**NUCLEAR CHEMICAL COPPER MINING AND REFINING:
RADIOLOGICAL CONSIDERATIONS**

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NUCLEAR CHEMICAL COPPER MINING AND REFINING: RADIOLOGICAL CONSIDERATIONS

Abstract

A preliminary radiological safety analysis of the proposed nuclear chemical mining and refining process for copper has been carried out. Exposure sources and paths, exposed populations, and dose levels are all discussed. Existing data were utilized in the formulation of this safety analysis, and conservative (i.e.,

"worst case") assumptions were used where data were unavailable. Despite the "upper limit" formulation of hypothetical radiological exposures to members of the public, all such exposures were found to be far below (much less than 1% of) current radiation protection guides.

Introduction

A preliminary radiological safety analysis of the proposed nuclear chemical mining and refining of copper is required in order to define potential problem areas. Studies designed to take us toward this goal are underway.

A large amount of data has been obtained which pertains to the behavior of radioactivity under conditions similar to those expected in certain copper leaching situations. However, recently suggested improvements in the method by which copper-containing minerals (mainly chalcopyrite) may be dissolved through in situ treatment of copper ore have resulted in changes in the prevailing view of the chemical environment which may

exist during copper leaching. Since this evaluation of radiological safety has used existing data together with conservative assumptions where pertinent information is lacking, the results should be viewed as preliminary and conservative in nature. It is desirable that additional data be obtained in order that a more realistic evaluation of radiological safety may be prepared. While it is expected that the conclusions of such further work will indicate even lower potential radiation exposures to workers and nearby residents, better grounded radiological safety analyses are desirable, both for plant design studies and for future environmental impact assessments.

The Mining Process

Considerable success has been attained in the recovery of copper from copper sulfide and oxide ores in waste dumps by means of both heap leaching and solution mining.¹ The sulfuric acid and ferric sulfate solutions used in such mining processes cannot, however, be utilized for recovery of copper from primary sulfide ores. Not only are such ores generally too impermeable for successful leaching, but also the primary ore minerals of interest (mainly chalcopyrite) are essentially insoluble in these commonly used leach liquors.

Recently, it has been proposed that relatively impermeable copper ore bodies be fractured by means of nuclear blasting, and that this fractured ore be leached with water containing dissolved oxygen under relatively high hydrostatic pressure²⁻⁴ (see Fig. 1). As has been discussed in Refs. 2, 3, and 4, the conditions existing in a typical nuclear chimney during such copper ore leaching would be as shown in Table 1. The rate of chalcopyrite (copper ore) dissolution is dependent upon temperature, oxygen pressure, and acidity. Since the

Fig. 1. Chemical mining of chimney rubble.

Table 1. Physical and chemical conditions during copper leaching.

Temperature	60°C - 150°C
O ₂ pressure	400 - 1000 psi
pH	1.0 - 2.5

oxidation reactions of the sulfide ores are highly exothermic, dissolution of these ores will maintain the requisite high temperature, and the rate at which copper is recovered will be dependent on the rate of diffusion of solution into (and out of) the chunks of ore present in the nuclear chimney.

The present concept of the mining process calls for the flooding of the ore-containing nuclear chimney with water (which can be pre-heated to the desired temperature for rapid initiation of the leaching process), immediately followed by the introduction of oxygen gas at hydrostatic pressure through pipes introduced into the bottom portion of the chimney. When the copper content of

the leach liquor has reached a pre-selected value, a portion of the pregnant liquor will be tapped off for processing (copper removal), and the stripped liquor will be returned to the cavity. This process of dissolution and recovery is estimated to continue for approximately five years, or until the return of copper drops below that which is required for economic operation of the process.

It should be noted that during the dissolution of the copper in the chimney rubble and prior to extraction, a complex chemical system is established. While the copper and iron sulfides (chalcopyrite and pyrite) are being dissolved, the breakdown of rock minerals is progressing and byproducts such as anhydrite (CaSO_4), jarosite ($\text{H}_2\text{Fe}_6(\text{OH})_{12}(\text{SO}_4)_4$), alunite ($\text{K}_2\text{Al}_6(\text{OH})_{12}(\text{SO}_4)_4$), montmorillonite, etc. are being precipitated. It is expected that the chemical state and availability of the radioactive elements in the cavity will be markedly affected by the complex reactions taking place in the leach liquor.

The Refining Process

Preliminary investigations⁵ indicate that the most suitable refining process which can be used with the proposed chemical mining technique involves organic solvent extraction of copper from the leach liquor, stripping the copper from the organic phase and subsequent electrowinning. A typical flowsheet for this process is shown in Fig. 2.

As noted in this figure, the pregnant leach liquor (at approximately pH 2) is subjected to batch extraction with a patented chelating agent dissolved in

kerosene (or a kerosene-like solvent). The organic phase containing the copper may be scrubbed with water as necessary, while the raffinate (depleted leach liquor) is recycled to the nuclear chimney as noted in the preceding section. Next, the copper is stripped from the organic phase with an aqueous solution of sulfuric acid and the chelate-organic solution is reused. Finally, the acid-copper solution is used as feedstock for a conventional electrowinning system, where the copper is electrolytically deposited. As processing

Fig. 2. Schematic flowsheet for processing of chimney rubble.⁶

Table 2. Chemical characteristics of the solvent extraction/electrowinning copper refining process.

Solution	Copper concentration (g/liter)	Acid (H_2SO_4) concentration
Leach liquor	>2	pH 1.0-2.5
Raffinate (leach liquor after extraction)	~0.02	pH 1.0-2.5
Organic extract solution (0.10 g/ml LIX 64 in Varsol)	~2	—
Stripped organic	~0.05	—
Scrub solution after scrubbing (water wash of organic phase)	~0.01	pH 2.4
Strip solution (electrolyte before Cu electrodeposition)	~23	4 <u>N</u>
Depleted electrolyte	~18	4.2 <u>N</u>

proceeds, the depleted electrolyte is used to strip succeeding batches of organic extract in order to build up the copper concentration in the electrolyte for further recovery by electrowinning.

The chemical concentrations shown in Table 2 are typical of the various solutions used in the solvent extraction/electrowinning copper refining process.

As indicated in the preceding section, complex breakdown products of rock minerals will be present in the leach liquor. It is possible that some of these

substances may be precipitated or otherwise removed from solution in the course of the refining process. Some radioactivity could be carried along with these mineral breakdown products. Since pockets of radioactive solids could build up at some places in the refining plant, appropriate experimental studies should be carried out to identify possible concentration mechanisms. From this information, the refining process can be designed so as to minimize the formation of radioactive "hot spots" in the operating refinery.

Radioactivity Source Term

PRODUCTION OF RADIOACTIVITY

The specific radionuclides produced by a nuclear detonation of the type that could be used to fracture copper ore are much the same irrespective of the design of the nuclear explosive which is used; however, the relative amounts of the various radioactivities formed is critically dependent on the fission-to-fusion ratio of the explosive. Table 3 shows the fission-product radioactivity present 180 days after the detonation of a 100-kt all-fission explosive. Should a 100-kt thermonuclear explosive with a 5-kt fission trigger be employed, the amounts of all of the radionuclides shown in Table 3 should be reduced twentyfold. With regard to the radioactive products formed when neutrons emitted from the explosive are captured in the surrounding rock, the amounts of these products are influenced both by the composition of the rock and by the number of neutrons escaping from the nuclear explosive. Since both fission

and thermonuclear explosives emit* approximately 2×10^{23} neutrons per kt of yield,⁷ the same amounts of the various activation products are produced, irrespective of the type of explosive. Thus, the list of radioactivities given in Table 4 represents rock activation produced by either a fission or a thermonuclear explosion. The one radioactive species shown in Table 4 which is sensitive to the type of explosive used is ^3H (tritium). Since approximately 2 g of this nuclide is formed per kiloton of thermonuclear yield,⁹ use of a fusion explosive of the type previously noted (5-kt fission trigger) would produce some 2,000,000 Ci of tritium, as against the less than 2000 Ci produced in a typical gas-well environment by the specially designed gas-stimulation explosive.¹⁰

It should be noted that the radioactive species formed by neutron absorption

* Assuming that the explosives do not employ neutron "shields" to diminish the number of neutrons released into the surroundings.

Table 3. Radionuclide source at D + 180 days after detonation of a 100-kt all-fission explosive (assumes ^{235}U fission).^a

Nuclide	Fission Products		Predominant location of radioactivity
	Half-Life	Curies	
^{85}Kr	10.8 yr	2,200	gas
^{89}Sr	52 da	220,000	chimney - on rock surfaces
^{90}Sr	28 yr	15,000	chimney - on rock surfaces
^{91}Y	59 da	360,000	chimney - on rock surfaces
^{95}Zr	65 da	440,000	melt - in vitreous matrix
^{95}Nb	35 da	800,000	melt - in vitreous matrix
$^{103}\text{Ru}^b$	40 da	105,000	chimney - on rock surfaces
$^{106}\text{Ru}^b$	1.0 yr	38,000	chimney - on rock surfaces
$^{115\text{m}}\text{Cd}$	43 da	120	chimney - on rock surfaces
^{123}Sn	125 da	600	chimney - on rock surfaces
^{124}Sb	60 da	50	chimney - on rock surfaces
^{125}Sb	2.7 yr	6,000	chimney - on rock surfaces
$^{125\text{m}}\text{Te}^b$	58 da	1,100	chimney - on rock surfaces
$^{127\text{m}}\text{Te}^b$	109 da	7,000	chimney - on rock surfaces
$^{129\text{m}}\text{Te}^b$	34 da	1,700	chimney - on rock surfaces
$^{137}\text{Cs}^b$	30 yr	18,000	chimney - on rock surfaces
$^{140}\text{Ba}^b$	12.8 da	900	chimney - on rock surfaces
^{141}Ce	33 da	130,000	melt - in vitreous matrix
^{143}Pr	13.6 da	1,800	melt - in vitreous matrix
$^{144}\text{Ce}^b$	284 da	360,000	melt - in vitreous matrix
^{147}Pm	2.6 yr	70,000	melt - in vitreous matrix
^{151}Sm	87 yr	500	melt - in vitreous matrix
^{155}Eu	1.8 yr	3,000	melt - in vitreous matrix

^aThe amount of each fission product is consistent with the assumed production given in Ref. 8.

^bRelatively short-lived radioactive daughters are in transient equilibrium with these fission products; however, the total activity (in curies) listed for these fission products represents only that from the parent nuclide.

which are listed in Table 4 represent only those resulting from activation of the rock surrounding the explosive. Additional radioactivity can be produced by neutron activation of the materials used in the construction of the explosive itself.

The large amounts of such nuclides as ^{181}W , ^{188}W , and ^{182}Ta , which have

been observed in debris from nuclear cratering experiments,¹¹⁻¹³ indicate that care must be used in the design of a thermonuclear explosive to prevent the formation of radionuclides which could adversely affect the feasibility of nuclear chemical mining techniques.

Table 4. Soil activation products (assumes Hardhat granite with 1 wt% FeS₂, 1 wt% CuFeS₂, and 5 wt% water).

Nuclide	Half-Life	Curies	Predominant location of radioactivity
³ H	12 yr	<2,000 ^b	water
¹⁴ C	5730 yr	7	gas
³² P	14 da	170	chimney - on rock surfaces
³⁵ S	88 da	15,000	chimney - on rock surfaces
³⁷ Ar	35 da	7,200	gas
³⁹ Ar	270 yr	8	gas
⁴⁵ Ca	165 da	20,000	melt - in vitreous matrix
⁴⁶ Sc	84 da	1,400	melt - in vitreous matrix
⁵⁴ Mn	303 da	180	melt - in vitreous matrix
⁵⁵ Fe	2.6 yr	49,000	melt - in vitreous matrix
⁵⁹ Fe	45 da	1,700	melt - in vitreous matrix
⁶⁰ Co	5.3 yr	120	melt - in vitreous matrix
⁶⁵ Zn	245 da	1,000	melt - in vitreous matrix
⁸⁵ Sr	64 da	160	melt - in vitreous matrix
⁸⁹ Sr ^a	52 da	550	melt - in vitreous matrix
⁹⁵ Zr ^a	65 da	90	melt - in vitreous matrix
⁹⁵ Nb ^a	35 da	150	melt - in vitreous matrix
¹³⁴ Cs	2 yr	1,500	chimney - on rock surfaces
¹⁴¹ Ce ^a	33 da	130	melt - in vitreous matrix
¹⁵² Eu	12 yr	600	melt - in vitreous matrix
¹⁶⁰ Tb	72 da	830	melt - in vitreous matrix
¹⁷⁰ Tm	130 da	2,900	melt - in vitreous matrix
¹⁹² Ir	74 da	900	melt - in vitreous matrix

^aAlso formed as a fission product.

^bPublished values for gas-stimulation fission explosive detonated in Wagon Wheel environment. While the lithium content of Hardhat granite is higher than that of Wagon Wheel rock, the lithium content of certain copper ores (such as that from Safford) is approximately equal to that of Wagon Wheel rock. Hence, for this study, we will use the Wagon Wheel tritium source term. For a 100-kt thermonuclear explosive, on the order of 2,000,000 curies of tritium would be formed.

INITIAL DISTRIBUTION OF RADIOACTIVITY

The sequence of events that occur when a buried nuclear explosive is detonated is shown schematically in Fig. 3. Approximately 70-100 tons of rock, as well as the explosive itself, are vaporized for each kiloton of nuclear yield. As the shock wave moves through the surrounding

material, still more rock is melted; as the cavity expands, superheated molten and vaporized rock melts still more rock in the immediate cavity environs. The cavity stabilizes in size in less than 1 second after the detonation, with the cavity pressure approximately equal to the lithostatic pressure at the detonation point; at this time, the spherical cavity

is lined with about 1000 tons* of molten rock per kiloton of nuclear yield.¹⁴

Shortly thereafter, most of this molten rock flows downward to form a puddle at the bottom of the cavity, and still later, the roof of the cavity begins to collapse. This collapse continues to propagate upward until the top of the shock-fractured rock region surrounding the detonation point is reached, or until the cavity bulks full of rubble (thus supporting the rock at the top of the chimney).

The distribution of the various radioactive nuclides formed by the nuclear detonation is dependent on the chemical

characteristics of each of these species at the time during which chimney formation takes place.¹⁵ That is, those radioactive substances which act in a refractory manner will largely be found in the so-called "puddle glass" or melt at the bottom of the chimney, while those which display some degree of volatility will be found to a greater or a lesser extent to be deposited on the surfaces of the chimney rubble or in the void volume between the rubble fragments. Table 5 gives a simple categorization of various typical radionuclides which could be found in debris from a nuclear detonation.

Those radioactive isotopes that are described as volatile appear to behave as such because either they themselves or one of their predecessors in a radioactive decay chain exists in a volatile chemical

*This molten rock may be at a temperature above the melting point of the surrounding rock; hence, additional rock may be melted at later times as heat flows from the molten rock into its surroundings.

Fig. 3. Cavity and chimney formation history.

Table 5. Chemical behavior of selected radioactive species.

	Volatile	Nonvolatile
Fission products	^{89}Sr , ^{90}Sr , ^{91}Y ^{103}Ru , ^{106}Ru ^{137}Cs	^{95}Zr , ^{95}Nb , ^{97}Zr ^{144}Ce ^{147}Nd , ^{151}Sm
Activation products	^{32}P	^{45}Ca , ^{54}Mn ^{56}Mn , ^{55}Fe , ^{59}Fe
Fusion products	^3H	

form for some time during the distribution process. The former situation may be exemplified by such species as ^{103}Ru or ^3H while the latter is typified by the mass-90 decay chain: $^{90}\text{Br} \rightarrow ^{90}\text{Kr} \rightarrow ^{90}\text{Rb} \rightarrow ^{90}\text{Sr}$, where, of course, the ^{90}Kr is a noble gas and the ^{90}Br and ^{90}Rb may be volatile under the conditions prevailing at the time of cavity collapse.

The refractory radioactive substances, in an analogous way, must themselves be refractory in nature or must be the end product of a radioactive decay chain all the members of which are also refractory.

On the basis of this chemical differentiation, the predominant location of those nuclides listed in Tables 3 and 4—within the glass matrix or on the rubble surface—can be established as shown in the fourth column of those tables.

CONCENTRATION OF RADIO-ACTIVITY IN "CHIMNEY GAS"

In the first few days following the nuclear detonation, the void spaces present in the chimney are filled with gas; principally CO_2 released from carbonate minerals which may be present in the region around the detonation point. On the basis of experimental determina-

tions carried out in conjunction with both the Gasbuggy and Rulison nuclear gas-stimulation experiments, we will assume that approximately 2000 tons per kiloton of the rock surrounding the explosive will be heated to a sufficiently high temperature so that all of the carbonate present will be released as CO_2 . In the following calculations it has been necessary to assume certain of the characteristics for a nuclear chimney resulting from a nominal 100 kt nuclear detonation; these are presented in Table 6. If it is further assumed that the rock heated by the explosion contains the equivalent of 2 wt% CO_2 , some 4×10^9 g of CO_2 would be present in the cavity. Again based on Gasbuggy¹⁶ and Rulison¹⁷ experience, some water vapor should be present in

Table 6. Chimney properties assumed.

Device yield—100 kt
Depth of burial—750 m
Cavity radius—41 m
Chimney height—300 m
Void volume } — $2.9 \times 10^5 \text{ m}^3$
Water volume }
Mass of chimney rock— 3.8×10^6 metric tons
Mass of melt—1000 tons/kt

Table 7. Fission-product and rock-activation radionuclides present in cavity.

Nuclide	Half-life	Ci(D+180)	Concentration in cavity gas ($\mu\text{Ci/ml}$)		(MPC) _a ^e ($\mu\text{Ci/ml}$)	Dilution ^f necessary
			Downhole conditions ^a	STP ^b		
³ H	12 yr	<2,000 ^{c,d}	7×10^{-6}	1×10^{-6}	2×10^{-7}	5
¹⁴ C	5730 yr	7	2.4×10^{-5}	3×10^{-6}	1×10^{-7}	30
³⁷ Ar	35 da	7,200	2.5×10^{-2}	3×10^{-3}	1×10^{-4}	30
³⁹ Ar	270 yr	8	2.8×10^{-5}	4×10^{-6}	— ^g	—
⁸⁵ Kr	10.8 yr	2,200 ^d	$8. \times 10^{-3}$	1×10^{-3}	3×10^{-7}	3000

^aAt 10 atm pressure and 90°C.

^bStandard temperature and pressure.

^cAs noted in the text, only 0.1% of the tritium will be present (as water vapor) in the gas.

^dShould a thermonuclear explosive be employed, the tritium source term and concentrations should be multiplied by 1000, while the analogous values for the ⁸⁵Kr should be multiplied by 0.05.

^eMaximum permissible concentration (MPC)_a in air for a member of the public (Ref. 18).

^fAmount of dilution required for the concentrations shown in Column 5 to be reduced to the (MPC)_a values given in Column 6.

^gNo value quoted.

the gas. Using the approach delineated in Ref. 8, the ambient temperature of the gas would be about 90°C, and the gas would thus contain approximately 1.2×10^8 g of water vapor.

While all of the gaseous radioactive species ⁸⁵Kr, ³⁷Ar, and ¹⁴C (presumably as ¹⁴CO₂) are assumed to be present in this cavity gas, the ³H would be distributed between water vapor and liquid water. If it is assumed that the exchangeable water pool is represented by the unbound water in the chimney rubble (about 2-1/2 wt%, or $\sim 10^{11}$ g), then about 0.1% of the ³H would be present in the cavity gas. On this basis, then, the concentrations of radioactive species in the gas can be calculated; these are shown in Table 7.

CONCENTRATION OF RADIO-ACTIVITY IN LEACH SOLUTION

For the purposes of this treatment, it will be assumed that those radioactive substances deposited on chimney rubble are completely soluble,^{*} while those present in the melt are soluble to the extent of 1%.[†] On the basis of data obtained

^{*}Note that this assumption actually is very conservative due to two factors:

- 1 not all of the so-called "volatile" radioactive species are present on rubble surfaces; actually, a portion of each of these species will be incorporated into a glass matrix; and
2. even that portion of the radioactivity which is surface-deposited on the chimney rubble is not necessarily completely soluble in a leach solution.

[†]This assumption is consistent with data on the dissolution rate of powdered melt samples.

at the Oak Ridge National Laboratory (see Ref. 5) these assumptions appear to be between a factor of 2 and a factor of 5 too high. However, it should be noted that while the Oak Ridge determinations were carried out using sulfuric acid solutions, the temperatures and oxygen content of the solvent were significantly different from those expected for the *in situ* mining process as discussed earlier. Thus, the solubilities used in this work were chosen in an attempt to take into account the different solvent properties. However, the concentration of radioactivity in the circulating leach solution is dependent not only on the initial solubility of the radionuclides present in the chimney but also on the degree to which these species remain in solution in the complex chemical environment of the leach liquor. Not only will some of the dissolved radioactive substances be adsorbed onto the large rock surface area of the chimney, but also a number of the radionuclides in solution would be expected to be coprecipitated with the breakdown products of rock minerals which are produced in the course of the oxidation of copper and iron sulfides. For instance, it has been experimentally verified that a significant fraction of the dissolved ^{90}Sr is carried down with precipitating anhydrite.

However, since the behavior of all the various radioactive substances in this solution containing rock mineral breakdown products has not yet been established, it will be conservatively assumed here that the only removal process is the sorption of these radioactive substances in the rock surfaces of the chimney rub-

ble.* For a number of species, this assumption may result in estimated amounts in solution which are too large by as much as a factor of 10. Work reported in Ref. 5 and shown in Fig. 4 indicates that the degree of sorption of a number of radioactive contaminants will be relatively high. Applications of these data, and of data obtained at the Lawrence Livermore Laboratory (LLL), to all species of interest and the estimated concentrations of these radioactivities in the leach solution are shown in Tables 8 and 9. Note that for species where no data are available, it has been conservatively

*With the exception of $^{89,90}\text{Sr}$, for which preliminary measurements of the coprecipitation effect have been made.

Fig. 4. Adsorption of radionuclides.

Table 8. Chimney radionuclides: estimated concentrations in solution at 180 days (assumes: 100% availability; $2.9 \times 10^5 \text{ m}^3$ solution; 25% of rock is effective in sorption; no coprecipitation with rock mineral breakdown products except as noted).

Nuclide	Half-life	Ci (D+180)	K_d^a	$\frac{1}{(1+rK_d)^b}$	Chimney concentration ($\mu\text{Ci/ml}$)	(MPC) $_w^c$ ($\mu\text{Ci/ml}$)	No. of MPC's	Time to decay to 1 MPC	Time to decay to 0.1 MPC
^3H	12 yr	<2,000	0^e	1	$<6.9 \times 10^{-3}$	3×10^{-3}	2.3	< 15 yr	< 55 yr
^{32}P	14 da	170	0^d	1	5.9×10^{-5}	2×10^{-5}	29	0.2 yr	0.3 yr
^{35}S	88 da	15,000	0^d	1	5.2×10^{-2}	6×10^{-5}	860	2.4 yr	3.2 yr
^{89}Sr	52 da	220,000	$\sim 100^f$	0.003	2.3×10^{-3}	3×10^{-6}	760	1.4 yr	1.8 yr
^{90}Sr	28 yr	15,000	$\sim 100^f$	0.003	1.6×10^{-4}	3×10^{-7}	520	250 yr	350 yr
^{91}Y	59 da	360,000	1.5	0.2	0.23	3×10^{-5}	7500	2.1 yr	2.6 yr
^{103}Ru	40 da	105,000	50	0.006	2.2×10^{-3}	8×10^{-5}	27	0.5 yr	0.9 yr
^{106}Ru	1.0 yr	38,000	50	0.006	7.9×10^{-4}	1×10^{-5}	79	6 yr	10 yr
$^{115\text{m}}\text{Cd}$	43 da	120	0^e	1	4.1×10^{-4}	3×10^{-5}	14	0.4 yr	0.8 yr
^{123}Sn	125 da	600	15^e	0.02	4.1×10^{-5}	—	—	—	—
^{124}Sb	60 da	50	300	0.001	1.9×10^{-7}	2×10^{-5}	0.01	—	—
^{125}Sb	2.7 yr	6,000	300	0.001	2.3×10^{-5}	1×10^{-5}	2.3	3.2 yr	12 yr
$^{125\text{m}}\text{Te}$	58 da	1,100	0^d	1	3.8×10^{-3}	2×10^{-4}	19	0.7 yr	1.2 yr
$^{127\text{m}}\text{Te}$	109 da	7,000	0^d	1	2.4×10^{-2}	6×10^{-5}	400	2.6 yr	3.6 yr
$^{129\text{m}}\text{Te}$	34 da	1,700	0^d	1	5.9×10^{-3}	3×10^{-5}	200	0.7 yr	1.0 yr
^{134}Cs	2 yr	1,500	5	0.06	3.1×10^{-4}	9×10^{-6}	34	10 yr	17 yr
^{137}Cs	30 yr	18,000	5	0.06	3.7×10^{-3}	2×10^{-5}	190	230 yr	330 yr
^{140}Ba	12.8 da	900	0.7^d	0.3	9.3×10^{-4}	3×10^{-5}	31	0.2 yr	0.3 yr

^aMeasured at LLL except as noted.

^bThe fraction of a given species remaining in solution in a unit volume of solution, where r is the ratio of the effective mass of rock upon which the given species is adsorbed to the volume of water in contact with the rock. ($r \approx 3$).

^cMaximum permissible concentration in water (MPC) $_w$, Code of Federal Regulations, Title 10, Chapter 1, Appendix B-Table II.

^dAssumed.

^eFrom ORNL data (Ref. 5).

^fIncludes effects of coprecipitation with rock mineral breakdown products.

assumed that the species is not sorbed.* The characteristics of the assumed detonation, as well as of the resulting chimney, that were used in calculating the concentrations shown in Tables 8 and 9 are given in Table 6. In calculating the degree of

*Some nuclear explosives which could be used contain the fissile material ^{239}Pu . The behavior of this nuclide in the chimney environment, as well as in the refinery, is discussed in Ref. 6.

sorption, it has been assumed that the effective mass of the rock in the chimney that is in contact with the radioactive solution is 25% of the actual mass. This assumption is necessary to account for the difference in size between the chimney rubble fragments and the material used in the determination of the distribution coefficient (K_d) values (see Ref. 5; experiments conducted at LLL used rubble

fragments which were similar in size to those employed in the Oak Ridge work). While this choice of the effective rock mass is somewhat arbitrary, in view of the large effective surface enhancement of this rock which is created during the dissolution process, the assumption appears to be conservative in nature.

More realistic estimates will probably not decrease the results presented here by more than a factor of 2 or 3.

With regard to the values of K_d given in Ref. 5, it should be emphasized that, while these values are based on data from experiments conducted with a sulfuric acid leach solution having a pH of about

Table 9. Melt radionuclides; estimated concentrations in solution at 180 days (assumptions: $2.9 \times 10^5 \text{ m}^3$ solution and 1% of melt nuclides soluble, 25% of rock is effective in sorption; no coprecipitation with rock mineral breakdown products except as noted).

Nuclide	Half-life	Ci (D+180)	K_d^a	$\frac{1}{1+rK_d}^b$	Chimney concentration ($\mu\text{Ci/ml}$)	(MPC) $_w^c$ ($\mu\text{Ci/ml}$)	No. of MPC's	Time to decay to 1 MPC	Time to decay to 0.1 MPC
^{45}Ca	165 da	20,000	0^d	1	6.9×10^{-4}	9×10^{-6}	77	2.8 yr	4.3 yr
^{46}Sc	84 da	1,400	0^e	1	4.8×10^{-5}	4×10^{-5}	1.2	0.06 yr	0.8 yr
^{54}Mn	303 da	800	0^d	1	6.2×10^{-6}	1×10^{-4}	0.06	—	—
^{55}Fe	2.6 yr	49,000	0^e	1	1.7×10^{-3}	8×10^{-4}	2.1	2.8 yr	11.4 yr
^{59}Fe	45 da	1,700	0^e	1	5.9×10^{-5}	6×10^{-5}	1.0	—	0.4 yr
^{60}Co	5.3 yr	120	0^e	1	4.1×10^{-6}	5×10^{-5}	0.08	—	—
^{65}Zn	245 da	1,000	0^e	1	3.4×10^{-5}	1×10^{-4}	0.34	—	1.2 yr
^{85}Sr	64 da	160	$\sim 100^f$	0.003	1.7×10^{-8}	1×10^{-4}	0.0002	—	—
^{89}Sr	52 da	550	$\sim 100^f$	0.003	5.7×10^{-8}	3×10^{-6}	0.019	—	—
^{95}Zr	65 da	460,000	50^e	0.006	9.5×10^{-5}	6×10^{-5}	1.6	0.12 yr	0.7 yr
^{95}Nb	35 da	800,000	50^e	0.006	1.7×10^{-4}	1×10^{-4}	1.7		
^{141}Ce	33 da	130,000	20	0.016	7.0×10^{-5}	9×10^{-5}	0.82	—	0.3 yr
^{143}Pr	13.6 da	1,600	1	0.25	1.4×10^{-5}	5×10^{-5}	0.28	—	20 da
^{144}Ce	284 da	360,000	20	0.016	2.0×10^{-4}	1×10^{-5}	20	3.4 yr	6.0 yr
^{147}Pm	2.6 yr	70,000	1	0.25	6.0×10^{-4}	2×10^{-4}	3.0	4.1 yr	13 yr
^{151}Sm	87 yr	500	1	0.25	4.3×10^{-6}	4×10^{-4}	0.01	—	—
^{152}Eu	12 yr	500	2	0.14	2.9×10^{-6}	8×10^{-5}	0.04	—	—
^{155}Eu	1.8 yr	3,000	2	0.14	1.4×10^{-5}	2×10^{-4}	0.07	—	—
^{160}Tb	72 da	830	1	0.25	7.2×10^{-6}	4×10^{-5}	0.18	—	0.2 yr
^{170}Tm	130 da	2,900	1	0.25	2.5×10^{-5}	5×10^{-5}	0.5	—	0.8 yr
^{192}Ir	74 da	900	0^d	1	3.1×10^{-5}	4×10^{-5}	0.78	—	0.6 yr

^a Measured at LLL except as noted.

^b The fraction of a given species remaining in solution in a unit volume of solution, where r is the ratio of the effective mass of rock upon which the given species is adsorbed to the volume of water in contact with the rock ($r = 3$).

^c Code of Federal Regulations, Title 10, Chapter 1, Appendix B-Table II.

^d Assumed.

^e From ORNL data (Ref. 5).

^f Includes effects of coprecipitation with rock mineral breakdown products.

2, the experiments were carried out at room temperature and only that amount of oxygen in equilibrium with the solution at atmospheric pressure was present during the determination of the distribution coefficients. Also, the investigations reported in Ref. 5 allowed the radioactive solution to remain in contact with the rock for only 16 hours; hence, there are no data to determine whether or not the system had reached equilibrium. Since, under actual in situ copper-leaching conditions, it might be expected that the values of K_d would be somewhat different from those noted in Ref. 5, a series of experiments was carried out at LLL to ascertain the K_d values in a chemical environment similar to that expected for in situ leaching of copper sulfide ore. Most of the LLL experiments were carried out on San Manuel sulfide ore and were conducted at about 90°C and under an oxygen pressure of 400 psi. The systems were ionically similar to those expected in chemical copper mining, with a pH of about 1.7-2.0*; equilibration times of four to eight weeks were used. It should be emphasized that the K_d values measured at LLL and used in this study are appropriate only for the assumed chimney conditions (both chemical and physical) and should not be assumed to be generally applicable.

The degree to which strontium is coprecipitated† with the breakdown products of rock minerals was also investi-

*In solutions with pH $\approx$ 1, lower K_d values were observed.

†The term "coprecipitation" is here used to designate a removal process which undoubtedly combines true coprecipitation with sorption.

gated; it might be expected that other radioactive elements would also be removed from solution by coprecipitation to some extent.

Table 10 compares the K_d values calculated from LLL data with those reported in Ref. 5. It can be seen that, with the exception of cesium, the K_d values determined under conditions approximating those expected in the present nuclear chimney concept are somewhat higher than those given in Ref. 5. Parenthetically, it should be noted that oxygen pressure apparently has little effect on the various K_d values which were measured at LLL.

Table 10. Comparison of K_d values determined at Oak Ridge (ORNL) and at Lawrence Livermore Laboratory (LLL).

Nuclide	K_d	
	ORNL (Ref. 5)	LLL
85,89,90Sr	0.7	$\sim 100^a$
91Y	0 ^b	1.5
103,106Ru	0 ^b	50
124,125Sb	50	300
134,137Cs	50	5
141,144Ce	0.5	1
143Pr	—	1
147Pm	—	1
151Sm	—	1
152,155Eu	0 ^b	2
160Tb	—	1
170Tm	—	1

^aIncludes effect of coprecipitation.

^bReported as less than 0.25.

Potential Radiation Exposures to Plant Workers and to Off-Site Populations

Exposure of plant workers and off-site residents to radiation resulting from the nuclear solution mining of copper would generally be by somewhat different processes. The plant worker could be exposed to gamma radiation fields which would surround that processing equipment in which radioactive materials were contained. Also, he could possibly inhale small amounts of radioactive substances, and thus receive internal radiation exposure. In addition, when the worker is inhaling such gases, he would be effectively immersed in them, and thus could be exposed to the radiation being emitted from the cloud of radioactivity in which he is immersed.

Members of the general public could also be exposed to small amounts of gaseous radioactivity emitted from the processing plant while copper refining was taking place; radiation doses would result from the same processes as those discussed for plant workers exposed to airborne radioactivity. While it is extremely unlikely that a nuclear chemical mining and refining operation would be located where the groundwater regime

would permit radioactive solutions to enter drinking or agricultural water supplies, the potential exposure path involving ingestion of radioactivity in drinking water will be examined. Finally it is possible that radioactivity could enter some food chains in the vicinity of the mining/refining operations; however, this method of exposure is so site-specific that it will not be discussed in this paper. As specific cases are evaluated, this potential exposure pathway will be considered.

In evaluating the consequences of exposure to radiation fields or to low concentrations of radioactivity in air or water, the guidelines published by the AEC¹⁸ (which are generally in accordance with those put forth by the EPA and other radiological protection organizations¹⁹) will be used for reference. Briefly, these guidelines for workers and the general public are consistent with those noted in Table 11. The so-called MPC (maximum permissible concentration) values given in Ref. 18 are computed for each radioactive nuclide, and represent that concentration of the particular radioactive substance in question which, if

Table 11. Permissible annual doses to occupational workers and individual members of the public.

Organ or tissue	Occupational worker	Individual member of the public
Total body (gonads and red bone marrow)	5 rem	0.5 rem
Skin, bone, thyroid	30 rem	1.5 rem
Hands, forearms, feet, ankles	75 rem	7.5 rem
All other organs	15 rem	1.5 rem

ingested by a person over a period of one year, will result in a dose which is approximately that shown in Table 11 being delivered to the critical organ of the person at risk. It should once again be emphasized that these MPC values apply to a chronic exposure situation that presumably persists over a period of one year, and not to a short term "pulse" of radioactivity in the environment.

With this background, then, it is possible to examine the various potential exposures to occupational workers and to individual members of the public which could result from nuclear chemical mining and refining of copper ore.

CHIMNEY REENTRY AND CAVITY FLOODING

For this analysis, we will assume that the gas contained in the chimney shortly after the nuclear detonation does not disperse into the fractured rock region surrounding the chimney. As a consequence of this assumption, we consider that this gas will all be released when the chimney is reentered about six months after the detonation. Thus, the concentrations of gaseous radioactivity shown in Column 5 of Table 7 are appropriate for use as the "source term" for radioactivity introduced into the atmosphere. If these concentrations are divided by the appropriate MPC values for members of the public (obtained from Ref. 18 and given in Column 6 of Table 7), then the relative biological effect of each of these radioactive species can be assessed. It can be seen that when a fission explosive is used, ^{85}Kr is the "limiting" radionuclide. Should a thermonuclear explosive be employed, tritium would be the principal isotope of concern.

The "dilution factors" given in Table 7 indicate the number of volumes of air with which the cavity gas would have to be diluted before the nuclide under consideration would be present in a concentration no greater than the MPC.

In practice, this dilution of cavity gas would take place as the gas diffuses into the air from the wellhead release point. Even if below-normal atmospheric diffusivity is assumed, the factor-of-3000 dilution required for ^{85}Kr should be attained before the gas moves one kilometer away from the wellhead.^{20,21}

However, there are several factors which render the factor-of-3000 dilution requirement unrealistically conservative. It has been assumed that none of the "chimney gas" has been dispersed into the fractured rock surrounding the chimney. Such dispersal could effect as much as a tenfold reduction in the amount of gas (and radioactivity) which would actually be released to the atmosphere upon chimney reentry. A second conservative aspect implicit in this approach lies in the calculation of the potential exposure to a member of the public living in the vicinity of the wellhead. The calculation assumes continuous exposure to the radioactive gas; however, normal shifts in wind direction would certainly indicate intermittent rather than continuous exposure. Finally, MPC values are predicated on continuous exposure of a member of the public over long periods of time. Since the chimney "bleed down" should actually require only a few days, the radiation exposure is transient in nature, and the postulated dilution requirements are unnecessarily stringent.

In summary, it can be concluded that the time-integrated concentrations of radioactivity resulting from chimney gas releases will result in potential exposures to members of the public that are well below the applicable guidelines.

No analysis of exposure for occupational workers has been made here, since this procedure (cavity reentry and flooding) can be planned so that no worker need be exposed to the effluent gases.

EMISSIONS DURING RELEASE OF OXYGEN FROM THE NUCLEAR CHIMNEY AND FROM PLANT OPERATION

As noted earlier, copper dissolution by this chemical mining process involves the introduction of oxygen into the water-filled nuclear chimney. Oxygen in excess of the stoichiometric amount needed for the copper and iron oxidation reactions must be supplied to the system; the excess oxygen will accumulate at the top of the chimney and can either be recycled or allowed to escape to the atmosphere. If it is assumed that the latter alternative is chosen, the radioactive species contained in this "off-gas" must be considered from the standpoint of potential population exposure downwind from the effluent release.

Nuclides which could be present in the released gas include only those which would persist after chimney flooding, and which would be volatile under the oxidizing conditions prevailing in the chimney. Certainly, tritiated water would be present in the chimney, and presumably would appear as water vapor in the oxygen releases. With regard to those elemental species which have volatile oxides, per-

haps the best known is ruthenium, with the compound RuO_4 displaying a boiling point of approximately 40°C . The simple approach to the effects of possible volatility of ruthenium or its compounds which is presented in this paper is only intended to serve as an example; laboratory experiments are required to verify this treatment. Also, other elements such as antimony, tellurium and cesium display tendencies toward volatility, depending upon the chemical and physical conditions to which they are exposed. Consequently, the behavior of these and other elements should be carefully examined in connection with a thorough safety evaluation of nuclear chemical copper mining.

Considering first the tritiated water, if it is assumed that the oxygen off-gas rapidly equilibrates to an ambient temperature of 30°C , this saturated gas will contain 3.4×10^{-5} g of water per milliliter of gas. Since the concentration of tritium in the chimney is 6.9×10^{-3} $\mu\text{Ci/ml}$ (see Table 8), the concentration of tritium (as HTO) in the off-gas stream will be about 2.3×10^{-7} $\mu\text{Ci/ml}$. If this value is compared with the Federal Standards for tritium concentrations in gas (2×10^{-7} $\mu\text{Ci/ml}$), it will be seen that this gas need be only slightly diluted in order to meet this standard. Hence, such a release does not appear to compromise the safety of members of the public located downwind from the off-gas release point.

In analyzing the concentrations of ^{103}Ru and ^{106}Ru which might be expected to be in the off-gas stream, the volatility of RuO_4 under the conditions prevailing in the chimney must be evaluated. A preliminary calculation of the equilibrium constant

$$K_{eq} = \frac{P_{RuO_4}}{P_{O_2}} \sim 2.6 \times 10^{-15}$$

has been made,²² assuming a cavity temperature of 100°C and that the RuO₄ gas is in equilibrium with RuO₂ solid. Effects that would lower the chemical activity of the ruthenium, such as reactions to form stable mixed oxides and possible formation of an unsaturated solution in the leach solution, were not considered. If it is conservatively assumed that only the ruthenium* produced as fission products is available for exchange with the radio-rutheniums, the specific activities of the two nuclides of interest are:

$$^{103}\text{Ru} - 3.5 \times 10^{10} \text{ } \mu\text{Ci/mole of RuO}_4$$

$$^{106}\text{Ru} - 1.3 \times 10^{10} \text{ } \mu\text{Ci/mole of RuO}_4.$$

Then, the concentrations of these species in the oxygen off-gas are:

$$^{103}\text{Ru} - 9.1 \times 10^{-5} \text{ } \mu\text{Ci/mole of O}_2 \\ \text{or } 3.7 \times 10^{-9} \text{ } \mu\text{Ci/ml of O}_2$$

$$^{106}\text{Ru} - 3.3 \times 10^{-5} \text{ } \mu\text{Ci/mole of O}_2 \\ \text{or } 1.4 \times 10^{-9} \text{ } \mu\text{Ci/ml of O}_2.$$

Here, it is assumed that the oxygen gas is at 30°C and 1 atm pressure. These concentrations may be compared with the standards for members of the public.

* Approximately 3 moles, or about 300 g.

	Calculated concentration in off-gas, $\mu\text{Ci/ml}$	(MPC), ¹⁸ $\mu\text{Ci/ml}$
¹⁰³ Ru	3.7×10^{-9}	2×10^{-8}
¹⁰⁶ Ru	1.4×10^{-9}	3×10^{-9}

Thus, it can be seen that no dilution of the oxygen off-gas stream is required to meet the established standards.

Should a thermonuclear explosive be used, tritium concentrations would be a factor of 1000 higher, and considerable dilutions would be required to meet the effluent standards listed above. On the other hand, ruthenium concentrations would be lower by a factor of 20.

With regard to the exposure of occupational workers to airborne radioactive substances, a similar analysis is appropriate. The HTO concentrations in air over leach solutions in the extraction plant would be the same as previously noted, or about $2.3 \times 10^{-7} \text{ } \mu\text{Ci/ml}$. Since allowable radiation exposures for occupational workers are a factor of ten higher than for members of the public, the allowable concentration of HTO in air for such workers is $5 \times 10^{-6} \text{ } \mu\text{Ci/ml}$; hence, minimal precautionary measures would be required. However, good plant design practice would require adequate ventilation to avoid problems with the buildup of non-radioactive chemical contaminants; thus, actual HTO concentrations to which workers would be exposed would be small.

Since the temperatures of open solutions in the processing plant would be well below 100°C, and the concentration of oxygen gas would be low, the concentrations of radiorutheniums in the air above the leach solution would be even smaller

than those calculated earlier and should be well below the guideline values for occupational workers.

Again, should a thermonuclear explosive be employed, tritium concentrations 1000 times higher than those previously indicated could result; in this case, ventilation design for the leach plant would have to be carefully evaluated.

RADIOACTIVITY PRESENT IN LEACH SOLUTIONS

Since it seems highly unlikely that an occupational worker will ingest any of the radioactivity present in these solutions (other than by inhalation, which can be greatly mitigated by good industrial safety design),^{*} the only method of exposure would be from the gamma-ray fields surrounding the processing equipment which result from the radionuclide content of the solutions contained in such equipment. Since processing of leach liquor will probably not begin earlier than one year after the construction of the nuclear chimney, maximum potential worker exposures will be calculated for that time after the detonation. If it is assumed that solutions contain radioactive isotopes consistent with the concentrations shown in Tables 8 and 9 (but corrected for further radioactive decay), then it can be calculated that a worker standing three feet from a tank containing such a solution would receive about 4 mrem/hr. As shown in Table 12, approximately 60% of this exposure would be due to the radiation from ¹³⁷Cs-Ba. Thus, two years following the detonation, the radiation field at this

point would have decreased only to about 3 mrem/hr.* It should be emphasized that this calculation does not assume any shielding by the materials from which such a tank would be constructed. Therefore, with appropriate plant design, incorporating either equipment shielding or process design obviating the need for close approach of the operator to the equipment, radiation exposure to plant personnel could be kept well within the appropriate safety standards.

Use of a thermonuclear explosive would, of course, decrease the radiation field surrounding the processing plant equipment by a factor of about 20, but increase potential HTO exposures by a factor of 1000. Thus, while less attention might be required with regard to equipment shielding, ventilation of working areas would have to be carefully evaluated.

This radiological safety analysis has been made only for an aqueous leach solution with the same composition as that prevailing in the nuclear chimney. Studies reported in Ref. 5 indicate that the amount of radioactivity following the copper purification process through the extraction and electrowinning steps is small; hence, radiation fields existing around tanks containing these solutions would be relatively smaller than those indicated in Table 12: certainly less than 1 mrem/hr. As noted earlier, this estimate is based on no shielding by the tank wall. A more realistic evaluation, taking such shielding into account, would reduce the calculated

* The solutions are chemically hazardous, due to the presence of large amounts of acid and dissolved copper.

* It should be noted for comparison that radiation levels in uranium ore milling and processing plants can be on the order of 100 mrem/hr in some locations.

Table 12. Estimated external dose rate from nuclides in solution (assumes 100% of chimney nuclides in solution and 1% of melt nuclides in solution).

Nuclide	Half-life	γ -MeV/sec-ml	
		At +1 yr	At +2 yr
⁴⁶ Sc	84 da	0.8	0.04
⁵⁴ Mn	303 da	0.1	0.06
⁵⁹ Fe	45 da	0.2	—
⁶⁰ Co	5.3 yr	0.4	0.3
⁶⁵ Zn	245 da	0.4	0.2
⁹¹ Y	59 da	3.5	0.06
⁹⁵ Zr	65 da	0.4	0.01
⁹⁵ Nb	35 da	0.7	0.01
¹⁰³ Ru-Rh	40 da	1.8	0.003
¹⁰⁶ Ru-Rh	1.0 yr	4.1	2.1
^{115m} Cd	43 da	0.03	—
¹²⁵ Sb	2.7 yr	0.4	0.3
^{125m} Te	58 da	2.3	0.03
^{127m} Te	109 da	25	2.5
^{129m} Te	34 da	0.8	—
¹³⁴ Cs	2 yr	15	11
¹³⁷ Cs-Ba	30 yr	84	82
¹⁴¹ Ce	33 da	9.004	—
¹⁴⁴ Ce-Pr	284 da	0.3	0.1
¹⁵² Eu	12 yr	0.1	0.1
¹⁵⁵ Eu	1.8 yr	0.03	0.03
¹⁶⁰ Tb	72 da	0.05	0.003
¹⁹² Ir	130 da	0.4	0.05
Σ		141 ^a	99 ^a

^aThe approximate dose 3 ft from a tank of infinite volume is 4 mrem/hr at 1 yr and 3 mrem/hr at 2 yr. This dose rate is assumed to be approximately one-half of that which would be received by an individual completely immersed in a solution having the radiological composition indicated in the table.

radiation field by a factor of between 2 and 10.

The only mechanism whereby members of the population could be exposed to radioactivity from the chemical mining leach solution would be by the introduction of a portion of this solution into some

aquifer in the vicinity of the nuclear chimney. However, location of a nuclear chemical copper mining/refining operation in an area where contaminants could eventually enter drinking or irrigation water supplies would in all likelihood never be either proposed or approved.

For the sake of completeness, the consequences of introducing radioactivity into groundwater in the vicinity of the nuclear chimney will be explored.

In order to evaluate the "critical" radionuclides which would be the largest contributors to any such potential exposure of members of the population, it must be recalled that, in order to engage in a successful chemical mining enterprise, dilution or removal of the leach solution from the nuclear chimney must be prevented over the period of time during which leaching is to take place. Current estimates indicate that this period of time should be about five years. During this period of time, if any leakage path exists it must be controlled by pumping in nearby control wells to eliminate groundwater movement in the vicinity of the chimney.

Examining Tables 8 and 9, it can be seen that radioactive decay alone would reduce the concentrations of most species in solution to below the guideline levels given in Ref. 18; the only nuclides which remain in significant amounts are ^3H , ^{90}Sr , ^{106}Ru , ^{125}Sb , ^{134}Cs , and ^{137}Cs . Of these species, only ^3H and ^{90}Sr have small values of K_d in groundwater,* and hence would travel at greater than 1% of the groundwater flow rate. From the results shown in Table A-3 in the Appendix, it can be seen that the "limiting" radionuclide is ^{90}Sr , which would not decay to the guideline level for about 250 years. Adopting the approach used in Ref. 23 to ascertain the potential effect

of dissolved radioactivity on groundwater quality, a calculation can be made of the distance over which the water would have to travel before the "limiting" radionuclide had decayed to an acceptable level (according to current radiation protection guides). For the purposes of this illustrative example, we will assume that the average groundwater flow rate is 50 ft/yr.* On the basis of the data presented in Table A-3 it can be seen that ^{90}Sr (the "limiting" nuclide) moves at a velocity only about 8% of that of the groundwater flow, or less than 4 ft/yr in this instance. Thus, the water would move less than 1000 ft before reaching the concentration specified by the Radiation Protection Guide (RPG) for ^{90}Sr .

Thus, while radioactivity in groundwater does not appear to compromise the inherent radiological safety of nuclear chemical copper mining, it does represent an aspect of the technique which must be carefully evaluated for each proposed experiment or application.†

Should a thermonuclear explosive be employed, the critical radionuclides now would be ^3H , ^{90}Sr , and ^{137}Cs . While ^{90}Sr would decay to guideline level in about 130 years, and ^{137}Cs would require about 100 years, tritium (being 1000 times more abundant than in the case of a fission explosive) would require about 140

* Note that the K_d values appropriate for groundwater are listed in Table A-3 in the Appendix; these are significantly different from those which should be used for chimney conditions (Tables 8 and 9).

† As previously noted, some nuclear explosives contain ^{239}Pu . Should such an explosive be used in conjunction with nuclear chemical copper mining, the behavior of this nuclide should also be considered (see Ref. 6).

* This estimate is taken only as an example. An actual determination of this velocity would be made at any proposed experimental site (see following section).

years to decay to guideline level. HTO is known to travel at essentially the same speed as the groundwater itself, while ^{90}Sr and ^{137}Cs do have appreciable K_d values. Thus tritium becomes the limiting radioactive species in the case of a thermonuclear explosive.

RADIOACTIVITY IN THE PRODUCT COPPER

Analysis of the product copper subsequent to an extraction-electrowinning refining process indicated the presence of a small portion of the ^{106}Ru which had been introduced initially into the simulated leach liquor (Ref. 5).^{*} If it is assumed that the amount of ^{106}Ru present in the product copper is proportional to the concentration of this radionuclide in the leach liquor, then the concentration of ^{106}Ru in copper recovered from a radioactive solution similar in composition to that presented in Tables 8 and 9 would be about $2 \times 10^{-6} \mu\text{Ci}$ per gram of metal. Of course, this represents an

extreme-upper-limit estimate, inasmuch as optimization of the copper refining process should prevent the incorporation of ^{106}Ru in the product copper. Also, the coprecipitation of ^{106}Ru with the breakdown products of rock minerals could significantly reduce the concentration of this radionuclide in the leach liquor below that noted in Table 8. However, even in the unlikely case that copper containing $2 \times 10^{-6} \mu\text{Ci/g}$ were the end product of the copper refining process, the exposure rate (from β particles to bare skin) to an individual would be about 0.002 mrem/hr. Perhaps more significantly, if such copper were assumed to be in contact with drinking water supplies and were dissolved to the extent allowed by pure water standards (1 ppm of copper in the water), the concentration of ^{106}Ru in the water would be $2 \times 10^{-12} \mu\text{Ci/ml}$. An adult drinking this water over a period of one year would receive a total radiation dose of about 0.0003 mrem. This represents about two ten-millionths of the RPG for a member of the public.

Laboratory and Field Data Required to Better Evaluate Radiological Safety of Chemical Mining

In formulating the preceding section, a number of conservative ("worst case") assumptions were used in calculating the radiological consequences of chemical copper mining and refining. Even this "worst case" radiological safety analysis

^{*} Note that entrainment of the ^{106}Ru with copper during the refining process is probably the reason for the presence of small amounts of this radionuclides in the product copper.

indicated that the total annual exposure of both plant workers and nearby residents would not be greater than 0.002 man-rem per ton of copper produced (see Conclusion). Thus, additional data are not required to prove the inherent safety of this concept. However, more data are required for optimum recovery plant design, and it would be desirable to employ more realistic assumptions in future

assessments of the safety of the entire process. The programs necessary to collect these data are briefly discussed in this section.

LABORATORY MEASUREMENTS NEEDED OR IN PROGRESS

Radionuclides Remaining in the Leach Liquor

More information is needed on this topic for two purposes. First, and most important, plant design will be determined to some extent by the amount of radioactivity in the solutions being processed. Therefore, the behavior of certain key nuclides (such as ^{103}Ru and ^{106}Ru) should be studied further. Secondly, in order to satisfy environmental concerns, the amount and behavior of a few long-lived nuclides (such as ^3H and ^{90}Sr) should be better characterized. In particular, it would be desirable to investigate radionuclide solubility and removal.

Radionuclide Solubilities

Although work has been carried out both at ORNL⁵ and LLL to determine the solubilities of a number of radionuclides—both from "puddle glass" and chimney rubble samples—the physical characteristics of the rubble and melt samples were undoubtedly different from the rubble and melt which would be encountered in a nuclear chimney. For example, the melt samples used in the LLL experiments were powdered and hence more amenable to leaching than would be the case with in situ puddle glass. With regard to the solubility of radionuclides contained in melt, preliminary measurements indicate that the fractional amounts of these isotopes which are dissolved are about the same

as the fraction of the silica glass which is dissolved. Studies are now in progress at LLL to further investigate these topics.

Radionuclide Removal From Solution

As noted earlier in this paper, radionuclides can be removed from the leach liquor in the chimney by two principal mechanisms: (1) sorption on rock surfaces in the chimney, and (2) coprecipitation with the breakdown products of rock minerals. Both of these processes must be investigated more thoroughly. With regard to the first process, although distribution coefficients have been measured for a number of nuclides of interest under conditions believed to be somewhat typical of those existing in a nuclear chimney, it should be emphasized that the experiments were conducted using "rubble" sizes which might not be representative of those present in a chimney. Also, it would be desirable to have data over a range of temperatures and solution compositions in order that the optimum conditions for radionuclide removal can be determined. In addition, only in the case of strontium have measurements been made in which coprecipitation of radionuclides with the expected rock mineral breakdown products takes place. Work in these areas of interest should be continued.

Distribution coefficient measurements made under conditions simulating those in an aquifer at a distance from a nuclear chimney are needed in order to study the possible consequences of groundwater transport. Some of these measurements have been made at LLL.

Volatility of Radionuclides Under Leaching Conditions

While even the "worst case" safety analysis presented in this paper indicated that volatile radioactivities would not represent a hazard either to workers or to the public, some minimal investigative program in this area would be desirable to further allay possible environmental concerns. Since the volatilities of radioactive elements (or compounds of these elements) are not well known under the conditions existing during the chemical mining of copper, a modest calculational program is planned to be carried out at LLL in order to identify any species which would warrant further investigation. Additional laboratory experiments should be carried out as needed to determine the behavior of any such species.

Behavior of Radionuclides During Processing

For this copper leach radiological analysis, it has been tacitly assumed that conditions in the processing plant will not contribute to local build-up of radioactivity during the copper purification. However, although plant design is being planned to minimize such build-up, the actual behavior of the radioactive solutions must be observed under realistic plant conditions of temperature, acidity, etc. This is necessary in order that process and equipment design may be optimized with respect to copper recovery and radiological safety. Such work is in the preliminary planning stages at LLL.

Product Contamination

Optimal copper refining conditions are required to insure both the chemical

and radiological purity of the product copper. Although, as noted in the preceding section, the presence of minimal quantities of radioactivity in copper does not pose a safety problem, it is still possible that public acceptance, as well as certain industrial uses of the metal, could be jeopardized. Thus, the normal chemical engineering approach to recovery plant design should give consideration to refining processes which will assure that there will be no radiological problems associated with the product copper.

FIELD DATA REQUIRED FOR PROJECT EVALUATION

In analyzing the potential radiological impact of any chemical mining experiment, especially upon the public, detailed information is required on any proposed site.

Groundwater Data^{*}

Locations and characteristics of all aquifers, and their positions relative to the proposed nuclear chimney must be determined. Data relating to the water quality and movement in these aquifers would also be required.

^{*}As noted elsewhere, the potential problem of radioactivity in groundwater is probably more apparent than real due to the inherent characteristics of copper deposits. Such deposits are usually found in relatively impermeable rock, where groundwater flow rates are relatively slow. Also, the water which passed through such a deposit would probably be unfit for consumption because of its mineral content (copper, sulfate, etc.). Thus, the contamination of potable water supplies by radioactivity from a nuclear explosive detonated in a copper deposit is unlikely.

Laboratory experiments measuring K_d values for critical radionuclides will be made under the appropriate chemical and physical conditions in order that the rate of movement of these radionuclides in aquifers containing potable water can be estimated. Such data are necessary for the preparation of required environmental impact statements.

Atmospheric Data

A data-gathering program would be necessary to document the wind patterns and atmospheric diffusivity characteristic of the proposed experimental site. The information collected would then enable a realistic assessment to be made of the potential exposures of nearby populations to airborne radioactive substances.

Conclusions

A large amount of data has been obtained pertaining to the radiological safety aspects of copper production using nuclear chemical mining and refining techniques.^{2,3} The existing information allows us to make a conservative ("worst case") safety assessment of the process. This assessment of potential radiation exposures to both occupational workers and members of the public as a result of nuclear chemical copper mining indicates that such exposures are well within existing radiation protection guides. These results are summarized in Table 13.

A more realistic radiological safety evaluation would, in all likelihood, result only in the prediction of still lower radiation exposures to workers and to members of the public. However, much of the data required for the preparation of such a realistic evaluation is also required for optimal recovery plant design. Hence, it is recommended that the appropriate calculational and/or experimental programs required to obtain these data be carried out in order that plant design questions may be speedily addressed and a more realistic radiological safety analysis may be prepared.

Table 13. Summary of the preliminary radiological analysis.

Potential source of exposure	Time during which exposure occurs	Principal population exposed	Exposure path	Estimated ^a exposure
1. Bleed-off of chimney gases at reentry and/or during chimney flooding.	Starting six months after detonation; duration is a few weeks at most.	Residents of the area downwind from the project. (Workers would receive negligible exposure.)	Inhalation of gaseous radioactivity.	0.000013
2. Off-gassing of excess oxygen during dissolution of ore.	Starting about one year after detonation and lasting for five years. ^b	Residents of the area downwind from the project. (Workers would receive negligible exposure.)	Inhalation of gaseous radioactivity.	0.0000001
3. Transport of soluble radionuclides out of the chimney area and into groundwater. ^c	Exposure could start many years after detonation.	Residents of the area using contaminated groundwater for drinking or irrigation.	Ingestion of soluble radionuclides.	<0.00000000000005
4. Radionuclides present in the product copper.	Exposure could start when copper reaches the market (about 1-1/2 years after detonation). Effective duration is no more than 1-1/2 years.	Population using copper products.	Ingestion of soluble radionuclides.	0.00006
5. In-plant radiation fields.	Starting about one year after detonation and lasting for five years.	Plant personnel.	External gases exposure; inhalation of tritiated water vapor, etc.	0.002

^aExpressed in man-rcms per year per ton of product copper. See Appendix for calculational details and supplementary information on radiation exposure.

^bFive years is the time interval during which the copper leaching/recovery operation is assumed to take place.

^cNote that of the listed sources of exposure to radiation, this is the one which is most amenable to prevention. Location of a nuclear chemical copper mining/refining operation in an area where contamination could enter drinking or irrigation water supplies would in all likelihood never be either proposed or approved.

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Appendix A

Evaluation of the Relative Importance of the Various Modes of Exposure to Radiation

The following mechanisms leading to possible radiation exposure as a result of nuclear chemical copper mining and refining were considered:

1. Bleed-off of chimney gases at reentry and/or during chimney flooding (see Table A-1).
2. Off-gassing of excess oxygen during dissolution of ore (see Table A-2).
3. Transport of soluble radionuclides out of the chimney area and into groundwater (see Table A-3).
4. Radionuclides present in the product copper.
5. In-plant radiation fields, inhalation and ingestion.

In the first four items, the primary concern would be with regard to potential exposure of members of the public; the fifth mechanism would result only in the possible exposure of plant personnel.

In estimating the relative importance of the five exposure paths noted above, it will be necessary to make a number of assumptions. These assumptions are not necessarily intended to represent conditions which would prevail at any given project site, but can be useful in obtaining some indication as to which of the five areas of interest warrants the most attention.

The assumptions made in evaluating the first three paths are:

- A town with a population of 10,000 is located 15 miles from the project site.

- The wind blows from the project site toward the town 50% of the time.
- Average atmospheric dispersion (between Pasquill Categories C and D) prevails in this area.
- The town water supply is obtained 5 miles from the project site, in the same aquifer and downgradient from the chimney.*
- No dilution of the leach liquor occurs as it moves between the chimney and the water intake for the town water supply.
- Exposure per ton of copper produced is based on a total copper production of 10^4 metric tons.

1. Chimney-Gas Bleed-off

Dose calculation results for chimney-gas bleed-off are shown in Table A-1. Since the town population is 10,000, the total population exposure would be 1.3×10^{-5} man-rem per ton of copper, delivered over a one-year period.

2. Oxygen Off-gassing During Ore Dissolution

Table A-2 gives the calculated dose resulting from oxygen off-gassing during ore dissolution. During dissolution of the copper from the chimney rubble, 13.06 tons of oxygen are required per ton

* As has been noted in the body of this paper, this assumption is very unrealistic; it is extremely unlikely that such a site for a nuclear chemical copper mining operation would be either proposed or approved.

of copper dissolved.²⁴ If it is assumed that twice as much oxygen is supplied to the chimney as is actually utilized in the copper dissolution, some 13.06×10^4 tons

of oxygen will be released in off-gassing over a five-year period. This is equal to about 6.1×10^5 ml of oxygen gas released per second.

Table A-1. Calculation for chimney gas bleed-off.

Nuclide	Total amount ^a released (Ci)	Release rate, ^b Q (Ci/sec)	Average ^c concentration at town, χ (μ Ci/ml)	Inhalation dose conversion constant ($\frac{R/yr}{\mu Ci/ml}$)	Total ^b dose (R)	Total dose per metric ton of Cu (R)
³ H	2	6.3×10^{-8}	6.3×10^{-15}	2.5×10^6	1.6×10^{-8}	1.6×10^{-12}
¹⁴ C	7	2.2×10^{-7}	2.2×10^{-14}	5×10^6	1.1×10^{-7}	1.1×10^{-11}
³⁷ Ar	7200	2.3×10^{-4}	2.3×10^{-11}	3×10^4	6.9×10^{-7}	6.9×10^{-11}
⁸⁵ Kr	2300	7.3×10^{-5}	7.3×10^{-12}	1.7×10^6	1.2×10^{-5}	1.2×10^{-9}
Total						1.3×10^{-9}

^aBased on sources considered (see Table 7).

^bAssuming that the release takes place over a period of one year. Although the actual duration of such a release could be much less than one year, the total calculated exposure is relatively insensitive to the rate of release, being determined principally by the total amount released.

^c $u \frac{\chi}{Q} = 4 \times 10^{-7}$ (see Refs. 18 and 19), where u is the wind speed in m/sec (assumed to be 2 m/sec in this case), χ is the concentration of a radioactive species in air after dispersion (at a distance from the source) in Ci/m³, and Q is the rate of release of the source radioactivity in Ci/sec. Thus, in the present instance, $\chi = 2 \times 10^{-7} Q$. If it is assumed that the radioactive plume passes over the town only 50% of the time, then $\chi = 1 \times 10^{-7} Q$.

Table A-2. Calculation for oxygen off-gassing.

Nuclide	Concentration in off-gas (μ Ci/ml)	Release rate (Ci/sec)	Average concentration at town (μ Ci/ml)	Inhalation dose conversion constant ($\frac{R/yr}{\mu Ci/ml}$)	Total ^b dose (R)	Total dose per metric ton of Cu (R)
³ H	2.3×10^{-7}	1.4×10^{-7}	1.4×10^{-14}	2.5×10^6	1.8×10^{-7}	1.8×10^{-11}
¹⁰³ Ru	3.7×10^{-9}	2.3×10^{-9}	2.3×10^{-16}	7.5×10^7	8.6×10^{-8}	8.6×10^{-12}
¹⁰⁶ Ru	1.4×10^{-9}	8.6×10^{-10}	8.6×10^{-17}	5×10^8	2.2×10^{-7}	2.2×10^{-11}
Total						4.9×10^{-11}

^aSee footnote (c) of Table A-1.

^bOver 5 years of exposure.

Since the town population is 10,000, the total population exposure would be 4.9×10^{-7} man-rem per ton of copper, delivered over a five-year period. Total population exposure rate would be about 1×10^{-7} man-rem per year per ton of copper.

3. Transport of Soluble Radionuclides in Groundwater

Calculational results for transport of soluble radionuclides in groundwater are given in Table A-3. A general treatment of radionuclide movement in groundwater is presented in Ref. 23.

As noted earlier, it has been assumed that the groundwater velocity is 50 ft/year. Since the radioactive species are assumed to remain in the immediate vicinity of the chimney during the leaching of the ore, they will only start to be carried by the

groundwater after the chimney leaching phase of the project has been completed, or about 6 years after the detonation.

Since the town population is 10,000, the total population exposure would be $<1.4 \times 10^{-12}$ man-rem per ton of copper, delivered over a 30-year period. Annual dose would thus be $<5 \times 10^{-14}$ man-rem per year per ton of copper. However, it should be noted that, because of the "hold up" time of the radioactivity in the ground water, this dose would be delivered long after the actual copper recovery operation took place.

4. Radionuclides in the Product Copper

The fourth exposure pathway considered represents potential dose from radionuclides present in the product copper. If the $\sim 10^4$ tons of copper recovered from the project under consideration were all

Table A-3. Results for groundwater transport.

Nuclide ^a	Chimney concentration ^b ($\mu\text{Ci/ml}$)	K_d ^c	$\left(\frac{1}{1 + rK_d}\right)^d$	Time required to reach town water intake (years)	Concentration at time of entry into town water system ^e ($\mu\text{Ci/ml}$)	Dose conversion constant (for drinking water) ($\frac{\text{R/yr}}{\mu\text{Ci/ml}}$)	Total dose ^f (R)	Total dose per metric ton of Cu (R)
³ H	$< 6.9 \times 10^{-3}$	0	1	528	$< 2.8 \times 10^{-16}$	170	$< 1.4 \times 10^{-12}$	$< 1.4 \times 10^{-16}$
⁵⁵ Fe	1.7×10^{-3}	0 ^g	1	528	2.5×10^{-65}	1.9×10^3	1.4×10^{-60}	1.4×10^{-64}
⁶⁰ Co	4.1×10^{-6}	0 ^g	1	528	1.9×10^{-36}	3×10^4	1.7×10^{-30}	1.7×10^{-34}
⁹⁰ Sr	1.6×10^{-4}	4 ^h	0.08	5860	2.2×10^{-78}	3×10^7	6.6×10^{-71}	6.6×10^{-75}
¹²⁵ Sb	2.3×10^{-5}	30 ^h	< 0.01	48000	$< 10^{-100}$	1.5×10^4	—	—
¹³⁴ Cs	3.1×10^{-4}	400 ^h	0.0008	630000	$< 10^{-100}$	5.6×10^4	—	—
¹³⁷ Cs	3.7×10^{-3}	400 ^h	0.0008	630000	$< 10^{-100}$	2.5×10^4	—	—
¹⁴⁷ Pm	6.0×10^{-4}	1000 ^h	< 0.0003	1600000	$< 10^{-100}$	7.5×10^3	—	—
¹⁵¹ Sm	4.3×10^{-6}	> 1000 ^h	< 0.0003	1600000	$< 10^{-100}$	3.8×10^3	—	—
¹⁵² Eu	2.9×10^{-6}	> 1000 ^h	< 0.0003	1600000	$< 10^{-100}$	1.9×10^4	—	—
Total								$< 1.4 \times 10^{-16}$

^aOnly those nuclides having a half-life greater than 2 years have been considered.

^bSee Tables 8 and 9 for concentrations 180 days after detonation.

^cValues appropriate for groundwater. Note that these K_d values are significantly different from those used for the chimney solution, because of the different chemical compositions of the two media.

^dThe fraction of a given species remaining in solution in a unit volume of solution, where r is the ratio of the effective mass of rock upon which the given species is adsorbed to the volume of water in contact with the rock ($r \approx 3$).

^eCalculated for a time 6 years longer after the detonation than noted in the preceding column — to allow for the time interval during which the radioactivity is localized in the chimney region.

^fAssuming a 30 year lifetime dose.

^gAssumed.

^hMeasured at L.L.L.

used in the fabrication of water pipes, and it is assumed that 10 lb of copper were used per person in the installation of such pipes, then a total of 2×10^6 persons could be exposed to ruthenium radioactivity, at an initial rate of 0.0003 mrem per year. To determine the total population exposure commitment, this initial rate should be multiplied by the average life of ^{106}Ru , which is about 1.4 years. Thus, the total population dose from this source would be

$$\frac{(2 \times 10^6)(3 \times 10^{-7})(1.4)}{10^4} = 8.4 \times 10^{-5} \text{ man-rem per ton of copper.}$$

The maximum annual dose rate would be about 6×10^{-5} man-rem per ton of copper. Once again, it should be emphasized that actual radiation exposure from the product copper can be essentially eliminated since pure product copper can be produced by means of an appropriately modified refining process.

5. In-plant Radiation Fields

The final source of exposure to radioactivity which could occur from nuclear chemical copper mining and refining is that resulting from in-plant radiation fields and radionuclides in air or drinking

Table A-4. Comparison of the relative importance of various radiation exposure paths.

Potential source of exposure	Time during which exposure occurs	Principal population exposed	Exposure path	Estimated radiation exposure			
				Individual		Group exposure	
				Exposure (R/yr)	Applicable RPG (R/yr)	Total (man-rem/yr)	Exposure per ton of Cu (man-rem/yr)
(1) Bleed-off of chimney gases at reentry and/or during chimney flooding.	Starting six months after detonation; duration is a few weeks at most.	Residents of the area downwind from the project (Workers would receive negligible exposure.)	Inhalation of gaseous radioactivity.	0.000013	0.5	0.13	0.000013
(2) Off-gassing of excess oxygen during dissolution of ore.	Starting about one year after detonation and lasting for five years.	Residents of the area downwind from the project (Workers would receive negligible exposure.)	Inhalation of gaseous radioactivity.	0.0000001	0.5	0.001	0.0000001
(3) Transport of soluble radionuclides out of the chimney area and into groundwater.	Exposure could start many years after detonation.	Residents of the area using contaminated groundwater for drinking or irrigation.	Ingestion of soluble radionuclides.	<0.000000000000003	0.5	<0.0000000005	<0.000000000000003
(4) Radionuclides present in the product copper.	Exposure could start when copper reaches the market (about 1-1/2 yrs after detonation). Effective duration is no more than 1-1/2 years.	Population using copper products.	Ingestion of soluble radionuclides.	0.0000003	1.5	0.5	0.00005
(5) In-plant radiation fields	Starting about one year after detonation and lasting for five years.	Plant personnel.	External gamma exposure; inhalation of tritiated water vapor etc.	2	5	20	0.002

^a Five years is the time interval during which the copper leaching/recovery operation is assumed to take place.

water. Since such fields and/or concentrations of radioactivity in air or drinking water can be controlled by many different factors, including equipment design, process modification, and even such approaches as personnel training, it is clear that any estimate of exposures from radiation fields must be arbitrary in nature. If it is assumed that workers are exposed to 1 mrem per hour throughout the working year, and that 10 workers are employed in the extraction plant, then the

total annual exposure over the five years during which copper was recovered would be

$$\frac{(0.001 \text{ mrem/hr})(2000 \text{ hr/yr})(10 \text{ men})}{10^4 \text{ tons of copper}} = 0.002 \text{ man-rem per year per ton of copper.}$$

Summary

On the basis of the foregoing estimates, a comparison of the relative importance of the various assumed radiation exposure paths is made in Table A-4.