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# **Impact of Actinoids on Environmental Chemistry**

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# ABSTRACT

Environmental actinide cleanup is commonly carried out due to a lack of understanding of the contaminated chemical forms. The levels of actinide contamination are especially important at the start and finish of the clean-up procedure. Final actinide concentrations based on expected dose or renegotiated levels of exposure are frequent at cleanup sites. Long-term behavior study and the development of new analytical instruments for identifying actinides in the environment require a thorough understanding of the chemical forms of actinides in the environment. Depending on the prevalent chemical form in the environment, actinide transport is influenced by solubility, colloid formation, or interactions with natural legends. This information can be utilized to research actinide fate and transportability. By learning about the most prevalent chemistries in the surrounding environment, it is feasible to develop analytical techniques that reduce counting error and improve statistics. Because counting errors are reduced, more certified garbage will be collected sooner and for less money. As a result, cleanup will be faster and expenditures will be lower. Actinide speciation is visible in both Am and Pu-contaminated and currently recovered sites. Because of the actinide speciation, cleanup tactics, regulatory responses, and costs have all been affected.

Keywords: Environmental actinide, chemical forms, solubility, colloid formation, Am and Pu-contaminated, cleanup tactics

# 1. INTRODUCTION

Cleanup and containment methods can only be predicted and devised with a solid grasp of actinide chemistry. When cations are present, four main processes occur: precipitation, complex reaction, adsorption, and colloid formation. This reduces actinide in solution and prevents its release and transport when a solid phase forms. Complexation is likely to increase release and migration rates. This slows the movement of actinides (Hugo Jaegler, 2021). These colloidal-sized particles can either facilitate or impede actinide movement depending on the colloids' composition and solution conditions. Depending on the actinide's oxidation state, four processes progress at varying rates. Absorbance spectroscopy can tell whether actinides are dissolved or colloidal in solution. This method can detect oxidized or complexed actinides. Scientists have developed ultrasensitive spectroscopes that use laser-induced photothermal and fluorescence spectroscopy to study actinides at a few parts per billion. With new fluorescence detectors and third-generation synchrotrons, X-ray absorption spectroscopy (XAS) is advancing environmental actinide speciation research (Talan, D. 2021).

As a result of uranium mining and nuclear testing, the reentry and disintegration of nuclear-powered satellites, reactor accidents and the storage and deconstruction of weapons-grade nuclear waste, activated actinides have leaked into the environment. Environmental effects on actinide properties and forms En aqueous solution, actinides interact with the surrounding geologic media, affecting their transport behavior. When assessing radiological risks, consider the volume and half-life of actinides produced by nuclear explosions and spent reactor fuel. Table 1 lists long-lived actinide isotopes' half-lives. These long-lived isotope actinides and their massive production are the main sources of current and future pollution (Jon plaue, 2002).

Polluted sites are being cleaned up to reduce actinide emissions into the atmosphere. The dose can be estimated using chemical sorption, distribution coefficient, and anticipated transport paths. While identifying the actinide chemical species present in the environment is critical, it is not currently the norm in actinide remediation. The Indian Air Force recently attempted to use actinide speciation into hazardous site remediation. The effects of actinide speciation were explored twice. There is a variety of material on cleanup. Actinide speciation impacts cleanup operations in two circumstances (Wolfgang Runde, 2000).

Radiation monitoring, testing, and verification are all part of comprehensive site remediation strategies. Regulators and other stakeholders are also involved. Final waste processing requires treatment and disposal of final waste.

In addition to risk assessors, the cleanup crew includes geologists and environmental engineers. The main goal is to determine the presence of pollutants and the immediate and long-term environmental conditions. Once contaminants of concern are identified, criteria, DCGLs, and evaluation methods can be defined. Calculate the DCGL using an isotope's predicted exposure pathway. Contamination sites may be opened to the public if their radioactivity levels meet standards. The maximum permitted dosages are determined by regulatory literature. For example, the NRC uses 0.25 msv/y to decommission nuclear reactors. Starting with criteria and measuring radionuclide transport is the first step (Morss, Lester R., 2006).

The next step is to assess the site's risk. Site-specific factors are emphasized in the dose-response study. The dose of each radionuclide is calculated individually. RESRAD can help us figure out how much medication to give the body. This method expresses radionuclide solubility as a concentration or distribution coefficient. Cleanup efforts usually use distribution coefficients. Actinide speciation is more reliable than distribution coefficients. In the planning of transportation routes, transfer factors and predicted travel patterns are considered. Actinide solubility in various environments is being

studied for dose assessment models. The annual doses of radionuclides are reflected in RESRAD output (Figure 1). The upper and lower dose limits for radionuclides can be compared (Harwood, 2007).

Then comes a characterization survey, this survey follows recognized norms. When examining the topic area, the DCGL was considered. Remedial action is now being taken Depending on the dosage, removing sediment or monitoring in-place are alternatives. Corrections are followed by dose and risk assessments.



# 2. RELATED WORK

Talan, D. and Huang, (2021). Thorium and uranium were the first actinides to be discovered. The metals actinium and prototactinium have been found in nature in very small quantities. Natural Actinides differ from man-made Actinides, which are all found in nature. Actinides are highly reactive, as evidenced by their low solubility in halide compounds, their instability in aqueous solutions, and their ease of precipitation from acidic and basic environments. Deniz Talan (2021). In this summary, it should be evident that actinides and their surrounding environments are tough to interact with. The particular environmental variables of each local ecosystem have an effect on the general movement and migration characteristics of adinide species. Norman M. et al. (2009). As part of our efforts to understand how these systems interact, we must study environmental chemistry, molecular science, and interfacial science to the fullest extent possible. When interpreting experimental results, it is necessary to take natural systems into account. Harwood, (2007). The aqueous phase stability and mobility of plutonium oxidation states are affected by the chemical conditions of the surrounding environment, regardless of whether Pu(VI) is present in a PuO2(s) matrix (water and rock compositions). Pu is unlikely to be stable in most aquatic environments (VI). For Pu(OH)4s in solution to precipitate as PuO2, the redox potential of the water must be strong enough to reduce their oxidation states (s). PuO2+x(s) and plutonium's solubility or mobility in the natural environment may be linked, however this is not always the case. Lester R., et al. (2006). It is becoming increasingly important to understand how actinides interact with the environment as stocks grow and containers age because of the increased risk of accidental leaks. More complex models are needed to account for all possible migration routes away from an actinide source. To build these models will be a challenge for theoretical and experimental scientists for many years. Cotton, Simon. (2006). The physical, chemical, and biological features of actinide elements, their unique compounds, and their ions in aqueous solutions are examined in greater detail in this paper than previously. These elements are distinct from those in the lanthanide and actinide transition series when compared to other 5f-block elements with similar characteristics. The electrical configurations, oxidation states, and thermo chemical data, crystal structures, and ionic radii of actinide elements have been studied. Morss, L. (2006). As part of this study, a flowchart was drawn up for the extraction of rare earths from coarse coal refuse. These potentially hazardous elements and rare earths were rigorously evaluated for their ability to separate from each other. While minimizing non-selective recovery of thorium and uranium into the product stream, several separation techniques, such as selective precipitation, solvent extraction, and adsorption, were used to extract rare earth elements. Each separation process was thoroughly examined in order to determine the possible use for various separation methods, and this was done through statistical and basic analysis of the test results.

## 2.1. Environmental reactions

A variety of elements must be taken into account when attempting to model or anticipate the release or transfer rates of actinides in watery settings. Processes such as precipitation of the actinide cation, complication, sorption, and the creation of colloids are all essential. If the concentration of solution actinide exceeds the solubility product constant required to form a solid phase, precipitation can occur. As less actinide is in solution near the solid phase, this effect delays release and migration. These reagents' legends, the actinide ions, can form complexes with both inorganic and organic compounds. Charges in solution complexes can vary widely.

# 2.2. CS-10 site (Pu and Am in Waste)

Samples of Pu and Am from Pokhran Site CS-10 were analyzed using an ion exchange, UV-Visible spectroscopy, alpha spectroscopy, mass spectroscopy, and NIST traceable standards as NIST standards. The inquiry focused on the materials' Pu chemical forms and Pu isotope compositions, which were examined (Harwood, 2007).

# **3. ANALYZE THE PROBLEM**

ICP-MS, a, 13, and y spectroscopy were used to determine the solution's isotopic composition. 99.5 percent of the samples had 239PU in them, according to the data gathered. There are 2.42n moles of 241Pu and 241Am in the solution. If the 241Am was not there when the Pu material was removed, the aged material had a 49.8-year-old history. There were  $[Pu] = 1.18 \pm 0.04 \times 10^{-4} \text{ M}$  and  $[Am] = 5.32 \pm 0.02 \times 10^{-7} \text{ M}$  in the answer (Norman M. Edelstein 2009).

Solution samples were analyzed using ion exchange and UV-Visible spectroscopy. To detect organic Pu species, one sample of the solution must be passed through a column containing XAD resin. The eluant's Pu activity fell little when it was tested. Analyzing the alpha activity of the XAD resin required dissolving a sample of the organic substance in 0.1 M NaOH. The solution contained  $9.4\pm3.5x104$  M of Pu organic species but no Am organic complex. The low solution pH is to blame for the absence of an Am organic complex. To create a new acid solution, a DOWEX anion exchange resin was added to the acidified 7.5M nitrate solution. This shows three-valent oxidation states. Pu was eluted with 0.1M. Nitric Acid instead of 2 M. Nitric Acid, leaving the Pu on the column after it was eluted, unlike the other method. Pu's oxidation state was likely quadruple tetravalent. The oxidation status was confirmed by an absorption peak at 470 nm in the UV-Visible spectrum. In solutions with a pH ranging from 6.0 to 7.0, Am can be found as Am3+. At pH 1.5, Pu4+ and Pu0H3+ are the dominant Pu species, and hydrolysis commences (Deniz Talan, 2021).

UV-Visible spectroscopy with 1-pheno1-2, 4-disulphonic acid was used to measure the solution's nitrate concentration. 410 nanometers is the maximum absorbance of the nitrophenoldisulphonic acid reaction in alkaline solution (nm). The nitrate concentration in the solution was determined to be 1.20.1 M using standards and a calibration curve. A search for NO3- in the solution was conducted based on previous laboratory separations at the area (Cotton, Simon. 2006).

In addition to radioactive materials, the solution is acidic and nitrate-heavy. Organic precipitation also covers the soil. Am3+ and Pu4+ and Pu0H3+ can be detected in solution during actinide speciation. Tests and calculations based on Pu hydrolysis constants were used to determine the Pu speciation. Pu behavior is heavily influenced by its immediate surroundings. Even if this material were to enter the atmosphere, it would be quickly degraded by the surrounding environment... Depending on their immediate environment, the Am and Pu could produce new chemical species. Hydroxide precipitation and subsequent precipitation would occur in the absence of significant complexing ligands. Inorganic hydroxides or colloids could then be formed as a result of the hydroxide.

#### 3.1. Solid Analysis

An airtight one-liter glass bottle was utilized to store the solid phase sample. The jar was divided into two sections: a metallic one and a nonmetallic one. A metallic phase, about 4 cm broad, was present. During the biological phase, there were a few metallic files present. There was nothing except thick and degraded oil remaining in the organic phase. In the solid state, 99.5 percent of the Pu is 239PU, just like in the solution phase. There were 0.129 g (0.539 mmoles) of total Pu and Am concentrations in the solid phase (2.42 micromoles). Only 0.003g of Pu 5.240,000 were found in the solid inorganic material, according to the testing. The metallic phase accounted for 26% of the system's overall mass. At Argonne National Laboratory, XAFS discovered that PuO2 is the atom's tetravalent state (Cotton, Simon. 2006).

The substance's density was estimated at 3 g/mL. Be, B, C, Al, Si, and Ca are all light elements with densities less than 3 g/mL. There is a good chance that this is the bulk of the content. Because of the lack of evidence for neutron emission, be was ruled out. Adding 5 M HC1 to the metallic phase dissolved it. The indicator dye Stilbazo bonded to the solution sample and appeared at 496 nm when it was further processed. This suggests that Al was the metallic phase. ICP-MS and ICP-AES both confirmed this. PuO2 made up 5% of the metallic phase of Al and was coated in aluminum oxide for the remainder (Morss, Lester R., 2006).

Prior to analysis, 7.5 M nitric acid was used to dissolve a sample of the organic phase. Two distinct phases were formed as a result of the organic phase's low density. The rest of the Am was found to be solid after it was diluted to  $94.2\pm5.3$  percent water. More than half of the Pu (61.3 - 3.2) was found in the aqueous phase. In this case, it's possible that the extractant has degraded the organic substance. Extractants containing triisooctylamine and/or tributylphosphate are effective at removing Am from 7.5 M nitric acid, but Pu is not removed. The organic substance may have used as an extractant in the purification process of Pu. The presence of phosphorus in the organic substance was detected using ICP-AES, indicating that tributylphosphate was employed.

#### 3.2. The Effect of Speciation on the CS-10 Site

Environmentally friendly behavior is determined by the type of rubbish that Pu sees on a regular basis. Pu oxide species in the Al matrix represent a low risk to the environment. Acidic or neutral solutions can be used to free the organic compound from Pu and Am (Morss, Lester R., 2006).

If the container explodes, Pu and Am could be released. Solution based on species has best likelihood of transferring Pu. The pH will rise rapidly if the solution is in close proximity to the user. Pu can precipitate and bind with environmental ligands because of its tetravalent nature. Colloids are expected to form in systems that are oversaturated with tetravalent Pu. If the typical distribution coefficients for pu are utilized, the concentration of Pu in the solution will rise (Morss, L. R., 2006).

The waste form speciation includes Pu environmental chemistry components that aren't seen when utilizing distribution coefficients to analyze Pu solubility for RESRAD input. Much more of the dosage is composed of the acidic aqueous solution than the organic solid phase. It is predicted that the Pu dose will be larger than expected if acid solution spills from waste form into the near field. Dose estimation can be made safer by using projected chemical forms as inputs, according to this study.

## 3.3. POKHRAN Site

Located in the Jaisalmer district of Rajasthan, Pokhran is both a hamlet and a municipality. Ballistic missiles, India's first nuclear weapons, were put to the test in the Thar Desert at the time. Data shows that the carbonate phase is capable of dissolving approximately 15% of the overall activity." Using the solubility constants for plutonium and americium carbonate species can help limit the amount of activity that can be dissolved. There was a significant amount of action in the insoluble portion of the drug. This heat- and acid-resistant substance could be used to make resilient actinide oxides. Rather than ashing samples, we recommend against it going forward due to the potential influence on speciation and dose assessment models.

Actinides from a nuclear weapons material incident damaged soil at India's nuclear Pokhran site. Dose assessment programmes used the ambient speciation as an input to determine the dose from actinides. The actinide fractionation in environmental medium was studied using sequential extraction techniques. As a starting point, NIST's traditional sequential extraction method will be used. Am and Pu concentrations in the environment will also be predicted using the chemical equilibrium and speciation using surfaces (CHESS) model. A major focus of CHESS is the chemistry of metal ions and colloids in the matrix. The CHESS double layer theory, which potentially explain it, relies on surface complexation as a key component. CHESS is heavily reliant on data gleaned from scholarly articles that have been vetted by their peers. More than 50 redox pairs live here, along with 700 species and 600 minerals (Gorden, A. E. V., 2003).

# 3.4. Analysis of Sediment

Alpha activity in the Pokhran sediment is closer to 2 Bq/g, with an alpha activity less than 30 Bq/g. Alpha-total alpha activity accounts for more than half of the overall alpha activity in 239p11/241AM (Harwood, 2007).

The U concentration in the sediment samples was 42 ng/g. ICP-MS, which was improved, determined that the mass ratio of 238U/235U was 1:1. A pre-heating procedure of about 450°C provided consistent samples for counting. It's possible that the removal of the sediment's natural organic makeup influenced the speciation of Pu and Am.

### 3.5. Method of sequential extraction

IAF soil samples were transported in plastic nalgene vials. The samples were weighed and then counted for 12 hours on 2g Canberra germanium gamma detectors. The concentration of 241Am was determined using a NIST traceable reference source and the spectrum analysis software Genie 2000.

S.No.	Phase	Solution	
1.	exchangeable	$0.4 M MgCL_2, pH = 4.5$	
2.	Carbonate	1.0M NH <sub>4</sub> Ac in 25% HAc,	
		pH = 4.0	
3.	Fe oxides	0.04M, NH <sub>2</sub> OH-HCL in	
		25% HAc, pH = 2.0	
4.	Residual	Conc. HNO <sub>3</sub> /HF	

Table.1. Conditions for sequential extraction

The extraction reagents, all of which were synthesized from scratch, are listed in Table 1. For a total treatment volume of 30mL, the specimens were centrifuged with 15mL of Phase 1 solution (0.4 M MgC1 pH 4.5). At 250 revolutions per minute, the centrifuge t ubes were manually shaken on a shaker table at room temperature. After being centrifuged for 10-15 minutes, the remaining liquid was filtered using 0.45-micron Gelman filters. Each sample was washed with 15 mL of DI water and centrifuged again in order to repeat the centrifuge operation. For testing purposes, rinse water was maintained in separate containers. A single technique was used to treat the carbonated extraction and the Fe oxide phases in this work (Morss, Lester R., 2006).

Because the NOM was removed during sample preparation, organic phase extraction was not required. During the extraction of the residue, the leftover soil matrix was digested by acid. In order to move the samples from the heated flask to the beaker, temperature-controlled plates were used. 15 mL HNO3 and 10 mL HF were the first acid quantities employed. A further 10mL of each acid was

added to the mixture over the course of three hours. This mixture was centrifuged for 15 minutes before the components were redissolved in 25mL of boric acid-saturated 50% HNO3. The collected supernatant will be used in future studies (Norman M. Edelstein 2009).

The gamma analysis was carried out on the residue that had been rinsed away." Various radio spectrometry methods were used to study the extraction process. Liquid scintillation counting was utilized to determine the total alpha activity of each sample by using 500 cc of each sample and 10 mL of Ultima Gold AB cocktail. 241Am concentrations were determined by doing a 12-hour radioactivity measurement on 10mL of each phase. After evaporation and electroplating, samples for alpha spectroscopy were monitored for 12 hours to determine their activity. To evaluate the solution's phases, ICP-MS was also used.

## 3.6. Geochemical Simulations

CHESS was used to analyze plutonium and americium samples found near the Pokhran nuclear facility. Table 2 illustrates the chemical composition of river water as well as the actinide concentrations that were used as input data for our inquiry (Deniz Talan, 2021).

S.No.	Elements	Concentration (ppm)
1.	$Na^+$	2.62
2.	$\mathbf{K}^+$	0.72
3.	$Mg^{2+}$	0.61
4.	Ca <sup>2+</sup>	1.10
5.	$\mathrm{Cl}^+$	5.93
6.	$SO_4^{2+}$	5.91
7.	$PO_4^3$	0.043
8.	SiO <sub>2</sub>	3.98

Table.2. The water composition of the Indian Rivers

Environmental speciation modeling was made easier with the help of the data in Table 2. Figure 2 depicts the outcomes of the CHESS simulation. There could be as many as nine different species in the given conditions, according to the computer's predictions. It is reasonable to expect that the free americium ion (Am3+) concentration will be at its peak at a pH of 4.5, which is about average for the area. Plutonium is unlikely to be soluble above a pH of 2.5. It seems unlikely that either of these actinides will be found in considerable concentrations. Solubility calculations can be made based on this information (Hugo Jaegler, 2021).



Figure.2. Indian River water composition diagram created with CHESS

# 4. RESULTS OF SEQUENTIAL EXTRACTION

To determine the quantity of 241Am present in each sample, a 12-hour gamma count was performed on each. It was then possible to calculate the 241 Am/total Pu ratio. Prior to this step, the soil samples had been treated to several extraction reagents, as had been indicated previously. Using liquid scintillation, we were able to accurately count each extract. The data was cleansed of reagent blanks. In each phase, the percentage of total alpha

activity that occurred was calculated (Figure 3). The carbonate and acid-soluble phases appear to harbor the majority of alpha activity, according to these findings (Harwood, 2007).



Figure.3. Extractable alpha activity distribution

To determine the appropriate 241Am/239PU ratio, alpha spectroscopy was performed on each extract in order to identify the best result. A distribution of isotope activity was calculated based on the information provided (Figure 4). Despite the fact that it appears to have a significant amount of activity left over, this residual phase does not appear to be acid-proof (Morss, Lester R., 2006).



Figure.4. The distribution of Am and Pu in sodium

# 4.1. Speciation Results and the Pokhran Test

It has been suggested that other sites' actinide carbonate dosages should be modeled and assessed using RESRAD in light of these findings Solubility ranges from 104 to 10-5 mol/L depending on the species. Based on the findings of these assays, the soluble actinides in these ashed samples are estimated to be 15%. As a result, fewer cleanups may be required, saving money in the long run (Morss, Lester R., 2006).

However, ashing must be taken into account. A better knowledge of actinide environmental behavior can be gained by studying native, unashed soils. The high-temperature ashing method can be used to alter species in a variety of ways. The primary advantage of this therapy is that it completely removes all biological material from the surface. Environmental migration of actinides, which is assisted by humic and fulvic acids that account for less than 5% of total mass, is supported.

The chemical form of trace actinide components may change throughout the ashing process. Metal oxides are more likely to develop at higher temperatures. High-fired chemically resistant actinide oxides are becoming increasingly prevalent. The considerable amount of activity that remained after the acid digestion confirmed the presence of these oxides. Measuring the actinides in oxide form is important to get a more accurate reading on the soil (Deniz Talan, 2021).

# 5. CONCLUSIONS

Assessment codes can be altered by using actinide speciation. It is possible to improve both safety and efficiency by speculating actinides. If the isotope exposure path changes, the remediation technique should take actinide speciation into account. An actinide solution's solubility and concentration can be determined by using specifications. Input codes for dose assessment like RESRAD can be made with this information.

Surveys and monitoring that rely on species identification are a boon. Speciation can improve methods for detecting the concentration of actinides in sediment. The existence of a known fraction of actinides in a sediment phase allows extraction and detection techniques to be customized to that phase. Remove actinides from the sample to lower detection limits and reduce experimental error if the organic sample has a substantial actinide phase. In order to reduce sample processing expenses, more samples can be processed in a shorter period of time.

Identification of species is a critical step in remediation planning. Modeling, tests in the lab and direct observation are only a few of the methods available for conducting research. ICP-AES and other commercially accessible technologies should be included. XANES and EXAFS are two cuttingedge research approaches that should be employed to improve remediation efforts during the survey phase. The most efficient strategy to remove actinides from the environment is to unite and utilize all current efforts.

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