Applied Research Center
FLORIDA INTERNATIONAL UNIVERSITY

U.S. DEPARTMENT OF ENERGY

ENVIRONMENT & ENERGY / ENVIRONMENTAL REMEDIATION

PROJECT: Rapid Deployment of Engineered Solutions to Environmental Problems: *Sequestering Uranium at the Hanford 200 Area Vadose Zone by In Situ Subsurface pH Manipulation Using NH_3 Gas*

CLIENT: U.S. Department of Energy
PRINCIPAL INVESTIGATOR: Dr. Leonel Lagos
LOCATION: Hanford Site, WA

Description:

FIU's Applied Research Center (ARC) is supporting the U.S. Department of Energy's Hanford Site in developing a strategy to improve the efficiency of the uranium stabilization process through pH manipulation using NH_3 gas injection.

Characterization of vadose zone (VZ) soil at the 200 Area has identified a number of radiological and hazardous contaminants, including technetium and uranium. This work is focused on long-lived uranium contamination, which is one of the key contaminants of concern that needs to be reduced to below levels that can cause harm to human health and the environment. Injection of reactive gases such as NH_3, is an innovative remediation technology shown to mitigate uranium contamination in soil.

The injection of NH_3 gas causes ammonia gas dissolution in soil moisture, with the formation of NH_4OH and a subsequent increase in pH. This manipulation will significantly alter the pore water chemistry and affect the dissolution of silica and aluminosilicate from soil minerals, followed by co-precipitation of U(VI) [as uranyl (UO_2^{2+})] and Al at higher pH conditions.

The main objective of the project is to evaluate the stability of the U-bearing precipitates created in the soil as a result of ammonia gas remedial actions. This information would help to accurately predict the mobility of U(VI) in the post-treated vadose zone soil.

Isopiestic measurements are the most appropriate to quantify mineral solubility for the unsaturated vadose zone conditions. This method is considered very accurate, helping to make more realistic predictions of contaminant fate and transport in vadose zone environments.

The isopiestic apparatus used for the experiments was fabricated from a pressure pot and contained an aluminum heat-transfer block that has a good thermal conductivity, able to maintain a uniform temperature distribution inside the chamber

Isopiestic chamber to conduct solubility experiments; aluminum block with holes to hold nickel crucibles

The main objectives are to:

- Examine the effect of concentration ratios of silicon to aluminum on the removal process of U(VI) in the presence of various bicarbonate and calcium ion concentrations.
- Evaluate the role of major pore water constituents such as Al, Si, bicarbonate and Ca on the formation of precipitates.
- Analyze mineralogical and morphological characteristics of precipitates by means of XRD and SEM-EDS to identify uranium-bearing solid phases.
- Examine the deliquescence behavior of multicomponent solids prepared from synthetic pore water solutions mimicking conditions at the Hanford Site.
- Examine the effect of temperature on the solubility of U(VI)-bearing precipitates.
- Determine the osmotic coefficient for multicomponent samples as a function of water activities, a_w, using $CaCl_2$ and LiCl as a standards.

ABOUT

Since 1995, the Applied Research Center (ARC) at Florida International University (FIU) has provided critical support to the Department of Energy's Office of Environmental Management (DOE-EM) mission of accelerated risk reduction and cleanup of the environmental legacy of the nation's nuclear weapons program. ARC's applied research is performed under the DOE-FIU Cooperative Agreement (under Contract # DE-EM0000598) and provides technical support to DOE EM in the area of environmental remediation and STEM workforce development and training.

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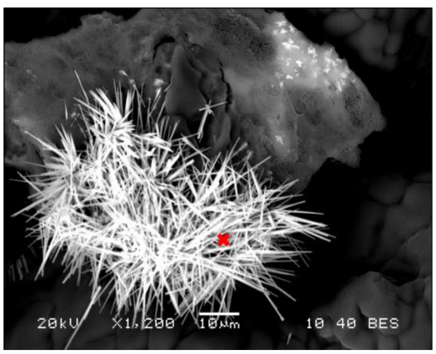
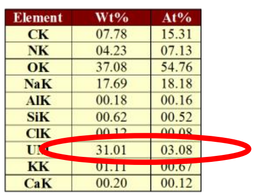
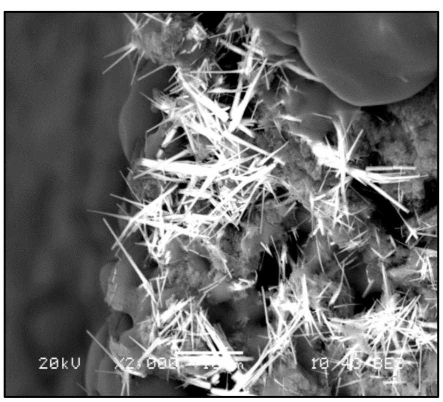
Benefits:

- Understanding the role of pore water constituents in the removal of U(VI) to predict the mineralogical changes and the formation of precipitates that would be created in the treated VZ soil.
- Yields information on the formation and solubility behavior of U(VI)-bearing precipitates under environmentally relevant Hanford Site VZ conditions.
- Attempts to investigate solid-liquid transitions of the synthetic multicomponent precipitate samples prepared from synthetic pore water solutions mimicking conditions at the Hanford Site.
- Evaluates the composition of U-bearing precipitates and relates them to the expected uranium species via solid phase characterization using XRD and SEM/EDS.





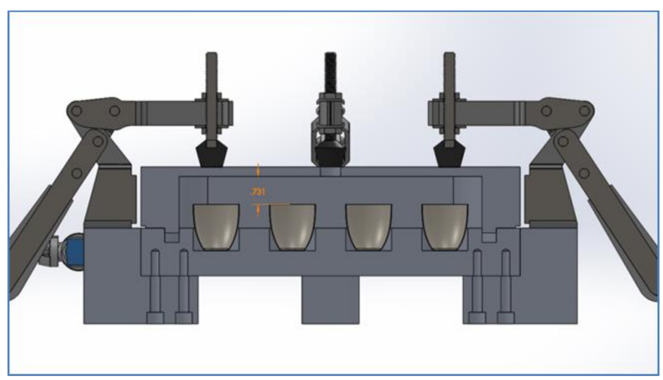
Element	Wt%	At%
CK	07.78	15.31
NK	04.23	07.13
OK	37.08	54.76
NaK	17.69	18.18
AlK	00.18	00.16
SiK	00.62	00.52
ClK	00.12	00.08
U	31.01	03.08
KK	01.11	00.67
CaK	00.20	00.12



Backscatter SEM images of samples showing the U-rich crystal-like forms

Accomplishments:

- Evaluated the effect of various silicon, aluminum, calcium and bicarbonate concentrations on the removal of uranium from the supernatant solution.
- Results have shown that the formation of Si gel always correlated with the removal of U(VI). If no Si polymerization and gel formation was observed, there was no U removal from the supernatant solution.
- Employed geochemical modeling software (Geochemist's Workbench & Visual Minteq) for speciation predictions and reaction modeling to supplement the characterization study.
- Initiated characterization studies of uranium phase using a range of analytical techniques including x-ray diffraction (XRD), electron probe microanalysis (EPMA), and transmission electron microscopy (TEM), and scanning electron microscopy with energy dispersive spectroscopy (SEM w/EDS).
- Initiated isopiestic measurements to investigate solid-liquid transition of no- uranium synthetic multicomponent precipitates combined from major pore water constituencies such as Na^+, SiO_3^-, Al^+, NO_3^-, K^+, HCO_3^-, Ca^{2+}, and Cl^- .
- Fabricated a new chamber to study the deliquescence behavior of U-bearing solids.



The full assembly of the isopiestic chamber with crucibles inside