THE MODES OF OCCURRENCE OF Ra-226 AND ITS IMMEDIATE DAUGHTER PRODUCTS IN URANIUM TAILINGS SAMPLES FROM THE MONTICELLO MILL, UTAH

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ABSTRACT

A nuclear-emulsion feasibility study of the modes of occurrence of Ra-226 and its immediate alpha-emitting daughter products has been completed. The study involved approximately 40 samples of drill core from the Monticello Mill, Monticello, Utah. The drill core was studied using the dispersed-grain nuclear-emulsion technique, a micro-autoradiographic procedure which permits the direct observation of the alpha activity of these radionuclides and their association with individual mineral grains or particles. This work was undertaken because nuclear-emulsion techniques have not been used in the study of uranium-mill-tailings and because it was important to explore the possible contributions that these untried techniques might make to the remedial-action program. Specifically, it was hoped that it would be possible to supplement the bulk measurements of uranium, radium, radon, and other radionuclide concentrations in the tailings with information on how these radionuclides are distributed, i.e., their modes of occurrence, mineralogy, and relative mobility.

Four modes of occurrence of Ra-226 and its immediate daughter have been described and photographed. The most abundent phase is believed to be a chemical combination of radium with sulfate most probably as a surface reaction with the mineral $(Ra,Ba)SO_4$. These "radiobarites" range in size from tens of microns to fractions of a micron. Some of the tailings samples contain tens to hundreds of "radiobarites" per milligram of sample. The second mode of occurrence is characterized by point sources of alpha activity which are not associated with visible grains or particles. At least some of these point sources are believed to be colloidal-sized particles of Ra-226 most probably coprecipitated with Ba and SO_4 following the extraction of the uranium in the milling process. The third mode is less common and is characterized by the association of relatively intense Ra-226 alpha activity with deposits of very fine red particles frequently attached to the surface of quartz grains. The identification of this mineral phase has not yet been established. The fourth mode is characterized by the presence of two-, three-, and four-track alpha stars whose vertices appear to lie on grain surfaces. These alpha stars are believed to result from the sequential decay of Ra-226 atoms and its immediate daughter products adsorbed on the surfaces of the quartz grains and other mineral fragments that make up the sample. This Ra-226 is believed to be much more weakly bound than the Ra-226 in the preceding three modes of occurrence. Closely related to this last mode of occurrence is the Ra-226 produced by the decay of its parent Th-230. The Th-230 has a very different mode of occurrence than the Ra-226 and appears to be adsorbed on aggregates of clay-like particles and other very fine-grain mineral particles. It is believed that these phases are the most likely sources of the mobile Ra-226 and Rn-222 observed in the samples.

It is suggested that a better understanding of the modes of occurrence of Ra-226 should contribute to new procedures that might minimize the further movement of Ra-226 and Rn-222 and aid in identifying those tailings piles which may present special health hazards because of the presence of intensely alpha-active Ra-226 particles in the colloidal to sub-micron size range.

INTRODUCTION

A nuclear-emulsion feasibility study[1] has been completed that includes modes of occurrence of Ra-226 and some of the other alpha-emitters in the U-238 decay chain in uranium mill tailings. Approximately 40 samples of drill core from the Carbonate, Vanadium, Acid, and East Tailings piles of the Monticello Mill, Monticello, Utah have been examined. The study was undertaken to determine if information on how these radionuclides were distributed, i.e., their modes of occurrence, their mineralogy and their relative mobility might make some contributions to the mill-tailings remedial-action program.

One hundred milligram splits of the drill-core samples were studied using the dispersed-grain, nuclear-emulsion technique[2]. This technique is a micro-autoradiographic procedure that permits the direct observation of the alpha activity of individual mineral grains or particles. The minimum particle size that can be studied is set by the resolving power of the optical system used. The technique employs a special alpha-sensitive photographic emulsion that records the passage of those energetic alpha particles, emitted during the radioactive decay process, that enter the emulsion. The trajectories of these alpha particles in the photographic emulsion usually appear, after exposure and development of the emulsion, as a straight chain of submicron-sized, closely-spaced silver grains. These linear arrays of silver grains or alpha tracks are examined under a microscope at a magnification between 200 and 1000

times. Such nuclear-emulsion techniques are remarkedly sensitive. This sensitivity is a consequence of the very low natural alpha-track backgrounds in the emulsions (essentially zero), the very high alpha-particle detection efficiency of the emulsions (essentially 100 percent), and the fact that exposures of many months can be used to ensure adequate alpha-track populations even though the radionuclide concentrations are very low.

The observational data consist of estimates of the populations of single alpha tracks and of the three- four- and five-branched alpha-star counts (per unit area), estimates of track lengths, and descriptions of the alpha activity associated with different mineral constituents of the sample. Using data of this type, inferences may be drawn on the mobility and modes of occurrence of such radionuclides as U-238, Th-230, Th-228, R-226, Rn-222 and Pb-210/Po-210.

In general, single alpha tracks are produced by U-238, U-235, Th-232, and their long-lived daughter products U-234, Th-230, Pa-231, and under certain circumstances by Pb-210/Po-210. Alpha-track stars are formed by the sequential decay of single atoms of such radionuclides in the Th-232 and U-238 decay chains as Th-228, Ra-226 and Rn-222 whose daughters are relatively short-lived. These atoms produce three-, four-, and five-branched stars, respectively, and the great majority of these stars are characterized by the property that the alpha tracks diverge from a common vertex or point of origin. From this observation it would appear that, at least in nuclear emulsions, the process of recoil and diffusion do not play significant roles in the movement and separation of the parent and its daughter products. Ra-226 and Pb-210/Po-210 have also been observed as intensely alpha-active radiocolloids and have been tentatively identified in chemical combination with such compounds as $(Ra,Ba)SO_4$, $(Ra,Ca)SO_4$, $(Pb,Pb\text{-}210)SO_4$ and $(Pb,Pb\text{-}210)CO_3$. These precipitates and mineral particles are also intensely alpha-active and usually are associated with hundreds to thousands of alpha tracks.

PREVIOUS STUDIES

The dispersed-grain, nuclear-emulsion technique proposed for use in this feasibility study was originally developed to identify the alpha-active mineral phases of Colorado Plateau uranium ores. The same technique was also used to describe, in a qualitative way and at the microscopic level, the equilibrium relationship between the long-lived alpha-emitting parents, U-238 and Th-232, and some of their alpha-emitting daughter products. It was clear from this early work that nuclear-emulsion techniques might provide descriptive and qualitative information that would be difficult, if not impossible, to obtain in any other way. In addition, the sensitivity of the dispersed grain technique was such that, under favorable circumstances, it is possible to uniquely associate an alpha track with a source particle less than a micron in size and, as has been mentioned, to identify the decay of single atoms of Th-228, Ra-226, and Rn-222.

Although the potential for use of the dispersed-grain nuclear-emulsion technique in the study of uranium-mill tailings seems promising, it does not appear to have been used. A search of both the Department of Energy (DOE) Energy Database and the GEOREF Database failed to produce any references to nuclear-emulsion studies on uranium-mill tailings since 1952. The only related geologic investigations recently reported in the literature have involved nuclear-emulsion studies of barren and mineralized drill core from uranium ore deposits in the Red Desert and Copper Mountain Test Sites, Wyoming (A Department of Energy/Bendix Field Engineering Corporation test facility). A continuing search of the literature has yielded only one additional nuclear-emulsion reference[4], a study of the radioactive hot spring mineral hokutolite.

A critical review of the extensive literature dealing with uranium-mill tailings which does not involve the use of nuclear-emulsion techniques has been prepared by E. Landa[5]. This comprehensive survey emphasizes the geologic and geochemical processes affecting the long term containment of uranium and its radioactive daughter products. The survey also identifies several areas where it has been necessary to approach fundamental questions concerning the distribution, localization, and mobility of the radionuclides in uranium mill tailings by indirect methods such as leaching. In these areas, it would appear that nuclear-emulsion techniques might be able to offer some insights based on direct, unequivocable observations.

For example, in the critical areas of the modes of occurrence of radium in uranium ores and uranium-mill tailings, Landa[5] reviewed and tabulated the leaching studies of a number of investigators including Shearer[6], Havlik and others[7,8] Borrowman and Brooks[9], Seeley[10], Moffett[11], Ryon and others[12], and Skeaff[13]. These indirect studies are not all mutually supporting and suggest that both adsorption and coprecipitation reactions may be involved in the retention of radium by mill tailings. Nuclear-emulsion studies should provide direct observational data on the relative importance of these two processes.

In addition, there appears to be some disagreement on the relative importance of the roles of $BaSO_4$ (Shearer and Lee[14]), $CaSO_4$ and $SrSO_4$ (Seeley[10], Ryon and others[12], and Kaiman[15,16]) and the group of hydrous iron-sulfate minerals including jarosite (Kaiman[15,16]). The immediate stability of radium in the mill-tailings environment is clearly influenced by the relative importance of the processes of adsorption and coprecipitation at the time the tailings pile was formed. It also seems likely that the long-term stability of radium within the pile will be influenced by surface desorption processes as well as by the differences in solubility of the host minerals with which the radium may be chemically combined. Nuclear-emulsion techniques, particularly when combined with other techniques which will yield positive mineral identifications, should be able to contribute in a direct way to the resolution of some of these important issues.

Control of radon emanation from the surface of a uranium mill-tailings pile is also an important objective of the Remedial Action Program. In view of the fact that the emanation of radon is a function, at least in part, of the specific chemical compound with which the radium is coprecipitated (Hahn[17]), it would seem once again that nuclear-emulsion studies of mill tailings

might provide direct information not readily obtainable in any other way. Intuitively, it would seem that Ra-226 atoms adsorbed, for example, on the surface of a quartz grain would, following their decay to Rn-222, be more easily lost that an atom of Ra-226 chemically coprecipitated with Ba and SO_4, a chemical form which has been shown to have a very low emanating power (Hahn[17]). The ability to characterize the different modes of occurrence of Ra-226 in mill-tailings samples should lead to a better understanding of the processes involved in the emanation and transport of Rn-222 in or near the surface of a pile and ultimately, its loss to the atmosphere. To the extent that such losses of Rn-222 to the atmosphere are affected by the mineralogy of the radium in the tailings pile, information of this type might lead to development of specific chemical procedures for minimizing radon emanation.

Finally, in addition to providing basic information on the modes of occurrence of Ra-226 in uranium-mill-tailings samples and estimates of the relative mobility of Ra-226 and Rn-222 in such samples, it is possible that nuclear-emulsion studies of this type may also contribute to the Remedial Action Program by providing a unique and direct means of observing the alpha activity of individual radium-bearing particles. It should be possible to study that population of radioactive particles in the size range usually retained in the lung and thus of special concern to health officials. The movement of tailings by the wind has been treated by Breslin and Glauberman[18] and, more recently, by Dressen[19]. It would seem that the information obtained from such nuclear-emulsion studies on alpha-active particulates might be of specific use in assessing some of the health hazards associated with this aspect of uranium-mill tailings.

THE MODES OF OCCURRENCE OF Ra-226, Rn-222, Po-218 AND Po-214

The alpha activity of Ra-226, and Rn-222, and especially its short-lived daughter products Po-218 and Po-214, is characterized by (1) the long length of the polonium alpha tracks (alpha-particle energies 6.0 and 7.7 MeV, respectively, compared to 4.3 MeV for U-238 and 4.7 MeV for U-234, Th-230 and Ra-226; (2) the Rn-222 and Ra-226 sequential decay schemes which produce in the emulsion the three-track and the four-track stars, respectively (representing the decay of single atoms); and (3) the relatively large numbers of alpha tracks associated with even colloidal sized particles which may contain only tens of thousands of atoms of Ra-226 and daughter products. This alpha activity is a function of the relatively short half life of Ra-226 compared to its parent Th-230 (1600y and 2.4×10^5y, respectively) and to the relatively short half-lives of Ra-226's immediate daughter products.

As has been mentioned, most of the tracks in the three-track Rn-222 stars and the four-track Ra-226 stars are oriented, i.e., they appear to diverge from a common point of origin. These intact points of origin or star vertices suggest that, at least in the emulsion, neither the processes of recoil nor diffusion appear to play a significant role in the movement or migration of the Rn-222 atoms between the time of their formation and their decay (mean life approximately 5.5 days). Similarly, there does not appear to be any evidence during the exposure period of the alpha plate (usually a minimum of three months) of the recoil or diffusion of Rn-222 atoms (three-track stars) into the emulsion in the immediate vicinity of the numerous sources of intense Ra-226 concentrations (radiocolloids and radiobarites) found in most Monticello samples. These observations support earlier studies in the literature and suggest that the Ra-226 and Rn-222 atoms in the radiobarites and radiocolloids are quite tightly bound and that solution or desorption from grain surfaces rather than recoil is the most probable source of the many randomly distributed three- and four-track stars which diffused into the emulsion during the preparation of the alpha plates and whose vertices lie between the upper and lower surface of the emulsion.

There are four principal modes of occurrence of Ra-226 and its accompanying daughters Rn-222, Po-210, and Po-214 in the Monticello drill-core samples.

Radiobarites

The most significant mode of occurrence of Ra-226 in the Monticello drill core, with the possible exception of some of the samples from the Carbonate Pile, is the chemical combination of the radium with sulfate, most probably as barite, $(Ra,Ba)SO_4$ (Fig. 1).

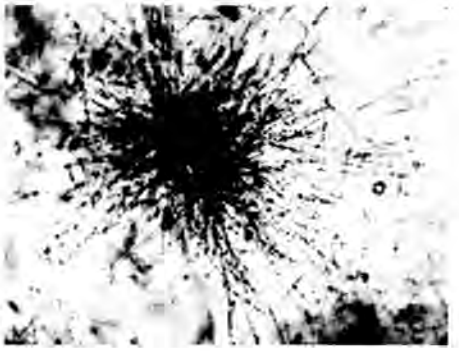

Fig. 1. A Photomicrograph of an Alpha-Active Barite Grain.

The tentative identification of these usually intensely alpha-active grains and particles as "radiobarites" is based on the birefringence of the grains as well as their high relief when viewed microscopically using an oil-immersion objective. The presence of the Ra-226 is confirmed both by the large number of alpha tracks that are associated with even very small particles (for long exposures almost all of the Ra-226 decays result in 4 alpha tracks) and by the preponderance of long tracks.

The size of the "radiobarites" in the tailings samples ranges from many tens of microns to fractions of a micron, the lower limit being set by the optical resolution of the microscope. There is a suggestion, based on track length, that the larger "radiobarites" have scavenged the Ra-226

from the spent mill liquors, possibly at the time the tailings were discharged to the piles, and that the radium is chemically combined with sulfate on grain surfaces. The smaller particles may be coprecipitates of radium with barium or other alkaline earth sulfates. The very large number of "radiobarites" in most tailings samples (many samples contain tens to hundreds of "radiobarites" per milligram) underscores the necessity of physically stabilizing the piles to prevent the wind-blown dispersal of alpha-active particles. If it is necessary to work in areas where the dust from the piles cannot be controlled, it may be prudent to take special precautions in order to minimize the inhalation of any alpha-active, radium-bearing particles but especially those particles in the size range usually retained in the lungs.

The fact that $(Ra,Ba)SO_4$ is chemically a very stable compound and that it is relatively insoluble even when the pH is low is of special significance when considering the long-term daughter-product stabilization of uranium mill-tailings piles. In addition, as has been noted, it is well documented in the literature that the Rn-222 emanation of $(Ra,Ba)SO_4$ is very low. These two properties of the sulfate compound suggest that if an excess of $BaSO_4$ is not present in the tailings, the addition of excess Ba or even barite, perhaps in a form similar to high density drillings muds, might be advisable. Recent experiments have reconfirmed that $BaSO_4$ precipitates are very effective scavangers of radium in solution. In addition, the presence of excess $BaSO_4$ should insure that the movement of the Ra-226 resulting from the decay of the widely distributed, adsorbed Th-230 would be held to a minimum. Finally, nuclear-emulsion studies of samples collected in the vicinity of a near surface, sandstone-type uranium deposit should provide a good basis for the evaluation of the long-term migration of uranium daughter products. Specifically, it should be possible to study in the adjacent sandstone the role of the clay minerals, detrital barite, as well as the interstitial barite cement in trapping mobile, unsupported Ra-226.

Radiocolloids

The second mode of occurrence of Ra-226 is much less common and is characterized by point sources of alpha activity, often intense, which do not appear to be associated with a visible grain or particle. The sources of this activity are believed to be colloidal sized particles of Ra-226 most probably coprecipitated with Ba and SO_4 (Fig. 2).

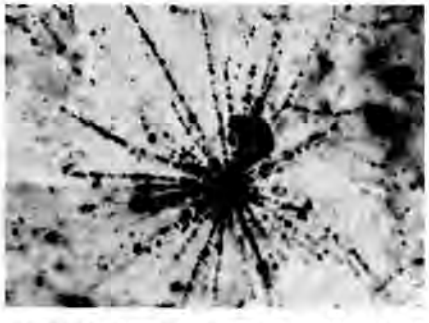

Fig. 2. A Photomicrograph of a Radiocolloid Containing Ra-226.

In this study these point sources of activity are called radiocolloids and are, by definition, those alpha-active particles which are so small that they cannot be optically resolved under a microscope at 1000 times magnification. In order that the alpha activity of the colloids can be unequivocably distinguished from the five-track Th-228 alpha stars, the lower limit for radiocolloids has been arbitrarily set at six alpha tracks. Radiocolloids grade imperceptibly in size into very small radiobarites and in many instances the distinction cannot be made because the source particle is obscured by the intense alpha-track activity. The smallest radiocolloid noted in this preliminary survey had eight tracks. Given an alpha-plate exposure of 118 days and assuming that, at a minimum, these tracks represented the decay of just two Ra-226 atoms (not a realistic assumption), it can be calculated that the colloid contained a minimum of approximately 14,400 Ra-226 atoms. This example suggests that useful information can be obtained on some types of particulate material even though the particle size is well below the resolving power of the microscope.

Mineral Coatings

The third mode of occurrence of Ra-226 is also much less common than the radiobarites and is typically characterized by the association of relatively intense alpha activity with deposits of very fine red particles frequently attached on the surface of quartz grains. (Fig. 3).

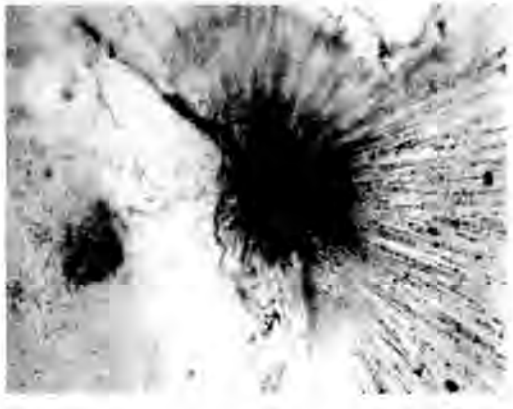

Fig. 3. A Photomicrograph of an Intensely Alpha-Active Coating on a Quartz Grain.

Unattached masses of fine-grained, alpha-active, red particles also have been observed. Under crossed polaroids these particles are birefringent and appear to be pink in color. The assignment of a chemical composition to this mineral phase has not been made and must await much more detailed mineralogic study.

Adsorbed Ra-226

The fourth mode is characterized by the presence of two-, three-, and four-track stars whose vertices appear to lie on grain surfaces (Fig. 4).

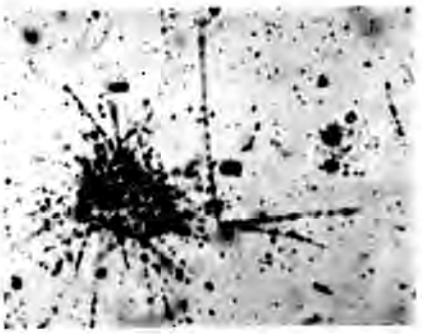

Fig. 4. A Photomicrograph of a Three-Track Alpha Star whose Vertex Lies on the Surface of a Small Quartz Fragment. The Adjacent Alpha-Active Grain is a Recent (?) Precipitate Containing Uranium.

Because of the fact that the nuclear emulsion records only the trajectories of those alpha particles which enter the hemisphere underlying the sample (i.e., the emulsion in this case has a 2π detection geometry), the probability, for example, that the trajectories of all four alpha particles from the sequential decay of a Ra-226 atom would fall within this hemisphere is not high. As a result, all two-track stars are interpreted as incomplete three-, four-, and five-track sequential decays. Similarly, three-track stars may be incomplete four- and five-track stars although a few of the three-track stars may, in fact, represent the full complement of a Rn-222 sequential decay.

The number of alpha stars that can clearly be associated with grain surfaces is relatively low compared to the alpha activity of the radiobarites and suggests that, with the exception of some samples from the Carbonate Tailings Pile where alpha stars appear to be more abundant, this mode of Ra-226 occurrence would appear to account for only a few percent of the total radium in the sample. The concentration level of this adsorbed radium seems consistent with the population of three- and four-track stars found in the emulsion. On the basis of this still tentative observation and the known relative insolubility of the radiobarites and radiocolloids, it is believed that the weakly-bound, adsorbed Rn-222 and Ra-226 on the grain surfaces is one of the more probable sources of the Rn-222 and the Ra-226 which enter the emulsion during the sample dispersal process.

Another possible source of relatively weakly-bound Ra-226 in the tailings is the Ra-226 produced by the decay of the chemically separated and unsupported Th-230. A significant fraction of the Th-230 originally in secular equilibrium with the uranium in the ore appears to be associated with masses of very small, birefrigent, clay-like particles often embedded in a clear to brownish matrix. The alpha activity associated with this type of material is characterized by mats of numerous, overlapping, randomly oriented, short, single tracks. The mats appear to be most abundant in samples from the East Tailings Pile, although they have been found in almost every tailings pile sample. If this Th-230 and the remaining U-238/U-234 are adsorbed by the clay in the sample, these atoms may be more firmly bound than their adsorbed counterparts on the quartz and feldspar grains. The Ra-226 produced by the decay of the Th-230 in this mode also may be less mobile than the Ra-226 adsorbed on the quartz and feldspar grains. What is suggested, however, is that the stabilization of the Ra-226 associated with this mode of occurrence of uranium and Th-230 presents a very different long-term problem than that presented by the Ra-226 originally present in the ore and chemically combined or coprecipitated with Ba and other alkaline earth-sulfate compounds.

CONCLUSIONS

This nuclear-emulsion feasibility study of the Monticello drill-core samples suggests that nuclear-emulsion techniques can provide a unique way of directly observing in uranium mill tailings the modes of occurrence of and the relative mobility of some of the different alpha-active radionuclides in the uranium and thorium decay chains. Even though this initial study was limited in scope and even though the identification of the associated alpha-active minerals could be only tentative at best, the qualitative, descriptive information on the localization of Ra-226 and Th-230 suggests that subsequent nuclear-emulsion studies combined with detailed mineralogical studies should be able to contribute in several important ways to the uranium-mill-tailings remedial-action program. These contributions might include:

1. Proposing specific remedial action based on the modes of occurrence of Ra-226 and Th-230.

2. Providing new techniques for assessing both the short-term and long-term effectiveness of certain remedial action.

3. Procedures for minimizing the further movements of Ra-226 and Rn-222 within the tailings pile.

4. Providing a means of identifying those tailings piles that may present special health hazards because they contain numerous, intensely alpha-active, Ra-226-bearing particles in the colloidal to submicron size range.

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ACKNOWLEDGEMENTS

This paper is based on work performed under the United States Department of Energy Contract DE-AC07-76GJ 01164 for the United States Department of Energy, Grand Junction Area Office, Idaho Operations, under Sub-contract 82-577-S for the Bendix Field Engineering Corporation.