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## **COLLOID GENERATION FROM METALLIC URANIUM FUEL**

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### **Abstract**

The possibility of colloid generation from spent fuel in an unsaturated environment has significant implications for storage of these fuels in the proposed repository at Yucca Mountain. Because colloids can act as a transport medium for sparingly soluble radionuclides, it might be possible for colloid-associated radionuclides to migrate large distances underground and present a human health concern. This study examines the nature of colloidal materials produced during corrosion of metallic uranium fuel in simulated groundwater at elevated temperature in an unsaturated environment. Colloidal analyses of the leachates from these corrosion tests were performed using dynamic light scattering and transmission electron microscopy. Results from both techniques indicate a bimodal distribution of small discrete particles and aggregates of the small particles. The average diameters of the small, discrete colloids are ~3-12 nm, and the large aggregates have average diameters of ~100-200 nm. X-ray diffraction of the solids from these tests indicates a mineral composition of uranium oxide or uranium oxy-hydroxide.

## Introduction

During the aqueous corrosion of uranium metal in an oxygenated atmosphere, uranium dioxide, hydrogen, various hydrated uranium products, higher valence uranium oxides, and uranium complexes (1-3) are commonly formed. The corrosion rate of uranium in water is rapid and dependent upon the solution conditions (temperature, atmospheric oxygen pressure, condition of original material, complexants, etc.). While studies on the corrosion behavior of uranium and solid corrosion products have been extensive (1-3), little work has focused on the colloidal corrosion products of uranium. The colloids formed during the corrosion of unirradiated uranium metal in a groundwater solution are the focus of this work.

Colloids have the potential to transport radioactive contaminants from a waste storage or disposal site and thus are extremely important to site evaluation activities for the proposed waste repository at Yucca Mountain. Sparingly soluble radionuclides associated with colloids can be present in solution at concentrations that substantially exceed the solubility of the radionuclide and can be transported greater distances than predicted based on solubility in groundwater. The migration of measurable concentrations of radionuclides in the groundwater systems from various waste sites has been underestimated by models that neglect colloidal transport (4). Recent studies at the Nevada Test Site have shown that over a 30 year period, plutonium associated with colloids composed of clays, zeolites, and cristobalite was transported 1.3 km from its origin (5). Thus, gaining an understanding of colloid formation and stability processes is essential for predicting source term release.

Previous work performed for the Yucca Mountain Program (YMP) on corrosion of high-level waste glasses demonstrated the generation of significant quantities of actinide-bearing colloids (6-7). This behavior was also observed with European waste glasses (8). While unsaturated tests of commercial spent fuel at Argonne have produced smaller quantities of colloids than similar tests with glass, colloids have been identified (9) in the test leachate. Moreover, the composition of some of the spent fuel colloids is similar to that of the clay colloids in the glass corrosion tests.

## Experimental

### Metallic Uranium Fuel

The metallic uranium fuel studied in this program is from an unirradiated Mark IV-E fuel element. Mark-IV fuel elements were fabricated as two concentric tubes of low-enriched (0.947%  $^{235}\text{U}$ ) metallic uranium alloy co-extruded in Zircaloy-2 cladding. The cladding was removed from the fuel, and the fuel was cut into coupons of 0.42 in. x 0.25 in. x 0.052 in. The coupon surface was polished to 600 grit finish, then cleaned and placed into the test vessel.

## Unsaturated Tests

The fuel holder, shown in Figure 1, was custom fabricated and consisted of three vertically stacked chambers. Each chamber has a metallic filter at its base to trap particles but allows passage of colloidal and dissolved species. The top chamber (Stage 1) has a 0.6-mm aperture gold mesh as its base; the middle chamber (Stage 2) has a 0.25-mm aperture gold mesh; and the bottom chamber (Stage 3) has 10  $\mu\text{m}$  holes in a Zircaloy-4 base. The assembled fuel holder rests on a ledge inside the stainless steel vessel in a configuration that allows injected water to directly contact the fuel sample. Solid corrosion products spalled from the fuel surface are size-fractionated and retained in the three chambers, while dissolved and colloidal species collect in the vessel base.

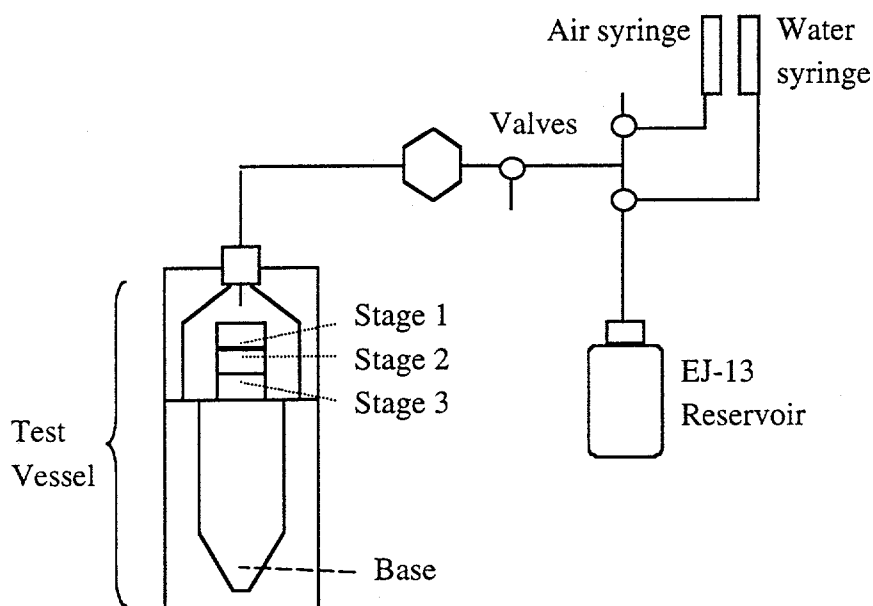


Figure 1. Unsaturated test vessel with three-stage fuel holder and injection system.

Unsaturated tests were conducted at 90°C with a water injection rate of 1.5 mL/week. The tests were performed by injecting tuff-equilibrated groundwater (EJ-13) onto a uranium sample placed inside a stainless steel vessel. The groundwater contacted the uranium fuel sample, flowed downward through a series of metallic filters, and collected in the base of the vessel.

The formation of colloids in the test leachates from the N-Reactor fuel was studied by DLS as a function of test duration. The test leachant injected onto the fuel was EJ-13 with a pH of 8 and major components,  $\text{HCO}_3^-$ , Na, and Si, at concentrations of 100, 54, and 45 mg/L, respectively. Sampling of the test leachates was done at 12, 28, 48, and 77 days to monitor the progression of the fuel corrosion. At each sampling, aliquots were taken from the base of the test vessel and from the stage 3 holder at the 12 day sampling (solution was not available from the other holders at other samplings).

On test days 12, 28, 48, and 77, the test was interrupted, and all the solution was removed from each chamber (if possible) as well as from the vessel base. On day 77, none of the uranium test coupon remained, so the test was terminated. Samples were analyzed to determine elemental content, pH, colloid composition, and mineral phase composition.

### Dynamic Light Scattering

Dynamic light scattering (DLS) is used extensively to probe the dynamics of colloidal particle motion. The principles governing the application of light scattering to study many systems are described in Chu (10) and Berne (11). The dynamic light scattering apparatus used in this study consists of a modified 4700c Photon Correlation Spectrometer from Malvern Instruments Ltd. The spectrometer has a Uniphase (variable power output of 3 to 75 mW) argon ion laser. All optical components of the system are mounted on an enclosed optical table. The correlator records the scattering intensity from small scattering volumes in time intervals as short as 50 ns. Particle size calibration was performed with a series of NIST-traceable polystyrene latex standards (nominal sizes of 30, 70, and 300 nm) from Duke Scientific Corp. Standard solutions were prepared according to certification instructions using filtered deionized water. Dynamic light scattering measurements of the polystyrene standards exhibited narrow size distributions, which were in excellent agreement with the manufacturer's specifications.

Unfiltered sample aliquots of approximately 500  $\mu\text{L}$  were taken for DLS measurements and placed in a temperature-controlled water bath (25°C). Laser power was varied from 3 to 25 mW with measurement durations of up to 5 min. The fundamental sample time ( $\tau$ ) or time window is dependent on the size of the colloidal particles present in each sample (10, 11). The colloids examined from leachates in this study had a fundamental sample time of 10 to 20  $\mu\text{s}$ , and the intensity autocorrelation functions were collected at a scattering angle of 90°. Data analysis was performed using the commercial software routine Contin (12-14), which involves LaPlace inversion of the correlation function. Contin is a well-known and powerful program for the analysis of photon correlation spectroscopy (or DLS) data. This program makes use of a constrained regularization method which is model independent to avoid the bias of fixed form models.

### Transmission Electron Microscopy

Transmission electron microscopy (TEM) is an excellent method for determining morphology, structure, and elemental composition (by energy dispersive x-ray spectroscopy (EDS), electron diffraction (ED), and electron energy loss spectroscopy (EELS)) of small, electron transparent particles. A 5  $\mu\text{L}$  aliquot is filtered through a microporous carbon grid to directly observe the colloids using TEM. The grid is analyzed with a JEOL 2000FX II transmission electron microscope equipped with two Noran energy dispersive X-ray fluorescence (EDS) detectors, plus an electron energy loss spectrometer (EELS) with a Gatan Imaging Filter. Identification of crystalline materials with TEM is often aided by electron diffraction (ED).

## X-Ray Diffraction

Sludge samples were analyzed by x-ray powder diffraction (XRD) using a Rigaku Miniflex diffractometer. Crystalline silicon was used as the calibration standard. Phases were identified by comparison with JCPDS-ICDD files. Owing to the small crystal coherence size in the  $\text{UO}_2$  sludge, substantial XRD broadening was observed for some phases. The peak search reports were generated by the program *Jade*, version 5, which estimates the peak width (full width at half maximum, FWHM) by the empirical formula  $0.9A/(I_p - I_b)$ , where  $A$  is the peak area,  $I_p$  is the peak maximum intensity, and  $I_b$  is the extrapolated background intensity. Each value of  $B(2\theta)$  must be corrected for instrument resolution by subtracting the FWHM of a peak from a known large crystal (for instance, the Si standard) at a nearby angle  $\theta$  in quadrature:

$$B(2\theta) = \sqrt{[FWHM(\theta)]^2 - [FWHM_{Si}(\theta)]^2} \quad (1)$$

Diffraction broadening originating from a particle size range is determined by a standard method (15):

$$B(2\theta) = \frac{0.94\lambda}{L \cos(\theta)} \quad (2)$$

where  $B(2\theta)$  is the FWHM (in radians) of the peak at angle  $\theta$ ,  $\lambda = 0.154056$  nm ( $\text{CuK}_{\alpha 1}$ ), and  $L$  is the size of the crystallite.

## **Results and Discussion**

Unsaturated tests with an unirradiated segment of N-Reactor fuel demonstrated that the fuel rapidly corrodes in simulated groundwater to form solids and colloidal-sized uranium oxides. Within 77 days, a 1.4 g sample completely reacted, leaving none of the original metallic uranium. The fast reaction progress observed in these unsaturated tests is consistent with observations by other researchers (1-3, 16).

All test leachates sampled exhibit at least two colloid populations with a mean diameter of ~3-6 nm and 100-200 nm (refer to Table I). A bimodal distribution of colloidal particles is not unusual for uranium-based weathering or reprocessing processes. In earlier work, for example, unsaturated tests on spent  $\text{UO}_2$  fuel generated micron-sized aggregates containing 20-100 nm individual particles (9). The colloidal phases identified using TEM were corrosion products (schoepite and soddyite) of the fuel. Particle releases from effluents of the Sellafield reprocessing plant included discrete 20 nm colloids and 100 nm pseudocolloids (17). In the N-Reactor fuel, the small-sized colloid population has a narrow distribution (peak width ~ 5-11 nm) and consistently makes up a large percentage of the total colloids (> 40%). The observation of abundant small-sized colloids is significant as the DLS method biases intensity distributions to large particles. The 100-200 nm particle population has a wide distribution (peak width



~200-400 nm). In addition, the 12-day sample from the spent fuel holder sample and the 48-day sample from the base of the vessel exhibit a large-sized component (mean diameter ~600 nm) of low amplitude. This large-sized component is attributed to a small population of large particles and will be discussed with the TEM results. These colloid samples have near-neutral pH (pH ~ 7.5) and a large polydispersity ( $Q = 0.4$ ). The polydispersity is lower ( $Q \leq 0.3$ ) for the colloidal solutions at higher pH (pH ~ 9). Average values from multiple DLS measurements are reported in Table I; refer to Figure 2 for representative size distribution plots from single DLS measurements.

Table I. Mean Particle Size and Polydispersity (Q) Determined by DLS for Colloids from Unsaturated Tests on Unirradiated N-Reactor Fuel with EJ-13 at 90°C.

Duration (days)	Leachate Location <sup>a</sup>	pH	Peak #	Mean Size <sup>b</sup> (nm)	Width <sup>b</sup> (nm)	Abundance (%)	Q
12	H-3	7.4	1	3.9 ( $\pm 0.8$ )	6.4 ( $\pm 1.7$ )	37	0.4
			2	100 ( $\pm 10$ )	120 ( $\pm 38$ )	57	
			3	580 ( $\pm 190$ )	410 ( $\pm 150$ )	6	
12	B	9.1	1	5.5 ( $\pm 1.3$ )	11 ( $\pm 3.6$ )	50	0.2
			2	190 ( $\pm 2$ )	180 ( $\pm 10$ )	50	
48	B	7.5	1	3.3 ( $\pm 1.1$ )	5.1 ( $\pm 2.4$ )	78	0.4
			2	140 ( $\pm 71$ )	140 ( $\pm 98$ )	15	
			3	620 ( $\pm 310$ )	390 ( $\pm 210$ )	7	
77	B	9.1	1	4.5 ( $\pm 0.3$ )	7.9 ( $\pm 0.7$ )	56	0.3
			2	150 ( $\pm 4$ )	180 ( $\pm 56$ )	44	

<sup>a</sup> Location where the leachate solution was sampled: H-3 = stage 3 (bottom) holder and B = base of test vessel.

<sup>b</sup> Standard deviations reported in parantheses are calculated from multiple DLS measurements.

Dynamic light scattering was used to estimate colloid concentrations in the N-Reactor tests (refer to Table II). Correlation of the scattering intensity with colloid concentration was performed by relating the scattering intensity of standard polystyrene nanospheres (70 and 300 nm diameter) to known concentrations of their colloidal suspensions. Applying this relationship to the measured scattering intensity of colloids from the tests yielded estimates of sample colloid concentrations. The results presented are considered approximate because differences in composition and refractive index between the sample and standard affect scattering properties. In this case, the refractive index for polystyrene is 1.6 (18), while the refractive index for  $\text{UO}_2$  is 2.4 (3). Thus, estimated concentrations will be presented only to show a relative trend and not absolute populations of the colloids. The highest concentration of colloids (3-8 mg/L) and total uranium ( $2.5 \times 10^{-4} \text{ M}$ ) was observed from the 12-day unsaturated test in the stage 3 holder. For the same test duration, the colloids in the base of the vessel were significantly lower

in concentration (1-3 mg/L). The total uranium concentration was also significantly lower ( $4.7 \times 10^{-6} \text{ M}$ ). As the unsaturated test progressed and the fuel was consumed, the concentration of colloids in the leachate at the base of the vessel decreased (0.1-0.3 mg/L). While colloids appear to be present in the EJ-13 groundwater sample, the colloid size distribution was unresolved due to the presence of large, unstable particles (composition unknown at this time).

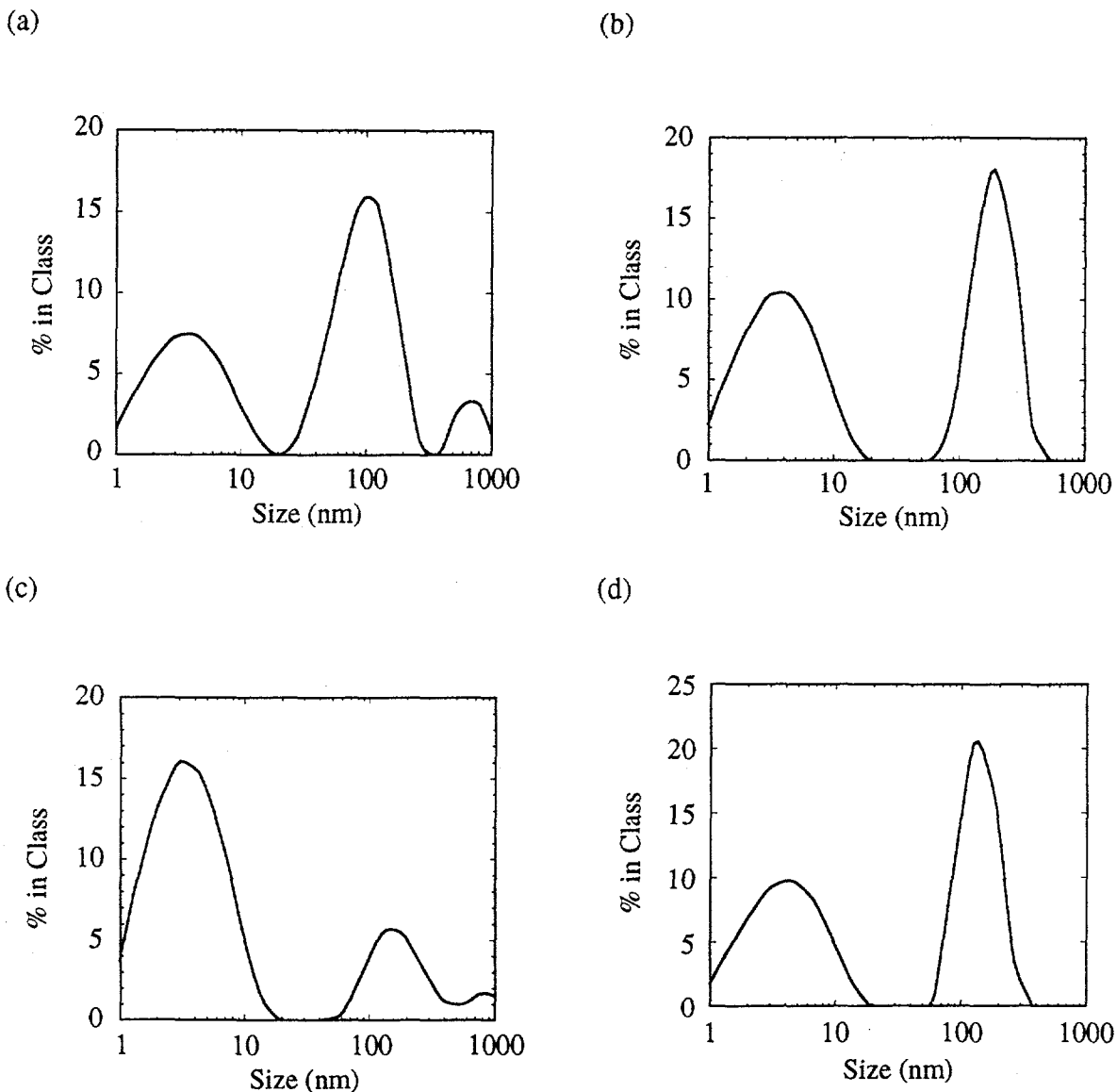


Figure 2. Intensity-Weighted Particle Size Distributions for the Colloids from the N-Reactor Unsaturated Tests as a Function of Test Duration. Representative colloid distributions are shown for leachates sampled (a) from the stage 3 holder at 12 days and from the base of the test vessel at (b) 12, (c) 48, and (d) 77 days.

Table II. Leachate Composition from Samples Collected from Unsaturated Tests on Unirradiated N-Reactor Fuel with EJ-13 at 90°C.

Property	Test Samples				
Duration (days)	0	12	12	48	77
Sample Location <sup>a</sup>	EJ-13	H-3	B	B	B
Scattering Intensity <sup>b</sup> (10 <sup>3</sup> counts/sec)	30	267	101	50	12
Estimated 70 nm Colloid Concentration <sup>c</sup> (mg/L)	0.9	8	3	2	0.3
Estimated 300 nm Colloid Concentration <sup>c</sup> (mg/L)	0.3	3	1	0.6	0.1
Uranium Concentration <sup>d</sup> (M)	1.4 x 10 <sup>-8</sup>	2.5 x 10 <sup>-4</sup>	4.7 x 10 <sup>-6</sup>	3.9 x 10 <sup>-5</sup>	2.4 x 10 <sup>-5</sup>

<sup>a</sup> Location where the leachate solution was sampled: EJ-13 = original groundwater leachant, H-3 = stage 3 (bottom) holder, and B = base of test vessel.

<sup>b</sup> The scattering intensity was collected at a scattering angle of 90° and at a laser power of 3 mW.

<sup>c</sup> Concentration of UO<sub>2</sub> colloids was estimated from polystyrene standards of known concentrations using two particle sizes.

<sup>d</sup> The uranium concentration in the leachate includes colloidal and dissolved uranium species.

To help explain the larger polydispersity observed for the lower pH leachates, the ionic strength was estimated (based upon major cations and anions). An increase in the ionic strength will increase particle-particle attractive forces by compressing the electric double layer and increase the polydispersity of the system by forming larger particles. Each of the leachates did not show any significant differences in the estimated ionic strength ( $2 \times 10^{-3}$  to  $3 \times 10^{-3}$  m). However, differences in solution pH have been shown to have a dramatic effect on colloid stability due to changes in the particle surface charge. The stability of UO<sub>2</sub> colloids solution under repository-relevant conditions has been examined by other researchers. The results show that the UO<sub>2</sub> particles are negatively charged in basic solutions. The zeta potential of uraninite is -22 to -30 mV at pH 7 to 9, respectively (19), with a zero point of charge of 3.5 (19, 20). Thus, the UO<sub>2</sub> colloids from the unsaturated tests are stable in solution pH values of 7 to 9, as evidenced by monitoring the samples over the several weeks. The colloid size distribution did not change and the count rate or concentration decreased only slightly over the course of several weeks. The minor but large-size component in the holder at 12 days and in the vessel base at 48 days may be attributed to another phase that is unstable at higher pH values. Results from TEM indicate that uranyl silicates are also present in the leachates; however, surface charge measurements are needed for the uranyl silicates to accurately describe their stability.

The particle size distributions obtained using DLS on colloids from the stage 3 holder in the 12-day test are consistent with the TEM results. The later analysis revealed copious amounts of ~100 nm uranium-oxygen-based colloids that were composed of smaller subunits with dimensions of ~5 nm (Figure 4). The microstructure of these colloids is consistent with XRD coherence size analysis of  $\text{UO}_2$  from sludge recovered from 48-day tests. These colloids were weakly diffracting in the electron beam, making positive phase identification difficult. Also observed was a relatively minor component of uranyl-silicate material (Figure 5), which presented a needle-like morphology. The composition and growth mode suggest a structure like that of the mineral *soddyite*.

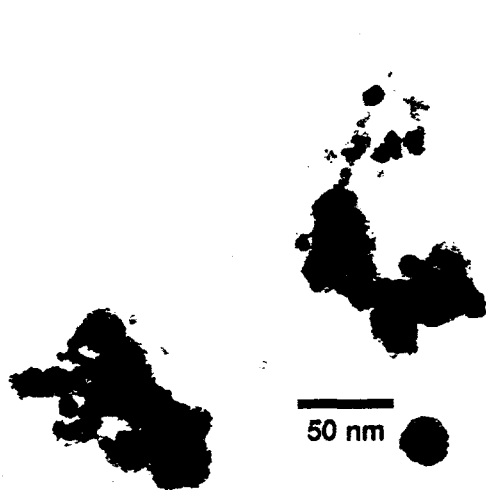


Figure 4. Bright-field TEM Micrograph of Uranium Oxide or Oxy-hydroxide Colloids from the Stage 3 Holder of the 12-Day Unsaturated Test of the Unirradiated N-Reactor Fuel. These particles are typical of many that were observed on the sample grid.

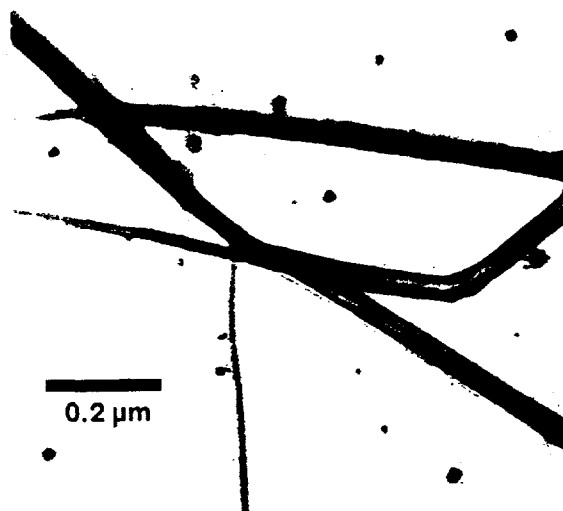


Figure 5. Bright-field TEM Micrograph of Uranium Silicate Colloids from the Stage 3 Holder of the 12-Day Unsaturated Test. The needle-like structure is typical of uranium-rich silicate hydrates; the composition of this material suggests *soddyite*.

The XRD results for solids recovered from the -stage holders at 48 and 77 days show that the predominant material formed in these tests is a black sludge with a mineral composition of  $\text{UO}_2$  and uranyl oxide hydrates. The XRD analyses indicate that the metal fuel is oxidized to  $\text{UO}_2$  with particles in the 7- to 12-nm size range. The lower value (7 nm) is obtained by reading the FWHM directly from the diffraction patterns, while the larger value (12 nm) is obtained from the peak search report. Owing to the extremely small particle coherence size of  $\text{UO}_2$  in the sludge, substantial line broadening was observed.

## Summary

The formation of  $\text{UO}_2$  colloidal particles from the corrosion of N-Reactor fuel has been examined under conditions likely in a repository setting. Work performed in this study demonstrates that unirradiated metallic uranium fuel degrades in simulated groundwater to a series of solid and colloidal uranium oxides ( $\text{UO}_{2+x}$ ). Dynamic light scattering (DLS) and transmission electron microscopy (TEM) indicate that the colloidal uranium oxides are present in solution predominantly as spherical,  $\sim 5$ -nm-sized  $\text{UO}_2$  colloids and  $\sim 100$ - $200$  nm aggregates of the smaller colloids. The DLS results indicate that the  $\text{UO}_2$  colloids are stable in solutions with pH values of 7 to 9 and low ionic strength ( $2\text{--}3 \times 10^{-3}$  M). These stable colloids may act as a vehicle for moving colloid-associated radionuclides through the repository. Other researchers have noted the existence of small colloidal  $\text{UO}_2$ , where uraninite colloids of  $<100$  nm have been reported (20, 21). Polycrystalline aggregates of  $\text{UO}_2$  from the natural fission reactor in Gabon exhibit dimensions of  $\sim 5$  nm (22). Colloidal-sized uranium oxide phases have also been identified as sorbed to bacterium (21), indicating another mode of potential subsurface transport of colloidal contaminants.

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