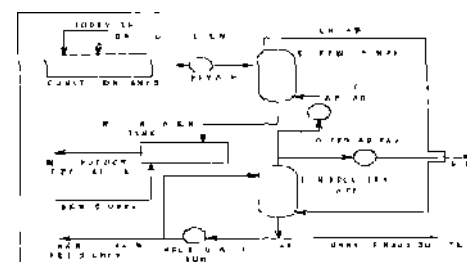
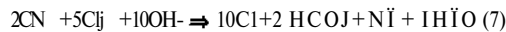


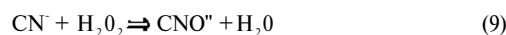
Figure 3 Scheme for the partial recycling of cyanide by the Cyan I sorb technology.



The problem with hypochlorite usage is that it generates (CNO⁻) which is objectionable. Application of chlorine gas needs provisions for the capture of fugitive gas while sodium and calcium hypochlorites are water-soluble powders.

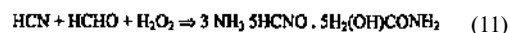
4.2.3.3 Hydrogen Peroxide

H₂O₂ as cyanide destructant, acts as follows :



Reaction (9) needs high pH, (which is already present in gold processing effluents) and Cu²⁺ ions to proceed, while reaction (10) occurs at slightly acidic aqueous environments, without need for cupric ions.

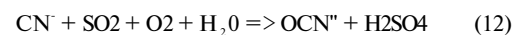
An alternative H₂O₂ - based reagent is commercially available (DuPont) and primarily consists of a formulation of hydrogen peroxide with formaldehyde (HCHO) with some proprietary additives. The mixing of reagents is conducted at about 50 °C and the net reaction is given as:



The problem is that, not all of the CN⁻ is removed from the medium. Additional treatment technologies are needed for the satisfactory de-cyanidation of effluents.

4.2.3.4 SO₂ / air oxidation

The use of sulfur dioxide-air mixtures in the presence of Cu²⁺ as catalyst forms the basis of the INCO (International Nickel Company) process (Robbins, 1996) where the reaction is represented as follows:



One of the arguments made against alkali chlorination (and in favor of the INCO process) is that if the effluent slurries contain iron arising from pyrite or pyrrhotite, ferro and ferricyanides are formed. These solids in turn can generate HCN when exposed to solar radiation at solid disposal sites. The INCO process is said to be among the ones that generate the least of harmful compounds, though, it may generate large quantities of CaSO₄ - rich sludge which adds to the cost of the process. A

simplified flowsheet for the application of this technology is given in Figure 2.

There are more than 30 plants that use the INCO process in North America; worldwide, more than 50 projects are reported to have licensed this technology. The approach has been shown to be applicable to solutions, slurries and abandoned heaps (as heap rinse).

4.2.3.5 Volatilization from acidic solutions

The HCN speciation diagram given in Figure 1 shows that at pH < 9.3, HCN gas is the predominating hydrolytic species. Thus the HCN gas can be driven off from the aqueous medium by distillation, or displacement by sparged air. Then HCN in the gaseous mixture can be captured by an alkaline solution such as NaOH or Ca(OH)₂ in a subsequent process step. This is in principle, forms the basis of AVR (acidification-vaporization-re-adsorption) technologies. A simplified flowsheet for the use of this approach is given in Figure 3.

One potential drawback relates to the safety of operators of such plants because HCN gas which is highly toxic is present in concentrated form in the plant pipeline system though, no incident has been recorded in some 8 plants that have operated using this technology. A synopsis of

Treatment technologies with critical commentary are summarized in Table 8.

5 CYANIDE DESTRUCTION ECONOMICS

Two motivations that form the basis for cyanide abatement and control are obvious: 1) Recycling of a valuable resource and 2) Destruction of a toxic component added to nature by man.

One of the alternatives for the minimization of cyanide emission is to reduce its use whenever possible. For example in flotation technology where cyanide is used as a selective-depressant for certain minerals such as copper or say, chromium-containing minerals sodium oxalate or sodium thiosulfate have been shown to be usable. Similar in the recycling of scrap metal, or exposed photographic film, thiosulfate or nitric acid solutions can be readily used as alternative lixivants.

If the economics of cyanide usage in a given precious metal recovery approach is considered successful, the problem then boils down to the toxicity of cyanide and its compounds and the resulting management of this situation. The toxicity problem can conceptually, be solved by three approaches two of which were cited above. "Replacement of cyanide by alternative lixivants" would be an added method of cyanide abatement. (Yarar 1993; Yarar, 1999).

Table 8. Cyanide abatement and destruction technologies for CN⁻ - containing effluents.

<i>Method</i>	<i>Primary Mechanism(s)</i>	<i>Demonstrated scale</i>	<i>Advantages</i>	<i>Disadvantages</i>
Dialysis, E-osmosis	Membranes	Lab.	Future Potential	Cost; Maintenance
Ion Exchange Resins	Ion-exchange	Lab. ; pilot	Potential for cyanide recycling	Cost; Maintenance
Metal salts and/or Mineral powder addition	Precipitation, Adsorptions; Catalysis	Small scale and pilot	Capture of ions and suspended solids	Product solids removal and disposal; materials volume
Active carbon contact	Physical & chemical uptake; Catalysis	Lab. and small scale	Widely effective on ions and solids	Cost, Fouling of carbon; catalytic-byproducts
Ion or precipitate flotation	Surface chemistry of foams and solids	Lab. demonstration	Potential for treatment of large volumes	Large scale demonstration and; supplementary techniques needed
Direct and With O ₃ , H ₂ O ₂ or Sensitizing Solids : ZnO, TiO ₂	UV-and catalysis-induced redox	Commercially available small-scale	Demonstrated technology	Cost, UV radiation does not penetrate water, special reactor design needs.
Electrical potential	Oxidation	Lab and large pilot	Demonstrated technology, can be combined with O ₂ , O ₃ , Cl ₂ , or OCl	Potential for poisonous by products
Ozone	Oxidation of CN ⁻	Industrial	Cost -effective; minimal amount of harmful end-products	More effective at low pH, while CN ⁻ -effluents are alkaline
Hydrogen peroxide	Oxidation of CN ⁻	Industrial, can be used with accelerators and catalysts	Cost -effective; proven technology	Cost of pH control when used with acids; Can produce nitrite and nitrate as byproducts
SO ₂	Oxidation	Industrial	Proven technology can be accelerated with Cu ²⁺	pH-control needed; sludge formation and handling is problematic
Chlorine (Cl ₂) and hypochlorite (OCl)	Oxidation of CN ⁻	Industrial	Versatile, usable at small and large scale	Minimal
H ₂ SO ₄	HCN-generation	Pilot	Allows recycling of CN ⁻	
Bio-oxidation	Metabolic and enzymatic oxidation and metabolic adsorption	Industrial with some strains and pilot or lab. - scale with others	Partly proven technology, promising potential; partial removal of metal ions also occurs.	pH and nutrient control may be needed; biomass and bacterial strain control may create problems

The cost of conventional destruction technologies can be up to \$1.50 per ton of ore treated. Recycling, provided by the AVR technology is an obvious abatement technique which is only partial at the present.

6 CONCLUSIONS

A study of the published literature on cyanidation-based metal extraction technologies and their cyanide-based products, allows the following conclusions:

1. Analytical data reporting is not as best it could be because it omits numerous cyanide complexes and compounds.

2. It is assumed that free cyanide, in nature, breaks down completely to CO₂ or nitrated compounds, which is not often the case. Compounds with heavy metals do form and resist destruction in natural environments for long periods of time. Cyanide compounds assumed to be "destroyed" or "not-present" are in fact, in place and are continually emitting harmful/toxic components.

3. Presently, a universal panacea to all cyanide-related problems does not exist although numerous technological approaches are available in the marketplace and most commercialized cyanide destruction processes claim superiority to competitors, in some form or another.

4. Cyanide usage is not about to cease any time soon, nor is mere need for that although research is needed for a less controversial and equally-effective alternative lixiviant.

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