



Radioactivity in mines and mine water—sources and mechanisms

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Synopsis

Uranium is a naturally occurring radioactive material in the earth's crust. Geological processes have resulted in concentration of uranium into ore mined for its gold content in South Africa. This uranium is present in the form of discrete particles of uranium minerals, generally oxides, which are normally insoluble. Chemical and physical processes during gold mining and extraction, particularly when carried out in conjunction with uranium extraction and sulphuric acid manufacture, act on the uranium minerals and the radioactive decay products of uranium. This may result in significant accumulations of radioactive materials in certain parts of the process.

There are a number of naturally occurring radioactive materials in the earth's crust, such as Uranium-235, Thorium-232, and Potassium-40. This paper focuses on Uranium-238, together with its decay products, and how they affect the gold mines.

Water plays a significant role in the redistribution of radionuclides, both as a physical transport medium, and as a solvent in which chemical reactions take place.

Introduction

Uranium is a naturally occurring radioactive material in the earth's crust, with an abundance of 2.7 g/t. Table I shows the abundance of selected elements in the earth's crust, for comparison.

Because uranium is radioactive, it is found in nature in association with its decay products.

The chemistry and geochemistry of uranium is complex because of numerous oxidation states and the stability of various complexes.

The presence of uranium in ores currently mined for their gold content required at least three geological processes; namely, the concentration of uranium into almost pure blocks of uranium minerals, the weathering of these blocks into a granular detrital form, and finally the gravity concentration of these weathering products into the reefs in the Witwatersrand basin.

Water played a critical role in these geological processes, both by assisting in chemical transformations and by physical activities such as the breaking down of large rocks into smaller particles and the transport of these particles from one place to another.

Water plays an equally important part in the mining and processing of ores, both as a physical transport medium, and by selective chemical interaction with certain components of the ore.

The identification of uranium in Witwatersrand ores dates back to 1915¹. It was known that uranium was radioactive, but this was not considered to be particularly hazardous. An early reference to a radiation hazard associated with acid plants dates back to 1974². Acid plants again featured prominently in 1992, when the Council for Nuclear Safety (CNS) believed that there may have been overexposures to radiation of workers during annual maintenance activities at an acid plant, and introduced stringent safety measures to reduce such exposures.

Radioactivity in liquid effluents associated with the gold mining industry achieved some prominence in 1996 with the publication of an overview of radioactivity in water sources by the Institute of Water Quality Studies³.

Table I
Composition of the continental crust
Average content of some trace elements⁶

Symbol	Name	g/t
Ba	Barium	425
Sr	Strontium	375
Zr	Zirconium	165
Cu	Copper	55
Sc	Scandium	22
Pb	Lead	12.5
U	Uranium	2.7
Hg	Mercury	0.08
Ag	Silver	0.07
Au	Gold	0.004

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Table II

The uranium decay chain, and mass equivalents of the uranium decay series for 1 curie of activity

Nuclide	Half life	Decay mode	Mass required for 1 Curie activity
Uranium-238	4.468x10 ⁹ y	alpha	3.2 tons
Thorium-234	24.1 d	beta	32.2 micrograms
Protactinium-234	1.17 m	beta	1.48 nanograms
Uranium-234	2.4x10 ⁵ y	alpha	164 grams
Thorium-230	7.55x10 ⁴ y	alpha	51.4 grams
Radium-226	1.6x10 ³ y	alpha	1.00 grams
Radon-222	3.83 d	alpha	6.49 micrograms
Polonium-218	3.1 m	alpha	3.56 nanograms
Lead-214	26.8 m	beta	30.6 nanograms
Bismuth-214	20 m	beta	22.5 nanograms
Polonium-214	1.6x10 ⁻⁴ s	alpha	3.11 femtograms
Lead-210	22 y	beta	11.4 milligrams
Bismuth-210	5 d	beta	8.07 micrograms
Polonium-210	1.38x10 ² d	alpha	223 micrograms
Lead-206	Stable		

While the CNS and the Department of Water Affairs and Forestry are primarily concerned with the existence of radionuclides in water sources, this paper outlines mechanisms, associated with gold mines, which explain the presence of such radionuclides.

A Curie of Rock

Consider a block of rock, 100 m long, 100 m wide and 1 m thick. This slice of rock has a mass of about 27 000 tons. At a uranium grade of 142 g/t of U₃O₈, this mass of rock contains about 3.2 tons of uranium, and at a gold grade of 5 g/t, 135 kg of gold.

It also contains exactly 1 gram of Radium-226. The original unit of quantity of radioactivity was the Curie, being by definition, exactly 1 gram of radium. I therefore call this block of rock a 'Curie of Rock'.

Every second, in this Curie of Rock, 3.7×10^{10} atoms of uranium and each of its daughter products decay by emitting either an alpha or a beta particle. The amount of each daughter product in the Curie of Rock is related to the stability of that daughter product which is expressed as the half-life. The relative proportions of the daughter products are as indicated in Table II.

For practical purposes, over time periods measured in human life times, the quantity of each daughter product in the Curie of Rock will remain constant at the level indicated in Table II, as the rate of decay of each daughter product exactly balances the rate of formation from higher up in the series. Over geological time periods, the amount of uranium will decline. Over the next 4.5 thousand million years, half of the uranium atoms present in the Curie of Rock will decay, and be converted into lead. The current Curie of Rock will have turned into a half Curie of Rock.

How it got there

Uranium is a naturally occurring constituent of the meteorites⁴ from which the earth agglomerated some 5–10 thousand million years ago. Since then geological forces,

such as volcanism and the action of water, have reworked most of the constituents of the primordial crust a number of times.

World wide, uranium occurs naturally at enhanced concentrations in several common rock types, including granite, limestone and dolomite. Further research is required to quantify this in South Africa.

Although uranium is generally considered to be insoluble in natural waters, it is a naturally occurring constituent of sea water at concentrations of about 3 ppb, and its presence has been documented in many inland natural waters at concentrations about this order of magnitude.

One of the early geological processes resulted in the concentration of uranium into fairly pure bodies of uranium minerals. There is one well-documented example⁵ of a uranium ore body of sufficient size and purity to start a self-sustaining fission reaction, i.e. this ore body was a naturally occurring nuclear reactor for a period of some 1 billion years, becoming critical some 3 billion years ago.

Weathering broke up these accumulations of uranium minerals, as well as gold, pyrite and other similar heavy minerals, into discrete grains which were then washed from the mountains into the valleys and lakes. The current Witwatersrand basin is an example of such a prehistoric lake. The action of wind and water in this lake finally resulted in the gravity concentrations of the heavy minerals into thin layers we now refer to as 'Reef'.

The reefs currently being mined were laid down some 700 to 1 200¹ million years ago, and were covered by several kilometres of other products of weathering. There followed periods of crustal movement which caused the bottom of the lake to become the high ground which is now being weathered in turn. This process of faulting, upliftment and weathering has resulted in outcrops of reef on surface, which has caused widespread dispersal of uranium into the environment. The best known example of such an outcrop is the Witwatersrand itself. Most of the gold in the outcrops was mined many years ago, so that most of the existing gold mines are now extracting ore which has been buried under several kilometres of rock for several hundred million years, well away from the action of wind and water.

The significance of this isolation is that the daughter products have not had the opportunity to escape from the host rock and are now substantially in equilibrium in the concentrations indicated in Table II.

An important characteristic of radioactivity is that it is not possible to predict which atoms in a particular mass will decay and that it is not possible to influence an atom to either decay or not. This characteristic ensures that the daughter products are homogeneously dispersed in their host matrix.

Physical and chemical processes

Physical processes operate on bulk materials, for example breaking a large rock down into smaller units, or moving a rock from one place to another. Water acting as a physical transport medium is one of the principal agents responsible for geological reprocessing of the earth's crust.

Chemical processes operate on an atomic scale, and change the chemical nature of the material they operate on, for example dissolving a material in one place and precipitating it as a scale in another.

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Because water dissolves almost all known materials to some extent, it is an important agent in geological processes, and also in the processing of ore by man. The chemical potency of water can be significantly enhanced by changes in factors such as temperature, pH value and redox potential, as well as the presence of other dissolved materials.

The chemistry of uranium and radium

The chemistry, and the geochemistry, of uranium is complex because uranium has many oxidation states, and forms many stable compounds in nature. About 100 naturally occurring uranium minerals have been identified⁶.

It is dangerous to generalize about the chemical behaviour of both uranium and radium, because it is the exceptions to generally expected behaviour that contribute to finding radioactive materials in unexpected places.

Uranium in the environment is generally measured in concentrations expressed as parts per billion, or parts per 10⁹ parts, or micrograms per litre, which are numerically equivalent. Highly specialized and expensive laboratory techniques, such as neutron activation analysis, are required to carry out these measurements, particularly in the lower ranges.

Most uranium minerals are insoluble, but the uranyl ion (UO₂⁺⁺) is highly soluble, and uranyl sulphate, carbonate and nitrate may give rise to significant concentrations of soluble uranium in mines and mine waters.

Before uranium leaching, radium is homogeneously dispersed in the bulk of uranium mineral grains at a relative concentration of about 0.3 ppm of radium within the uranium matrix. Leaching of radium prior to uranium leaching is restricted to the surface of the uranium mineral grain. Uranium leaching destroys the uranium matrix which locks the radium in place, and therefore mobilises the radium so that it may interact with the surrounding aqueous environment.

The literature is ambiguous about the behaviour of radium at very low concentrations. Combrink⁷ states that 'Radium sulphate has a very low solubility: (0.02 microgram/l in cold water)', but in the same reference he states that 'Radium is extracted from uraninite with uranium in dilute sulphuric acid'. In the circuit shown in Figure 4, no sign of radium build-up is observed in the uranium leaching section of the pyrite stream, but radium precipitates as enhanced activity gamma emitting insoluble deposits or scales further downstream, as conditions such as the pH value change. The discovery of high activity gamma-emitting scales in the pyrite filter section of this plant came as a complete surprise.

It is worth noting that radium at these low concentrations is not detectable by conventional chemical techniques, and is detectable only by measurement of its radioactivity.

Radium dissolves under the conditions used for the leaching of uranium, but is generally insoluble in conditions found in the environment. Although uranium and radium tend to go into solution together, the differences in their chemical behaviour are sufficient to ensure that their subsequent movement is independent of each other. Radium moves through the environment very much more slowly than uranium. This applies particularly to leakage of radionuclides from tailings dams⁸, which effectively lock up radium but

allow some uranium leakage. When looking at radionuclides in waters, the ratio of uranium to radium is unpredictable.

Water reticulation

Mine water reticulation systems, both underground and on surface plants are complex with substantial internal recirculations, and a complex mix of ions in solutions, resulting from deliberate changes in pH value and redox potential. A full description of the movement of radionuclides in a typical mine requires a detailed understanding of the water reticulation system as well as the specific process metallurgy. A section or process which may have a radiation problem on one mine, may be free of such a problem on another mine.

Ore processing

Ore processing consists of a series of physical and chemical steps starting with mining, usually followed by crushing and milling, to liberate the individual mineral grains of interest, and then separating the mineral components by physical processes such as flotation, or chemical processes such as leaching, before the residue material is finally deposited on tailings dams.

In explaining the movement of radioactive materials, it is important to understand the chemical and physical forces which act on the ore as it passes through the recovery process. The sequence of the processing steps may have a significant influence on the movement of radioactive materials.

Figures 1 to 4 show a number of fairly typical gold recovery processes.

Figure 1 is one of the simplest processes for gold extraction, typified by modern Carbon in Pulp (CIP) plants. The ore is treated as a single stream in a single pass operation with no uranium or pyrite extraction.

Figure 2 typifies older circuits where the uranium grades were not sufficient to justify uranium extraction, but pyrite was extracted for supply to a pyrite burning acid plant. These acid plants, under the auspices of the Acid Distribution Committee of the Chamber of Mines, produced the acid



Figure 1—The simplest gold recovery process. Very low potential for radiation hazard from fortuitous gravity concentration (for example in launders)

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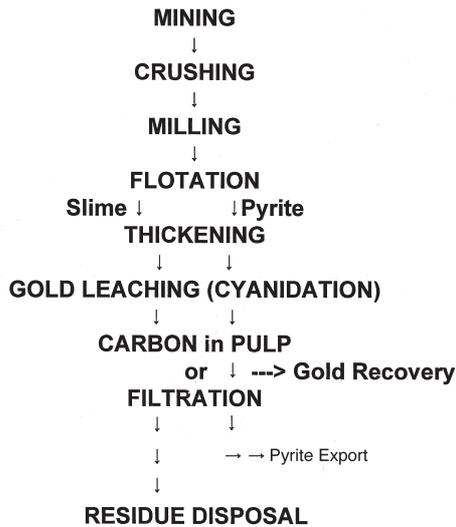


Figure 2—Simple gold recovery process with flotation. Increased potential for radiation hazard from uranium concentration into pyrite

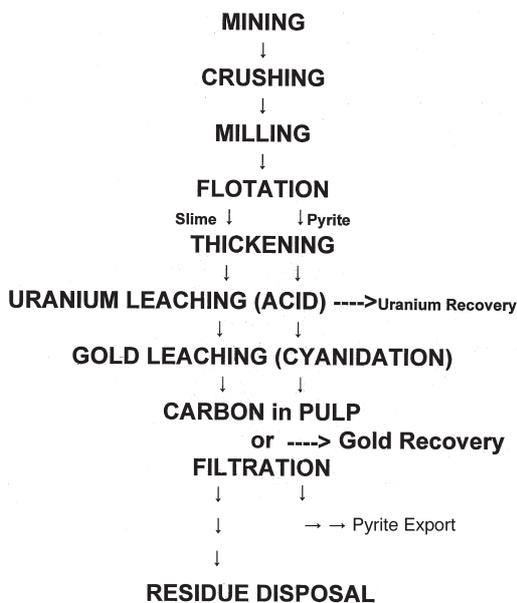


Figure 3—A gold recovery process with flotation and reverse leach for Uranium Extraction. High potential for radiation hazard from radium mobilized during uranium extraction

required by the uranium plants then in production. The two sets of arrows in the figure indicate that slime and pyrite are treated in separate but parallel processes.

Figure 3 introduces uranium leaching after flotation. Pyrite is supplied to an acid plant after gold and uranium extraction.

Figure 4. In addition to gold and uranium extraction, this diagram shows locally produced, and imported, pyrite being burned in an on-site acid plant. The calcine from this operation is usually recycled to the uranium and gold leaching processes (not shown in Figure 4).

These examples are far from comprehensive, and there are almost as many variations as there are gold mines. Some of the variations include uranium recovery after gold recovery, and flotation after gold and uranium recovery. In

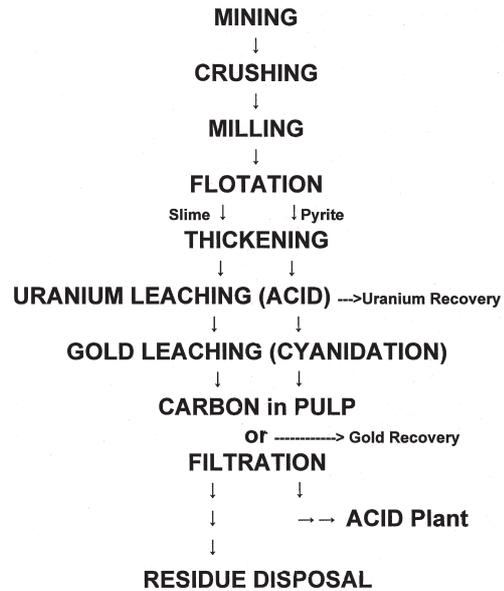


Figure 4—A gold recovery process with flotation, reverse leach for uranium extraction, and sulphuric acid manufacture. Additional high potential for radiation hazard from radium mobilized in solution streams in acid plants

addition there are a number of alternative uranium extraction technologies, including fixed bed ion exchange, and solvent extraction. A comprehensive analysis of all these variations is beyond the scope of this paper, some of these variations relating to uranium extraction are described by Ford⁹. The effect on radionuclides of the unit operations will be covered in the following sections.

Mining

As mining of our Curie of Rock starts, there is initial fracturing of the rock, which allows air to penetrate to the rock, and allows radon gas which was trapped in the rock to escape into the air-filled cracks in the rock. Radon is also highly soluble in water and can be transported over significant distances in the underground environment. This can represent a significant transport mechanism in very wet mines. This radon gas is a significant source of exposure to radioactivity in the underground environment but will not be considered further in this paper.

An important step in mining is the drilling of the rock face and blasting of the rock using nitrate based explosives. After blasting the rock comes into contact with water. Water in the underground environment contains a complex and highly corrosive mix of ions, some of which are capable of forming soluble uranium compounds.

Sulphur in pyrite may oxidize to form sulphuric acid. Iron derived from the oxidation of pyrite, reacting with dissolved oxygen will generate ferric iron in solution which enhances uranium leaching. This acidity may then be neutralized in underground water treatment plants, where the pH value may be adjusted with lime or sodium carbonate, thus providing a source of carbonate ions. The use of nitrate based explosives gives rise to significant concentrations of nitrate in mine water circuits.

The chemical nature of the uranium in underground water is as a uranyl ion associated with sulphate, nitrate or

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carbonate. Uranium has been found to precipitate out in certain underground water reticulation systems as a result of changes in pH value. There are some references¹⁰ in the literature to radium 'scales' (i.e. radium-bearing deposits on surfaces) in underground water systems, and these may occur in some mines. The author's experience of these scales on two gold mines indicates only the presence of chemically separated uranium, with no significant radium content. The uranium concentrations in the scale may be as high as several kg/ton, but no crystalline uranium phase could be detected by X-ray diffraction. These scales have a distinctive radiation signature when observed with a typical contamination monitor. There is almost no detectable alpha and gamma activity, but count rates of several hundred counts per second of beta activity are observed.

Note that the term 'Mining' could include dump reclamation, with the added complexity that the dumps could have been constructed from material which had only been subjected to gold recovery or to both uranium and acid recovery in the past. Analysis of these factors is beyond the scope of this paper.

Crushing

Crushing is primarily a physical process in which large rocks are broken down into smaller units. Although crushing itself is usually a dry process, it is associated with washing and screening processes, so that new rock surfaces come into contact with water shortly after they are formed.

Water in crushing circuits is usually maintained at neutral to slightly alkaline conditions to prevent scaling and corrosion. Under these conditions, there is little observed leaching or precipitation of uranium in the water reticulation systems. Because the water acts only on the outer surfaces of uranium minerals exposed during crushing, there is no mobilization of radium and no scales with enhanced radioactivity are found in crushing plants.

Gravity concentration effects, generated by the physical transport of grains of heavy minerals in flowing water, in the wash water circuit, may give rise to localized areas containing gold, uranium and pyrite.

There is little scope for chemical processes to occur in the water in the crushing circuit.

Milling

Milling is similar to crushing, except that it is normally a completely wet process, and that the particle sizes are much smaller. The pH value is normally in the neutral to alkaline region, and there is little evidence of uranium leaching and scale precipitation in the mill return water reticulation system.

Gravity concentration may give rise to isolated areas containing heavy minerals; for example, behind mill liners, and in pipes and launders, but there is little scope for chemical concentration of radionuclides to form high activity scales.

Note that, the unit operations of 'Crushing' and 'Milling' can in some cases be combined into a single unit operation 'Run of Mine Milling'.

Thickening

This is a wet process, in which the solids are separated from the liquid. The pH value is the same as in the milling circuit. Again there is little scope for chemical fractionation, and no evidence of enhanced activity scales in the water circuit associated with the thickeners.

Gold leaching by cyanidation

Cyanidation is a wet process which takes place at ambient temperature and high pH values, typically above 11. Cyanide is not known to attack any of the radionuclides of interest, so there is little scope for radionuclides to be removed from the original uranium mineral grains.

Filtration or carbon in pulp (CIP)

In this process, the gold-bearing solution is separated from the slime by filtration, or the gold is absorbed directly onto activated carbon. There is no change in pH value, and little reason to believe that any radionuclides could be dissolved.

It is worth noting that if radium has gone into solution upstream of a CIP plant, its chemistry is sufficiently similar to calcium to co-precipitate with calcium on the carbon. This could result in elevated levels of radioactivity on the carbon.

Residue disposal

The residue disposal operation itself is not a chemical process. The pH values are generally neutral to alkaline in the pumping system and pipe lines to prevent corrosion. To date no enhanced activity scales have been observed in residue pipe lines, and no radium absorption onto rubber linings has been observed.

The long-term chemical stability of radionuclides in tailings dams is another matter. Most tailings dams have residual pyrite in them, which is then subject to atmospheric oxidation to sulphuric acid and ferric iron in solution, both ideal conditions to encourage uranium dissolution. The extent to which this occurs is uncertain, as is the interaction of dissolved uranium with underlying soils and rocks. It is worth noting that those plants which operate flotation plants for the recovery of pyrite will reduce this source of potential environmental pollution.

The unit operations listed here indicate that apart from mining and residue disposal, there is little scope for mobilization of radionuclides in a gold recovery plant as indicated in Figure 1.

Flotation

In older plants constructed during the 1950s, when uranium extraction was high on the agenda, flotation was carried out immediately after milling as indicated in Figure 2. On plants of this nature, flotation usually takes place at high pH values, typically above 11. On newer plants, including dump reclamation plants, and plants where flotation was introduced as an afterthought to gold and uranium extraction, flotation may take place at a pH value of about 4, immediately after uranium leaching.

Flotation is a surface chemical phenomenon, where

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chemicals are added to the slurry to selectively make mineral surfaces attract or repel water. The minerals that are made to repel water will preferentially attach themselves to bubbles of air, and this allows separation from other unaffected mineral types.

In the high pH value circuits, the radionuclides are substantially insoluble, and no enhanced activity scales are found.

In the circuits at low pH values, radium may have been liberated during uranium leaching and may result in enhanced activity scales. The radium may also be absorbed by rubber linings in the circuit. The flotation tailings are neutralized with lime shortly after the flotation process for final disposal, and neutralization precipitates out the radionuclides. This may cause enhanced activity scale formation in, and immediately after, the neutralization process.

Uranium leaching

In South Africa, uranium recovery is carried out by leaching in sulphuric acid at elevated temperatures in the presence of oxidizing agents such as ferric iron and manganese dioxide. Uranium is brought into solution as uranyl sulphate.

Older uranium plants use the fixed bed ion exchange technique to concentrate the uranium first into an eluate containing uranyl nitrate followed by precipitation with ammonia as ammonium diuranate (ADU). Radium is not absorbed onto the resins in this process, and is excluded from the ADU circuit.

Some of the more modern plants use solvent extraction at some point in the ADU circuit. It appears that radium may be transferred to the ADU circuit and generate enhanced activity gamma-emitting scales, and cause contamination of stainless steel components by the process of recoil embedding.

The uranium-bearing solutions are generally contained within the bunded areas of the plant, and spillages are returned to the process. However, the recovery of dissolved uranium from the leaching process is never 100% complete, and some soluble uranium will pass into downstream processes. The fate of this uranium is unclear. It is believed that neutralization with lime after uranium leaching will precipitate the uranium out as a calcium uranate, but there are a number of complexes containing calcium, magnesium, carbonate and uranium compounds, and their behaviour in the environment is uncertain. The solubility of some complexes may be dependent on pH value, while other complexes are stable and independent of pH value, once formed.

Once radium has been mobilized during uranium leaching, it may interact with process components such as rubber linings and stainless steels.

Sulphuric acid manufacture

Sulphuric acid can be manufactured by roasting pyrite, and converting the resulting SO₂ to sulphuric acid. A number of acid plants were constructed during the 1950s to provide the acid required for uranium extraction. For some years it has been known that pyrite burning acid plants have been a source of radium-bearing scales with enhanced activity. Roasting takes place at a temperature of about 800 deg C. Under these conditions the iron sulphide matrix is converted to an iron oxide matrix, and the calcine formed is then

contacted with hot, usually acidic water at about 60 deg C. Under these conditions, it is not surprising that some radium goes into solution and subsequently precipitates out as scales when the temperature of the process streams is reduced. These scales in the 'Weak Acid' sections, together with associated downstream equipment for handling calcine are the major source of radioactivity on pyrite-burning acid plants.

Conclusion

Radioactive materials such as uranium and radium are a natural part of our environment. Human activities such as mining accelerate natural geological processes such as the reprocessing of the earth's crust, by transferring ore from underground to tailings dams on surface.

Water plays an important role in moving naturally occurring radionuclides through the environment, both as a transport medium for bulk ore, and as a medium for chemical reactions.

Man's activities enhance such water-borne transport of radionuclides into the environment. In some cases, radionuclides are separated from the bulk ore and may pass into the environment via water streams, for example, uranium in underground water pumped to surface, and uranium leakage from tailings dams resulting from oxidation of pyrite. (On the other hand, radium is effectively immobilised in tailings dams.) Where mines have uranium or acid plants, there is, in addition, significantly enhanced scope for uranium and radium, mobilised during the leaching process, to escape into the environment via water streams.

As our understanding of these processes improves, we can expect to develop cost effective methods of control. On at least one acid plant, methods have been found for chemically dispersing existing radium-bearing scales, and preventing their formation in the first place¹¹.

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