THE FATE OF URANIUM PROGENY IN URANIUM MINING AND MILLING OPERATIONS

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INTRODUCTION

Domestic uranium production has increased from a few hundred tons of U_3O_8 extracted from thousands of tons of raw ore in the late 1940's to approximately $2x10^4$ tons of U_3O_8 from $1.4x10^7$ tons of ore in 1978. Production levels have increased about 15% annually during the past two years and have surpassed the peak years of the early 1960's. However, because of lower grade feedstock, production rates have now doubled those experienced during the peak years. Increasing demands coupled with decreasing ore grades will continue the production rate trend (see Fig. 1). This creates a concomitant increase in mining and milling operations and in the quantity of radioactive tailings which must be properly sequestered. Proper management of the uranium recovery and waste disposal operations will require an understanding of the behavior and fate of progeny radioactivity.

URANIUM SOURCES AND RECOVERY METHODS

Currently uranium is recovered from approximately 400 sources; over 75% are underground mines, approximately 20% are open pit mines and the remaining sources include a few low yield-heap leach, byproduct recovery and solution mining operations. Whereas U_3O_8 production is currently split between underground and open pit mines, environmental and economic incentives may encourage expansion of uranium byproduct recovery efforts and in-situ solution mining in the future.

Raw ore or leach liquors are currently processed at the 35 operating mills shown in Fig. 2. Conventional mills account for 90% of the U_3O_8 capacity. The remaining capacity comes from 8

in-situ solution mining operations, 3 phosphate acid by-product recovery plants, and four heap leaching operations which extract uranium from dumps, used tailings or copper tailings. Not shown on the figure is a recently completed by-product recovery plant near Tucson which extracts uranium ($\sim$ 10 ppm) from copper leach liquors.

The preponderance of existing and planned uranium recovery operations are found in the western United States. About 50% of current production is in New Mexico, 25% in Wyoming and the remainder spread throughout the other states with operating mills. Many new mines and mills will be needed to satisfy the fuel requirements of nuclear generating projects now under construction. Nearly all the new facilities are expected to be located in the western United States with over 60% in Wyoming and New Mexico. A delineation of potential resources is provided in Table I.

The current annual production of about $2x10^4$ tons of U_3O_8 is equivalent to about $1x10^4$ Ci of uranium and a total of about $8x10^4$ Ci of activity from the uranium and progeny. It is estimated that by the turn of the century the active mines will contribute about $1x10^7$ Ci annually and this will be about the same release rate as from uncovered tailings piles and active mills. Thus, both mines and mills have the potential for releasing uranium and progeny particularly radon (Rn-222), to the environment. Releases may occur through exhaust or ventilation systems, dewatering operations, crushing, grinding and extraction processes, packaging and active or inactive workings and tailings piles. Whereas the type and quantity of any radionuclide release is site and mill circuit specific,it is possible to make some general observations. First, however, it is necessary to gain insight about the uranium decay series.

URANIUM AND PROGENY

Decay series found in the natural radiation environment are illustrated in Fig. 3. Natural uranium contains both the uranium and actinium series with U-238 and U-235 in the portions of about 140 to 1. Mass, activity and body burden information for natural uranium is provided in Table II. U-235 and progeny are present in such small amounts that they do not constitute a health hazard. Incidentally, very little thorium has been mined in the United States. However, if thorium production increases in the future, its more energetic decay products and their resultant lower biological tolerances might create a tailings disposal problem more

serious than uranium. Thus, the main concern is the fate of the uranium (U-238) series and its decay characteristics are further described in Fig. 4.

The uranium series may be reduced to three sub-series: decay of uranium and progeny to the production of radon (Rn-222) gas, the short-lived progeny of radon which terminate with the formation of relatively long-lived Pb-210 (21 yr.), and the decay of Pb-210 through the final chain to stable lead.

Regulatory guidance for releases of uranium and progeny may be in the form of dose or concentration limits for restricted or unrestricted areas as dictated in 10CFR20, total annual dose equivalence from the entire nuclear fuel cycle as set forth in the US EPA requirements found in 40CFR190, worker uranium bioassay and ALARA suggestions found in US NRC Regulatory Guides and the U.S. Public Health Service "Working Level" (WL) concept which assesses the impact of short-lived radon daughters. In the past the WL philosophy was used primarily for monitoring underground mine atmospheres but it has been extended to interpret the radiological impact of radon daughters released from other sources such as tailing piles. The short-lived daughters are heavy metals which rapidly attach to available surfaces such as small airborne particles or aerosols which, in turn, may be deposited in the respiratory tract. They provide the major source term for calculating inhalation dose. By definition, one WL is any combination of short-lived radon daughters in one liter of air that will result in the release of 1.3×10^5 MeV of alpha energy. For comparison, 100 pCi/ℓ Rn-222 in secular equilibrium with its short-lived daughters is equivalent to 1 WL; the 10CFR20 limit is 1 pCi/ℓ or 0.01 WL. However, equilibrium conditions seldom exist.

URANIUM MINE RELEASES

The principal environmental release from underground uranium mines is radon gas exhausted through the exhaust vents. About 15 Ci of radon are released to the atmosphere per ton of U_3O_8 produced. This source term ceases after the mine is properly decomissioned, but the raw ore carried to the surface will continue to emit radon from the tailings pile. It is possible to backfill the mine with tailings but this tactic is not economically attractive. In addition, milling operations yield a tails which occupies more volume than the original ore.

Open pit mines release about 3 Ci radon per ton of U_3O_8 during mine operation and about 0.2 Ci residual after shutdown. The residual release is about five times larger than naturally exhaled radon released from an equivalent undisturbed area which has a background uranium concentration of 4 ppm.

PROGENY IN THE URANIUM MILL CIRCUIT

Steps basic to all mills are crushing, grinding, chemical leaching and recovering the uranium from a leach solution. The circuits may be either acid or alkaline and use ion exchange and/or solvent extraction. About 80% of the current milling capacity involves the use of sulfuric acid leach, the preferred process for ores with low lime content (12% or less). Ores with high lime content require excessive quantities of acid for neutralization and are best extracted by alkaline (carbonate) leaching for economic reasons. Since acid and alkaline chemical processes do not solubilize the progeny to the same extent it is necessary to study each circuit separately.

A radium and thorium balance for a typical acid mill circuit is shown in Fig. 5. The low pH of the barren leach liquor has a tendency to extract significant amounts of radium and up to 50% of the thorium into the pregnant solution. Much of these products are extracted from the yellowcake and report in the liquid effluent, or raffinate, to the tailings pond. Liquid concentrations of radium and thorium may be several thousand times the MPC limits for releases to unrestricted areas. The yellowcake may have up to 5% of the initial thorium activity, and up to 0.2% of the radium,lead, and polonium activity. About 90% of the uranium is successfully extracted and precipitated into yellowcake and, in general, the quantity extracted is higher than for alkaline leach processes.

As shown in Fig. 6, the alkaline leach process is more selective than the acid leach. The thorium remains virtually insoluble and lesser amounts of radium are extracted into the leach liquor. However, most of the extracted radium reports to the yellowcake. The tailings effluent concentrations are above MPC, but far below the values experienced in acid leach. Water requirements and seepage of tailings solutions are about 30% to 80% to that of the acid mill.

TAILINGS PILE RADIONUCLIDES

Sands (particles between 75 and 500 microns) represent about 70% of the solids. Slimes (particles less than 75 microns) comprise the remainder of the solids and consist of various silica complexes; 70% of slimes are less than 45 microns. The slimes contain higher concentrations of uranium and progeny. Over 80% of the radioactivity in the tailings may be located in the slimes. In addition, uranium and progeny concentrations appear to increase as particle size decreases and makes the smaller, respirable particles potentially more hazardous. Furthermore, when the mill ceases operations and the piles dry out, the dissolved radioactive materials in the pond water will crystalize and become part of the slime fraction. The solid tailings contain about 60% or more of the total long-lived radioactivity originally present in the ore, including about 10% or the uranium, 90% of the thorium and over 99% of the radium, lead, and polonium.

The liquid portion of the tailings component is equal in weight to the solids and is used in the tailings system as a medium to transport the sand and slimes to the final place of deposition. Depending on the process and nature of the liquid, recycling varies from about 0 to 80%. Table III shows typical concentrations and limits for the various nuclides in the tailings solution.

The transport of tailings contaminants is a complex function of parameters including conductivity and dispersivity of the sub-soils and underlying strata, hydraulic variance and underlying ground water formation, ionic exchange, and amounts of precipitation and evaporation. Fortunately, most ponds or tailings disposal sites are located on clay type soils and in an arid environment with little rainfall. Water tables are usually well below the surface. Consequently, the environment provides a considerable safety margin. The fine particles usually seal the bottom of the pond so most of the seepage occurs during early life of the pond and does not continue indefinitely. Alkaline leach tailings can be expected to seal the pond more rapidly than acid tailings because the particle size of the tailings is much smaller (ore is more finely ground). Tailings from a clay type ore should form a seal more rapidly than a sandy ore. If the pond is sited on a clay soil the highly acidic effluent from the acid leach mill will disperse the soil particles which, in turn, clog the void spaces and stop further seepage. Conversely, the acid effluent can

dissolve a limestone soil, causing tunneling and an increase in the seepage rate.

The tailings may be neutralized or treated with compounds which precipitate the soluble radionuclides. Neutralization of acid tailings to pH 8 will precipitate up to 90% of the dissolved radium and almost all the thorium. Additional radium may be precipitated if the solution is treated with selected barium compounds, but the solutions must be free of particulates since the presence of solid materials in the solution has been found to be detrimental to the reaction. Calcium is found in many tailings effluents and is known to interfere with the precipitation of radium. Neutralization is of limited value in treating alkaline waste since most of the species found in acid wastes are not initially extracted into alkaline solutions. Copperas may be added to assist in radium removal. It should be noted that even stabilized dried tailings have the potential to release radium if contacted by rain or groundwater. One possible solution to the radium problem in acid circuit may be to leach both the uranium and thorium with nitric acid, then chemically separate the radium and dispose it as a concentrated radwaste.

Typical uncovered tailings will yield gamma radiation on the order of a few mR/hr at 3 feet above the ground. A radiation survey conducted at the inactive Tuba City tailings site is shown in Fig. 7. If the tailings are not properly covered or stabilized during decommissioning efforts, the solids may be blown onto adjacent lands. Fig. 8 shows the increased radiation downwind of the inactive Tuba City site. Though not shown, increased radium concentrations were also observed at downwind locations.

RADON EMANATIONS

Radon may be released through active mine ventilation systems as well as from active and inactive open pits, tailings piles or storage locations. Operating mills release radon during the crushing and grinding phases. Radon releases are affected by changes in barometric pressure, temperature, humidity and wind velocity. Radon generation in the tailings is dependent on the concentration of radium which is not consistent or constant in the vertical and horizontal profiles because of variations in the quality of the ores processed through the years. Also, the differential settling of the tailings solids after discharge to the pond results in spatial fractionation, the larger tailing sands being deposited closer to the point of effluent discharge. In addition, much of

the radium activity in the tailings is associated with the finer grains which are washed to lower areas and become buried at greater depths than the larger grains. The draft GEIS on Uranium Milling (NUREG-0511) model tailings pile serves a $1x10^3$ ton/yr yellowcake mill and releases $7x10^3$ Ci/yr Rn-222. Radon concentrations above the tailings may range from a few tenths of a pCi/ℓ upwards to perhaps 34 pCi/ℓ. Background concentrations range from a few tenths of a pCi/ℓ to perhaps 30 pCi/ℓ in energy efficient dwellings. The radon releases constitute the largest release and potential dose to populations, particularly those not living adjacent to or in the immediate vicinity of the tailings. Dose information for the model mill situation is found in Appendix G to NUREG-0511.

SUMMARY AND CONCLUSIONS

Considerable information is available in the published literature regarding the fate of uranium progeny under various conditions and a GEIS has been prepared for a model uranium milling operation. About half of the current uranium comes from underground mining and half from open pit operations. Most uranium extraction occurs at conventional mills, but solution mining and by-product recovery is receiving increased attention. Most of the uranium recovery activity occurs in the western United States. Many new mines and mills, primarily located in the western United States, will be required to satisfy future fuel requirements. With respect to uranium progeny:

- radon is the significant radionuclide released at the mine and mill and its short-lived daughters contribute to inhalation doses.
- radon concentrations are highest at underground mine exhausts, in open pits and atop the tailings piles.
- radon dissipates and levels drop rapidly at short distances from the release point.
- radon concentrations atop mill tailings are somewhat larger than concentrations which may appear in energy efficient homes.
- more thorium and radium is solubilized by acid leaching circuits.
- neutralization is effective in removing radium and thorium from acid tailings effluents. Barium chemicals may provide additional removal under specific conditions.

- dry tailings ponds may release radioactivity through windborne dispersion.
- rain or groundwater intrusion may redissolve and transport precipitated radium found in dry inactive tailings ponds.
- radioactivity concentrations are much greater in the slime portion of the solids.
- radioactivity concentrations increase as particle size decreases.

TABLES AND FIGURES

Table I. Potential Resources of Uranium from Conventional Sources*

State	Share of Probable Resources, %
New Mexico	30
Wyoming	15
Colorado	11
Utah	14
Texas	10
California	2
Arizona	4
South Dakota	1
Nevada	2
Washington	2

* From Draft GEIS on Uranium Milling, NUREG 0511, April 1979.

Table II. Characteristics of Natural Uranium and Progeny in Secular Equilibrium

Unat	g/gUnat	μCi/gUnat	MPBB (μCi)*
U-238	99.280	0.33	
U-235	0.715	0.01	5E-3+ Kidney
U-234	0.005	0.33	
U-238 Decay			
U-238 (α)	9.9E-1	0.33	5E-3 Kidney
Th-234	1.4E-14	0.33	4E 0 Bone
Pa-234	5.0E-16	0.33	-
U-234	5.4E-5	0.33	5E-2 Bone
Th-230 (α)	1.8E-5	0.33	5E-2 Bone
Ra-226 (α)	3.3E-7	0.33	1E-1 Bone
Rn-222 (α)	2.1E-12	0.33	-
Po-218 (α)	1.2E-15	0.33	-
Pb-214	1.0E-14	0.33	-
Bi-214	7.5E-15	0.33	-
Po-214 (α)	1.0E-21	0.33	-
Pb-210	4.0E-9	0.33	4E-1 Kidney
Bi-210	2.2E-12	0.33	4E-2 Kidney
Po-210 (α)	7.3E-13	0.33	3E-3 Spleen/ Kidney
Pb-206	-	-	-

Summary

Unat: 0.67 μCi/gUnat and 0.67 μCi(α)/gUnat†

U-238 and Progeny: 4.6μCi/gUnat or 2.6μCi(α)/gUnat

U-235: Mass insignificant but 0.11 μCi/gUnat or 0.07 μCi(α)/gUnat

* From U. S. Department of Commerce Handbook 69.

\+ 5E-3 = 5×10^{-3}

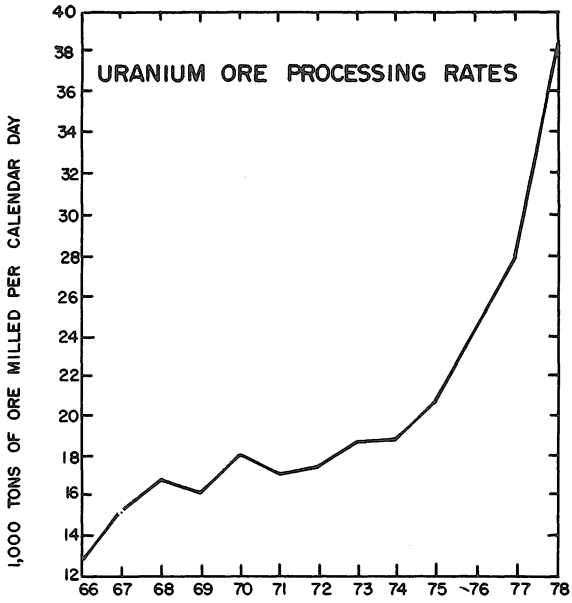
† μCi(α) implies activity from alpha particle decay

Table III. Typical Concentrations of Radionuclides in Acid Tailings Solution

Radionuclide	Concentration μCi/mL *	Maximum Permissible Concentration in Unrestricted Areas, μCi/mL+
U-238	5.4 E-6	4 E-5
U-235	5.4 E-6	3 E-5
Th-230	1.5 E-4	2 E-6
Ra-226	4.0 E-7	3 E-8
Rb-210	4.0 E-7	1 E-7
Bi-210	4.0 E-7	4 E-5
Po-210	4.0 E-7	7 E-7

* From Draft GEIS on Uranium Milling, NUREG 0511, April 1979.

+ From Rules and Regulations, Title 10 - Chapter I, Code of Federal Regulations, Part 20, Standards for Protection Against Radiation, U. S. Nuclear Regulatory Commission.



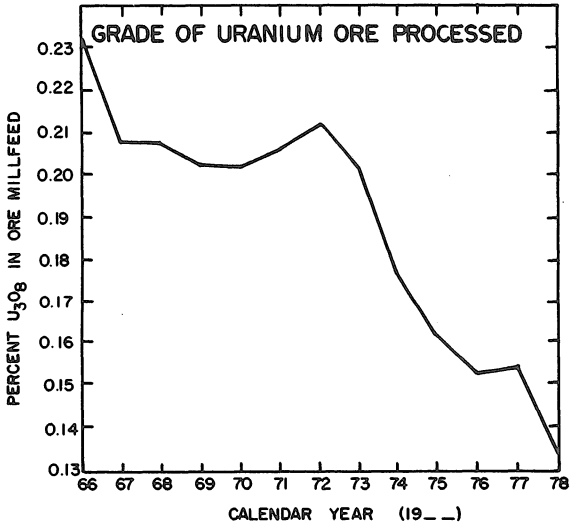
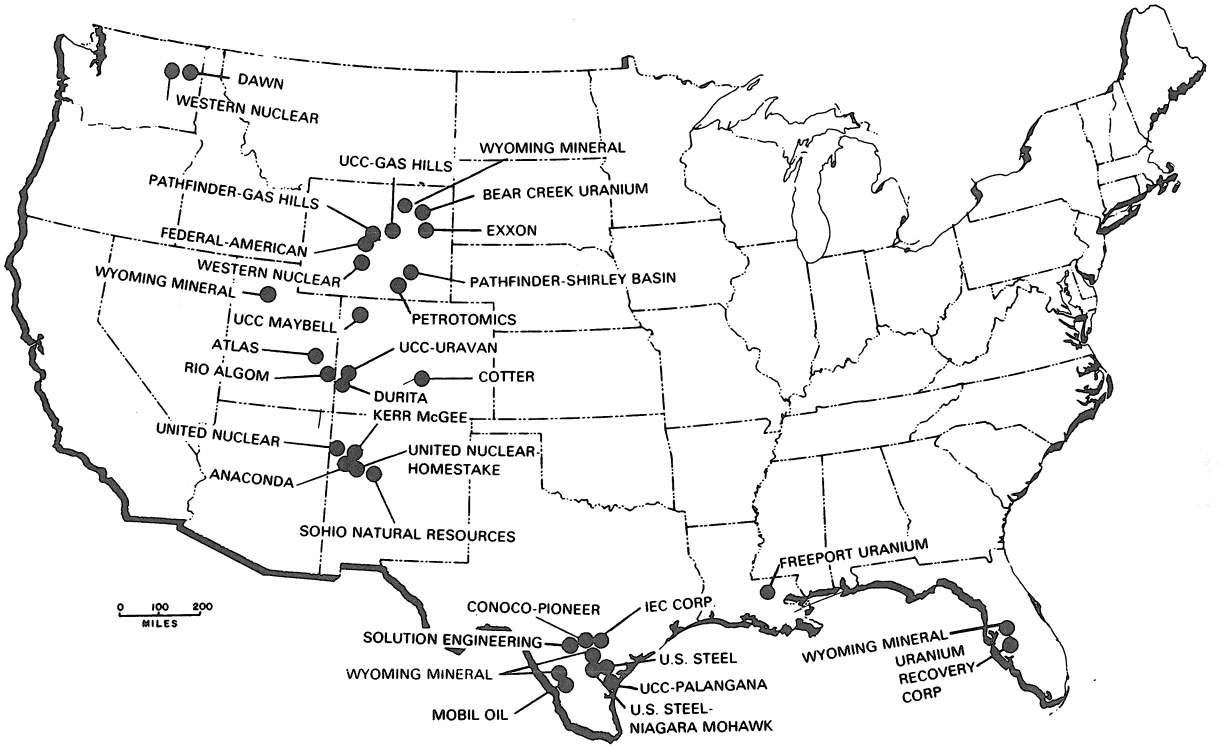


FIG. 1 - AMOUNT AND GRADE OF URANIUM ORE PROCESSED



Reference: Statistical Data of the Uranium Industry, U.S. Department of Energy, GJO-100(78), 1979.

FIG. 2 - **URANIUM MILLS IN THE UNITED STATES OPERATING AS OF 1/1/79**

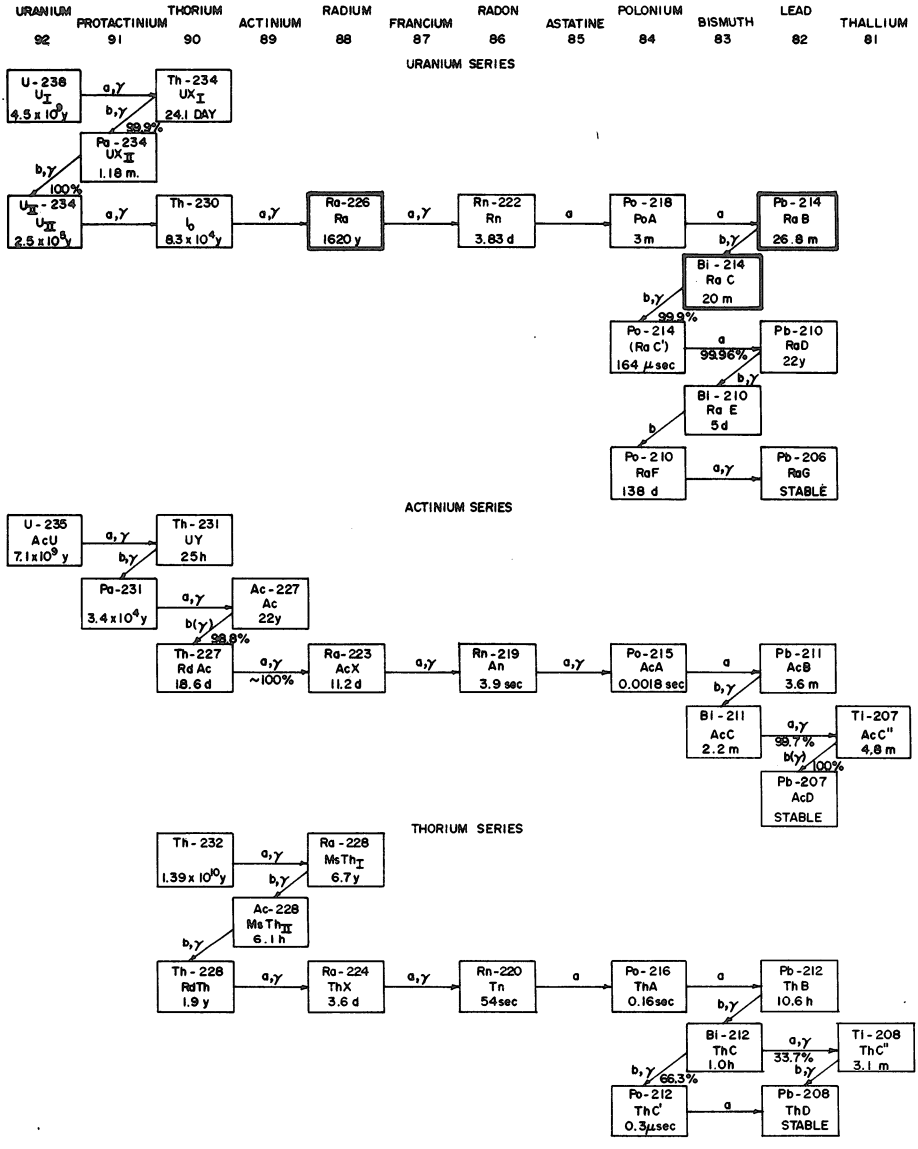


Fig. 3 - Radioactive Decay Chains for Uranium and Thorium

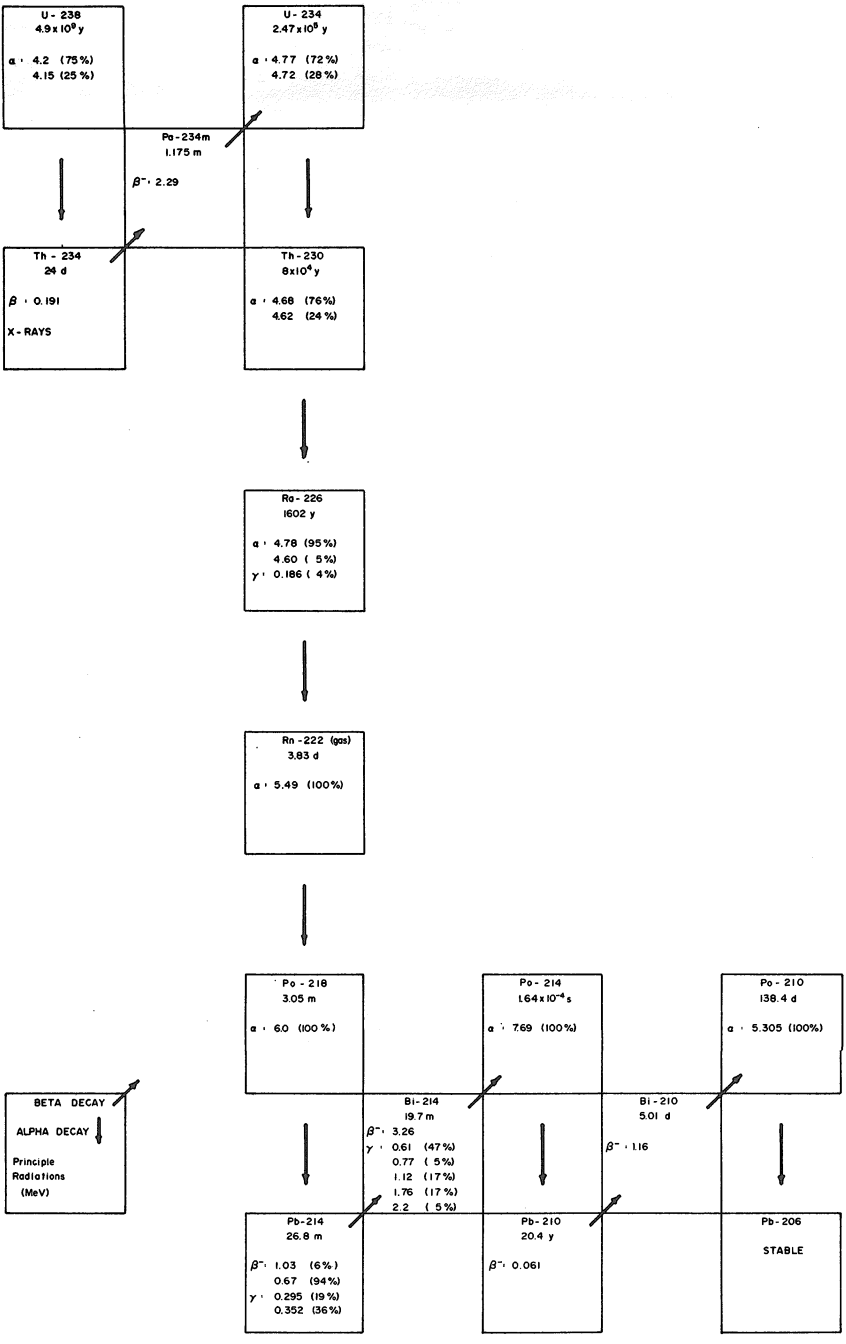


Fig. 4 - Principle Decay Scheme of the Uranium Series

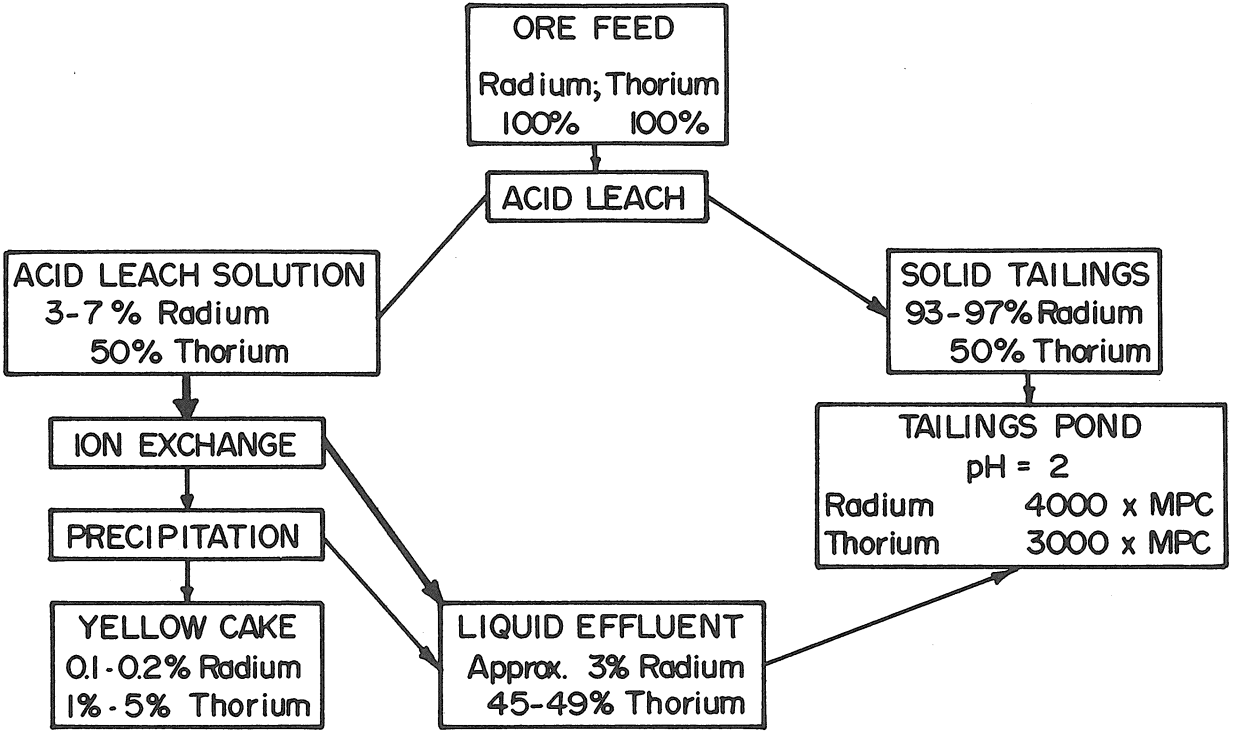


FIG. 5 - Radium and Thorium Balance in Acid Mill Circuits

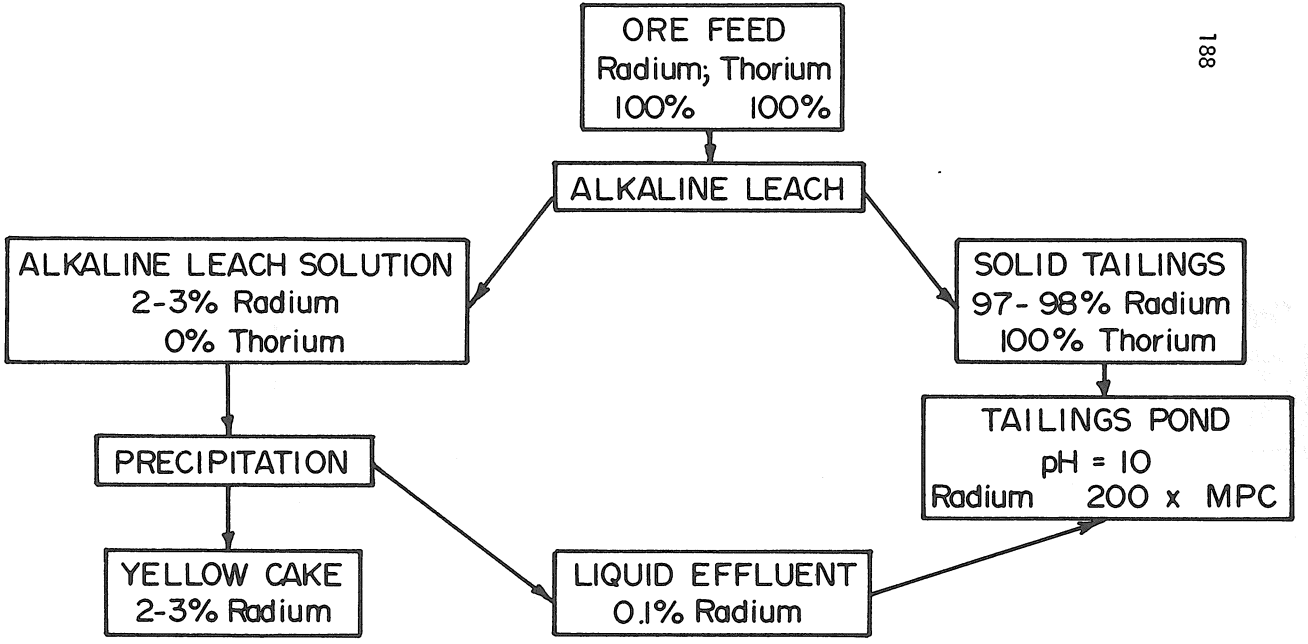


FIG. 6 - Radium and Thorium Balance in Alkaline Mill Circuits

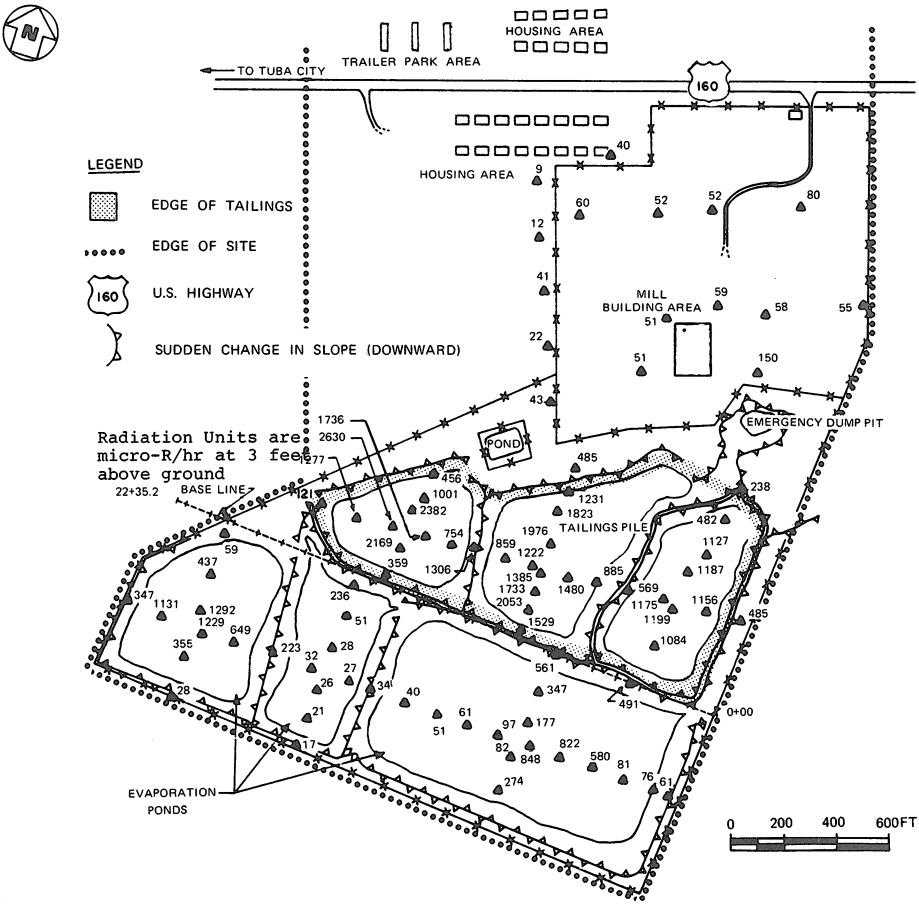
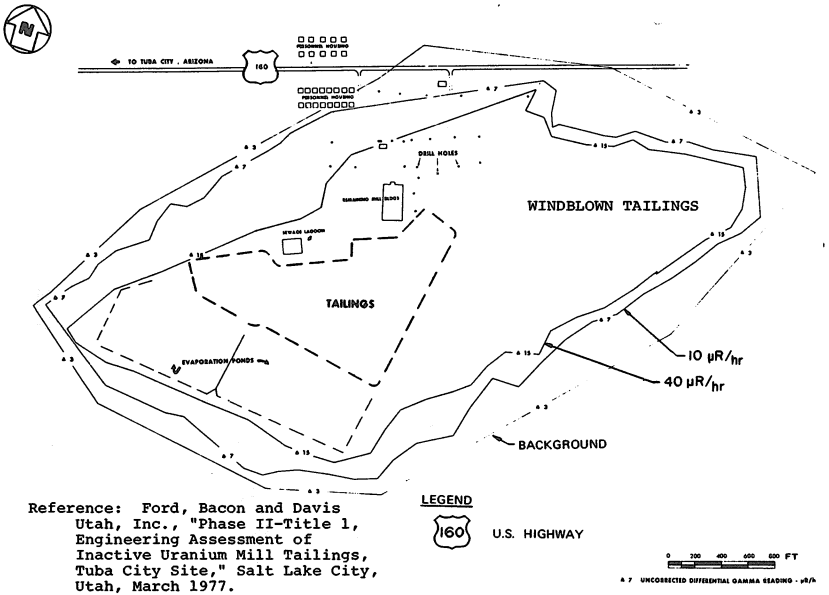


Fig. 7 - Gamma Survey at Inactive Mill Site, Tuba City Arizona



Reference: Ford, Bacon and Davis Utah, Inc., "Phase II-Title 1, Engineering Assessment of Inactive Uranium Mill Tailings, Tuba City Site," Salt Lake City, Utah, March 1977.

Fig. 8 - Gamma Survey Surrounding Inactive Mill Site, Tuba City, Arizona

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