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CERIUM, URANIUM, AND PLUTONIUM BEHAVIOR IN GLASS-BONDED SODALITE, A CERAMIC NUCLEAR WASTE FORM

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Abstract

Glass-bonded sodalite is being developed as a ceramic waste form (CWF) to immobilize radioactive fission products, actinides, and salt residues from electrometallurgical treatment of spent nuclear reactor fuel. The CWF consists of about 75 mass % sodalite, 25 mass % glass, and small amounts of other phases. This paper presents some results and interpretation of physical measurements to characterize the CWF structure, and dissolution tests to measure the release of matrix components and radionuclides from the waste form. Tests have been carried out with specimens of the CWF that contain rare earths at concentrations similar to those expected in the waste form. Parallel tests have been carried out on specimens that have uranium or plutonium as well as the rare earths at concentrations similar to those expected in the waste forms; in these specimens UCl_3 forms UO_2 and PuCl_3 forms PuO_2 . The normalized releases of rare earths in dissolution tests were found to be much lower than those of matrix elements (B, Si, Al, Na). When there is no uranium in the CWF, the release of cerium is two to ten times lower than the release of the other rare earths. The low release of cerium may be due to its tetravalent state in uranium-free CWF. However, when there is uranium in the CWF, the release of cerium is similar to that of the other rare earths. This trivalent behavior of cerium is attributed to charge transfer or covalent interactions among cerium, uranium, and oxygen in $(\text{U,Ce})\text{O}_2$.

1. Introduction

Scientists at Argonne National Laboratory (ANL) are developing an electrometallurgical process to convert spent sodium-bonded nuclear reactor fuel into repository-acceptable forms [1]. The electrometallurgical process is carried out in a molten salt (LiCl-KCl) electrolyte containing UCl_3 to facilitate electrolysis. Spent fuel rods are chopped into small segments, placed into anode baskets, and immersed in the molten salt for electrolysis. The process results in the following:

1. Purified uranium is deposited on the cathode.

2. Noble metal fission products are retained on the anode, and are converted to a metallic waste form.
3. Easily oxidized fission products and transuranic elements accumulate in the molten salt electrolyte used in the process.

The radioactive materials that accumulate in the molten salt electrolyte must be removed periodically and eventually conditioned (i.e., treated so as to be acceptable at a nuclear waste repository). To condition the salt, the salt is first blended at 500°C with granular dehydrated zeolite 4A (containing a clay binder) to incorporate most of the salt and radionuclides within structural cages of the zeolite. This "salt-occluded zeolite" is then mixed with a glass binder and processed at high temperature and pressure under an inert atmosphere in a hot isostatic press. The zeolite 4A converts to sodalite; the product is glass-bonded sodalite, a ceramic waste form (CWF) that consists of about 75 mass % sodalite, 25 mass % glass, and small amounts of halite, clay, and oxide phases [2].

To qualify the CWF for transport to and acceptance by a nuclear waste repository, the testing program at ANL must specify certain properties of the waste. In particular, it must identify phases containing radionuclides and provide methodology to monitor CWF product consistency. Also, to provide repository performance engineers with data they need to assess the performance of the repository, the program must measure the release rates of matrix components and radionuclides and must provide experimental parameter values for a mechanistic model.

This paper reports the structural characterization of rare-earth and actinide-containing samples of the CWF and presents the results and interpretation of tests being conducted to measure the corrosion rates at which the matrix components and radionuclides are released from the waste form. Scoping tests have been carried out with CWF specimens that contain yttrium, lanthanum, cerium, praseodymium, neodymium, samarium, and europium at concentrations similar to those expected in the waste form. Parallel tests have been carried out on monolithic specimens that contain uranium as well as the rare earths at concentrations similar to those expected in the waste form.

2. Experimental

2.1. Preparation of Glass-Bonded Sodalite. Granular dehydrated zeolite 4A, containing a clay binder, was mixed with process eutectic LiCl-KCl salt containing chlorides of simulated fission

products by mechanical blending at 773 K for several hours to occlude most of the salt into the zeolite cages. The blended zeolite was then mixed with a glass binder (in the ratio 75 mass % blended zeolite to 25 mass % glass binder) and processed at 1173 K and ~1500 bar pressure under an inert atmosphere in a HIP to produce CWF [2]. Parallel samples were prepared of CWF containing simulated fission products and uranium. Compositions of CWF and CWF containing uranium were determined by elemental analysis and are given in Tables 1 and 2.

Glass-bonded sodalite doped with plutonium (primarily ^{239}Pu) was prepared [3] from LiCl-KCl eutectic salt containing 1.5 mol % Pu and no fission products, or 1.5 mol % Pu and simulated fission products at concentrations similar to those in Table 1. (The simulated fission product concentrations were those expected for waste salt after processing 300 driver fuel rods.) Under an inert atmosphere these salt mixtures were blended with zeolite 4A powder that had been dried to 0.56 mass % H_2O and mixed with glass binder at a zeolite:glass ratio of 75:25 mass %. The mixture was then heated under inert-atmosphere hot uniaxial pressing (HUP) conditions (1023 K, 570 bar) to obtain "dry" CWF. Some of this CWF was exposed to air (50% relative humidity) for 4 days; this material is described below as "CWF made from sim. f. p. salt, 'wet'."

2.2. X-Ray Diffraction. Representative samples of eutectic LiCl-KCl salt containing rare earths, uranium, and/or plutonium, as well as samples of HIPed or HUPed glass-bonded sodalite, were pulverized and loaded into X-ray capillaries in an inert atmosphere box. X-ray diffraction patterns were collected on Debye-Scherrer films, which were optically scanned and interpreted by search-match computer programs.

2.3. X-ray Absorption Spectroscopy. The Pu-doped glass-bonded sodalite was analyzed by XAFS (X-ray absorption fine structure spectroscopy) and XANES (X-ray absorption near edge spectroscopy) at the Materials Research Collaborative Access Team's beam-line insertion device (undulator) at the Advanced Photon Source at ANL. Reference spectra were also obtained of PuF_3 , PuO_2 , $\text{NaPuO}_2\text{CO}_3$, and Ba_3PuO_6 , which contain Pu(III), Pu(IV), Pu(V), and Pu(VI), respectively. Further details are given by Richmann et al. [3].

2.4. Temperature-resolved X-ray Diffraction and Gas Analysis. A sample of UCl_3 mixed with LiCl-KCl eutectic salt, 28.5 mol % UCl_3 -48.2 mol % LiCl-23.2 mol % KCl, was melted and cooled under an inert atmosphere. Under an inert atmosphere the mixed salt was pulverized and blended with dehydrated, powdered zeolite 4A, i.e. $\text{Na}_{12}(\text{AlSiO}_4)_{12}$ containing <0.2 mass % H_2O .

Temperature-resolved XRD measurements from 300 to 900 K were collected at the beam-line insertion device (undulator).

Parallel evolved gas analysis (EGA) experiments were carried out from 300 to 800 K to identify gaseous products of the reaction during heating [4].

2.5. Corrosion Testing of Glass-Bonded Sodalite. Materials for the corrosion tests were obtained from the HIP products. Cores were removed from the HIP can with a diamond-coated core drill. Monolithic specimens were obtained by wafering the core with a diamond saw. Crushed materials were obtained by crushing the annular material and unused cores and then sieving to obtain the appropriately sized particles. Standard test protocols for nuclear waste forms were followed to measure corrosion of the glass-bonded sodalite: the Material Characterization Center (MCC-1) Test with monolithic (wafer) samples and the Product Consistency Test (PCT) with crushed material [5]. Details of the standard procedures for preparing samples and conducting the tests are available [6,7]. Modifications to the standard procedure have also been published [8]. Both types of corrosion tests were run in demineralized water at 90°C (363 K) for durations up to 91 days. Aliquots of each test solution were analyzed for dissolved ions by inductively coupled plasma-mass spectrometry (ICP-MS) at the termination of the test.

3. Results

3.1. X-Ray Diffraction. The LiCl-KCl eutectic salt containing 1.5 mol % Pu was shown to contain K_2PuCl_5 by powder X-ray diffraction. The eutectic salt with 28.6 mol % U contained $K_3U_5Cl_{18}$ and LiCl.

The dehydrated zeolite contained small amounts of water, which reacted with all of the uranium or plutonium chlorides to produce dioxides. The amount of water was also sufficient to react with rare earth chlorides during the salt blending to produce rare earth oxides and/or oxychlorides. These phases were routinely detected by X-ray diffraction (XRD) and also were identified in micrographs of polished wafer samples by scanning electron microscopy (SEM) accompanied by energy dispersive spectrometry.

When uranium or plutonium was added to the process salt in the form of UCl_3 or $PuCl_3$, and either the blended zeolite or the HIPed CWF was analyzed, small amounts of UO_2 or PuO_2 were identified by X-ray diffraction. In separate experiments these phases have been identified by

SEM and transmission electron microscopy. We conclude that some of the UCl_3 and PuCl_3 reacted with water expelled from the zeolite by LiCl and KCl entering the zeolite cages during the blending to produce UO_2 or PuO_2 . A typical reaction producing PuO_2 is



A similar reaction can be written for the conversion of UCl_3 to UO_2 . These reactions are exoergic at the temperature of the transformation (~ 800 K): For the uranium case, $\Delta_r G(1) = -301$ kJ mol^{-1} , and for the plutonium case, $\Delta_r G(1) = -27$ kJ mol^{-1} [9].

3.2. X-ray Absorption Spectroscopy. The XANES spectra showed only Pu(IV) in the glass-bonded sodalite. The XAFS spectra taken near the Pu L edges showed essentially the same chi-space spectrum as that of a reference PuO_2 sample (Fig. 1). Within limits of detection this spectrum corresponds to $\geq 95\%$ of Pu present as PuO_2 in the glass-bonded sodalite.

3.3. Temperature-resolved X-ray Diffraction. Temperature-resolved XRD measurements of the U-bearing salt and zeolite mixture showed the presence of zeolite and $\text{K}_3\text{U}_5\text{Cl}_{18}$, which decomposed to K_2UCl_5 and UCl_3 upon heating. As shown in Fig. 2, diffraction peaks of UO_2 began to appear at 600 K and the zeolite 4A peaks shifted to higher 2θ , indicating lattice contraction consistent with chloride occlusion. The $\text{K}_3\text{U}_5\text{Cl}_{18}$ peaks began to disappear at 600 K and were replaced by peaks of K_2UCl_5 (not shown in Fig. 2).

In parallel EGA experiments, gases with m/e 2 (H_2), 28 (N_2), 18 (H_2O), 36 (H^{35}Cl), and 38 (H^{37}Cl) were identified [4]. These observations are consistent with reaction (1) to produce UO_2 from UCl_3 and H_2O .

3.4. Corrosion Testing. The solutions from the glass-bonded sodalite tests were analyzed for dissolved CWF components at the completion of each test. Results were calculated in terms of normalized elemental mass loss, $\text{NL}(i) = (m_i - m_{bi})/f_i S$, where m_i and m_{bi} are the masses of element i in the test solution and in the blank, respectively; f_i is the mass fraction of element i in the CWF; and S is the surface area in m^2 . The value of f_i was calculated from the composition of the appropriate CWF (see Table 1 and 2).

Figures 3 and 4 show the NL results of the MCC-1 and PCT, respectively. The NL values for Ce and Nd from the CWF are open symbols, while the NL values for Ce, Nd, and U from the U-doped CWF are filled symbols. In both the MCC-1 tests (Fig. 3) and in the PCT (Fig. 4) with

the CWF made without U, the normalized release of Ce was 2-10 times less than that of Nd. The normalized releases of the other rare earths (La, Pr) were similar to that of Nd. When U is present, however, the releases of U, Ce, and Nd as well as the other rare earths were similar.

The releases of rare earths in dissolution tests were much lower than those of matrix elements (B, Si, Al, Na), regardless of whether uranium was present in the CWF. When no uranium was in the CWF, the release of cerium was about three to ten times lower than release of the other rare earths. We attribute the low release of Ce to its presence in the CWF as Ce(IV) in CeO₂ or in a mixed rare earth oxide. However, when U was present in the CWF, the release of Ce was similar to that of the other rare earths. We attribute this behavior to the presence of (U,Ce)O₂, a well-studied mixed oxide that may contain partially reduced Ce and partially oxidized U [10-13]. Efforts are underway to confirm the presence of this phase and to use the resultant information to aid in the modeling of radioisotope release from the CWF.

4. Discussion

4.1. Cerium in CWF Containing Uranium. It is well known that when CeO₂ and UO₂ mixtures are heated to form compounds in which the metal-oxygen ratio remains 2, i.e. Ce_yU_{1-y}O₂, they form a partial or complete set of solid solutions [10-13] that nearly obeys Vegard's Law (the lattice parameter of the cubic unit cell of the mixed oxide is almost the same as a weighted average of the lattice parameters of the cubic unit cells of pure CeO₂, 0.54112 nm, and UO₂, 0.54700 nm) at all Ce/U ratios. However, it has long been recognized that the "cerium-uranium blue" color of some CeO₂-UO₂ solid solutions, e.g., Ce₂UO₆ (2CeO₂-UO₂), is indicative of a partial charge-transfer process or orbital hybridization [13,14]. Magnetic studies [13] of CeO₂-UO₂ solid solutions were interpreted in terms of partial charge transfer, e.g.,



However, X-ray absorption spectroscopy of Ce₂UO₆ [14] produced two results: (1) XAFS spectroscopy showed that this material has cation disorder, consistent with a solid solution containing Ce⁴⁺ and U⁴⁺ ions, and XANES showed the presence of only Ce⁴⁺ and U⁴⁺. These results led to the conclusion that Ce₂UO₆ has significant covalent hybridization among Ce, U, and O orbitals.

With our knowledge of the formation of UO₂ and PuO₂ during formation of glass-bonded sodalite from salt that contained U³⁺ or Pu³⁺, we interpret our corrosion test results as follows. In

glass-bonded sodalite with rare earths, Ce behavior is different from that of trivalent rare earths but similar to that expected for Ce^{4+} . In glass-bonded sodalite with rare earths and uranium, Ce behavior is similar to that of other rare earths, i.e., Ce behaves as Ce^{3+} . The presence of Ce^{3+} in glass-bonded sodalite with rare earths and uranium may result from charge transfer as proposed to occur in the mixed oxide $(\text{U,Ce})\text{O}_2$ [13]. It is more appropriate to describe the phenomenon as a covalent interaction among Ce, U, and O, as described by Antonio et al. [14] for Ce_2UO_6 . The mixed oxide may have an oxygen stoichiometry less than 2 ($\text{Ce}_y\text{U}_{1-y}\text{O}_{2\pm x}$) since reducing conditions existed for the reaction stoichiometry of equation (1).

4.2. Cerium in CWF Containing Plutonium. Mixtures of CeO_2 and PuO_2 form a continuous set of solid solutions [11] that obey Vegard's Law [15, 16]. Since the lattice parameters of CeO_2 (0.54112 nm) and PuO_2 (0.53960 nm) are nearly identical, it is not possible to use the lattice parameter to infer the composition of a CeO_2 - PuO_2 oxide. After sintering to 1273 K in air and cooling, CeO_2 - PuO_2 solid solutions appear as "hard, glassy lumps, lavender in color"; after grinding the color was reddish-brown [16]. Our samples are uniformly gray; they may show no unusual colors because the cerium and plutonium concentrations are low. We have no evidence to expect that glass-bonded sodalite containing rare earths and plutonium might show $\text{NL}(\text{Ce})$ different from that of glass-bonded sodalite containing rare earths without plutonium.

We are unaware of any studies of the CeO_2 - UO_2 - PuO_2 system. Samples of glass-bonded sodalite containing rare earths, uranium, and plutonium have recently been prepared at Argonne. We will examine them by XRD, XANES, and EXAFS and will study their dissolution behavior. We plan to search for Ce^{3+} and Ce^{4+} in these samples by XANES at an appropriate Ce L edge. Because uranium is in all of these samples, it is probable that the release of cerium in corrosion tests of these samples will follow that of the other rare earths.

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TABLES

Table 1. Compositions of the Glass-Bonded (CWF) Sodalite and Its Constituents
(Units: mass %)

Element	CWF ¹	Zeolite 4A	Salt	Glass	Element	CWF ¹	Zeolite 4A	Salt	Glass
B	1.50	NM ²	NM	5.99	La	0.05	NM	0.64	NM
Al	13.60	18.83	NM	3.95	Ce	0.10	NM	1.22	NM
Si	19.90	18.70	NM	29.5	Pr	0.04	NM	0.54	NM
Li	0.44	NM	5.49	BDL ³	Nd	0.16	NM	1.96	NM
Na	11.58	14.9	5.23	4.83	Sm	0.03	NM	0.35	NM
K	1.81	0.04	21.08	0.37	Eu	0.003	NM	0.03	NM
Cs	0.15	NM	1.82	NM	Cl	4.77	NM	59.45	NM
Sr	0.05	NM	0.54	0.02	I	0.01	NM	0.12	NM
Ba	0.08	NM	0.79	0.02					

¹CWF = Ceramic waste form, consisting of 25 mass % glass and 75 mass % salt-loaded zeolite. The latter is composed of 67 mass % zeolite and 8 mass % salt. The salt-loaded zeolite is converted to sodalite during HIP.

²NM = Not measured.

³BDL = Below detection limit.

Table 2. Compositions of the U-doped Glass-Bonded Sodalite (CWF) and Its Constituents
(Units: mass %)

Element	CWF ¹	Zeolite 4A	Salt	Glass	Element	CWF ¹	Zeolite 4A	Salt	Glass
B	1.50	NM ²	NM	5.99	La	0.04	NM	0.55	NM
Al	13.51	18.7	NM	3.95	Ce	0.08	NM	1.03	NM
Si	19.90	18.7	NM	29.5	Pr	0.03	NM	0.43	NM
Li	0.43	NM	5.49	BDL ³	Nd	0.12	NM	1.54	NM
Na	11.56	14.9	5.23	4.83	Sm	0.02	NM	0.29	NM
K	2.12	0.04	21.08	0.37	Eu	0.002	NM	0.02	NM
Cs	0.12	NM	1.82	NM	U	0.40	NM	5.35	NM
Sr	0.05	NM	0.54	0.02	Cl	4.62	NM	57.55	NM
Ba	0.07	NM	0.79	0.02	I	0.01	NM	0.11	NM

¹CWF = Ceramic waste form (as in Table 1).

²NM = Not measured.

³BDL = Below detection limit.

FIGURES

Fig. 1. XAFS of Pu-doped glass-bonded sodalite: (a) χ -space data for three Pu-containing LiCl-KCl eutectic salts and PuO₂ and (b) radial shell data for the same materials with the k-range = 3.3-15.5 Å⁻¹, dk = 1 Å⁻¹, and k² weighting. "Sim. f. p. salt" refers to salt containing simulated fission products at concentrations similar to those expected after processing 300 driver fuel rods. "Dry" and "wet" refer to CWF held under inert atmosphere and CWF exposed to atmosphere (50% relative humidity) for 4 days, respectively.

Fig. 2. Sequence of X-ray diffraction patterns of uranium-doped zeolite during in-situ heating.

Fig. 3. Results of MCC-1 corrosion tests at 90°C (363 K) with monolithic (wafered) glass-bonded sodalite with and without uranium. Note that cerium release is lower than release of other rare earths in absence of uranium.

Fig. 4. Results of PCT corrosion tests at 90°C (363 K) with crushed glass-bonded sodalite with and without uranium. Note that cerium release is lower than release of other rare earths in absence of uranium.

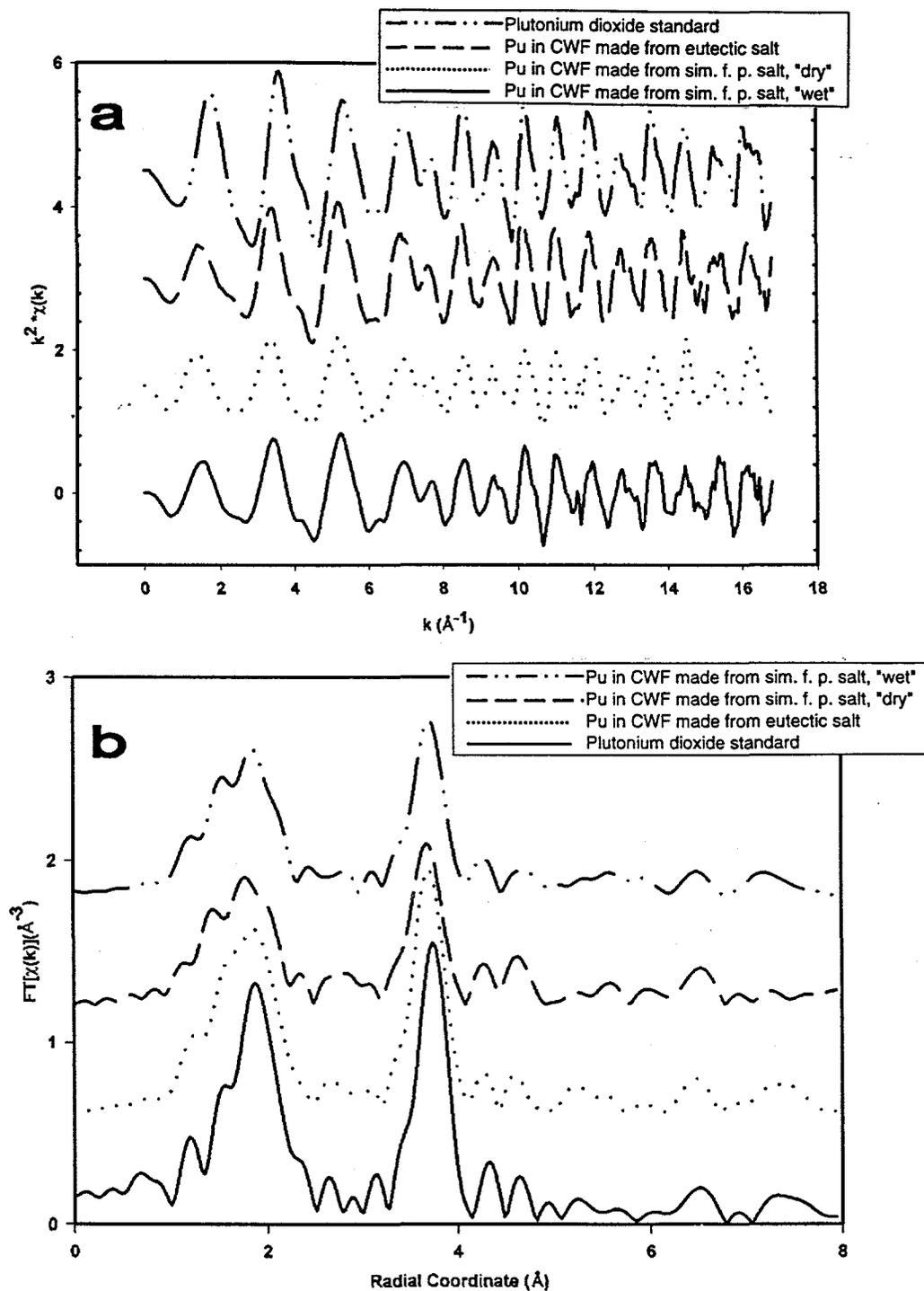


Fig. 1. XAFS of Pu-doped glass-bonded sodalite: (a) χ -space data for three Pu-containing LiCl-KCl eutectic salts and PuO_2 and (b) radial shell data for the same materials with the k -range = $3.3\text{--}15.5 \text{ \AA}^{-1}$, $dk = 1 \text{ \AA}^{-1}$, and k^2 weighting. "Sim. f. p. salt" refers to salt containing simulated fission products at concentrations similar to those expected after processing 300 driver fuel rods. "Dry" and "wet" refer to CWF held under inert atmosphere and CWF exposed to atmosphere (50% relative humidity) for 4 days, respectively.

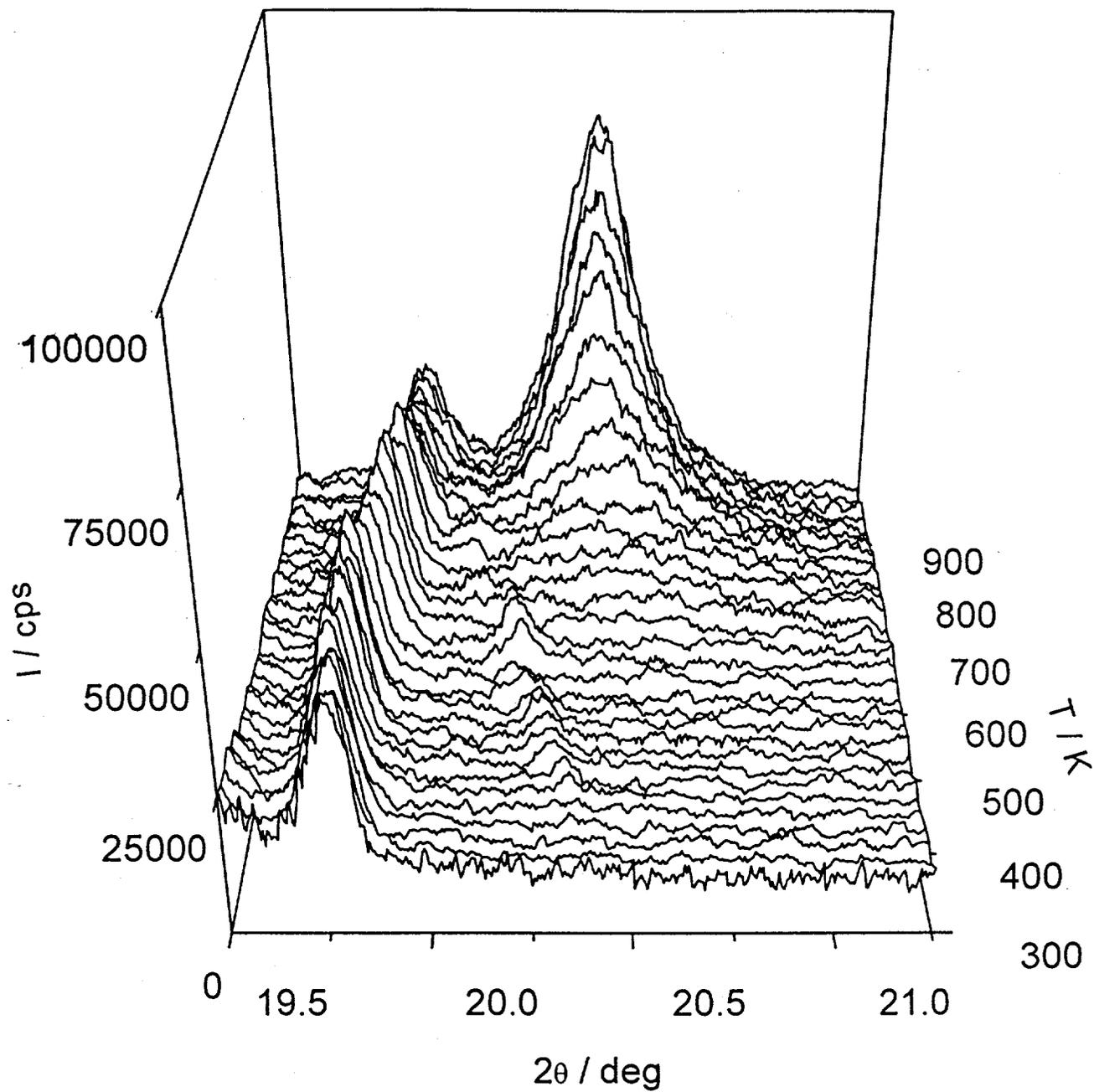


Fig. 2. Sequence of X-ray diffraction patterns of uranium-doped zeolite during in-situ heating.

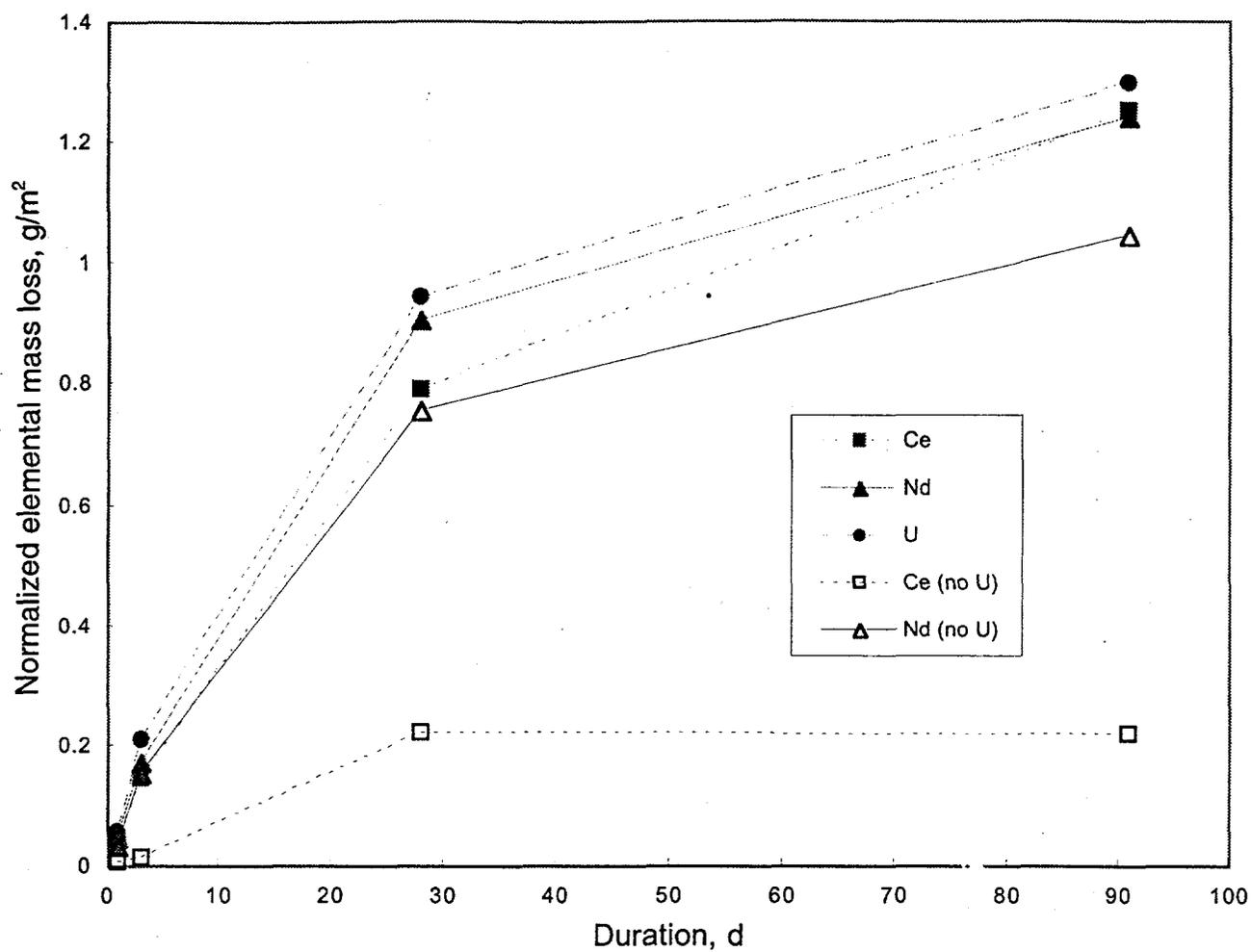


Fig. 3. Corrosion tests with monolithic (wafered) glass-bonded sodalite at 90°C with and without uranium. Note that cerium release is lower than release of other rare earths in absence of uranium.

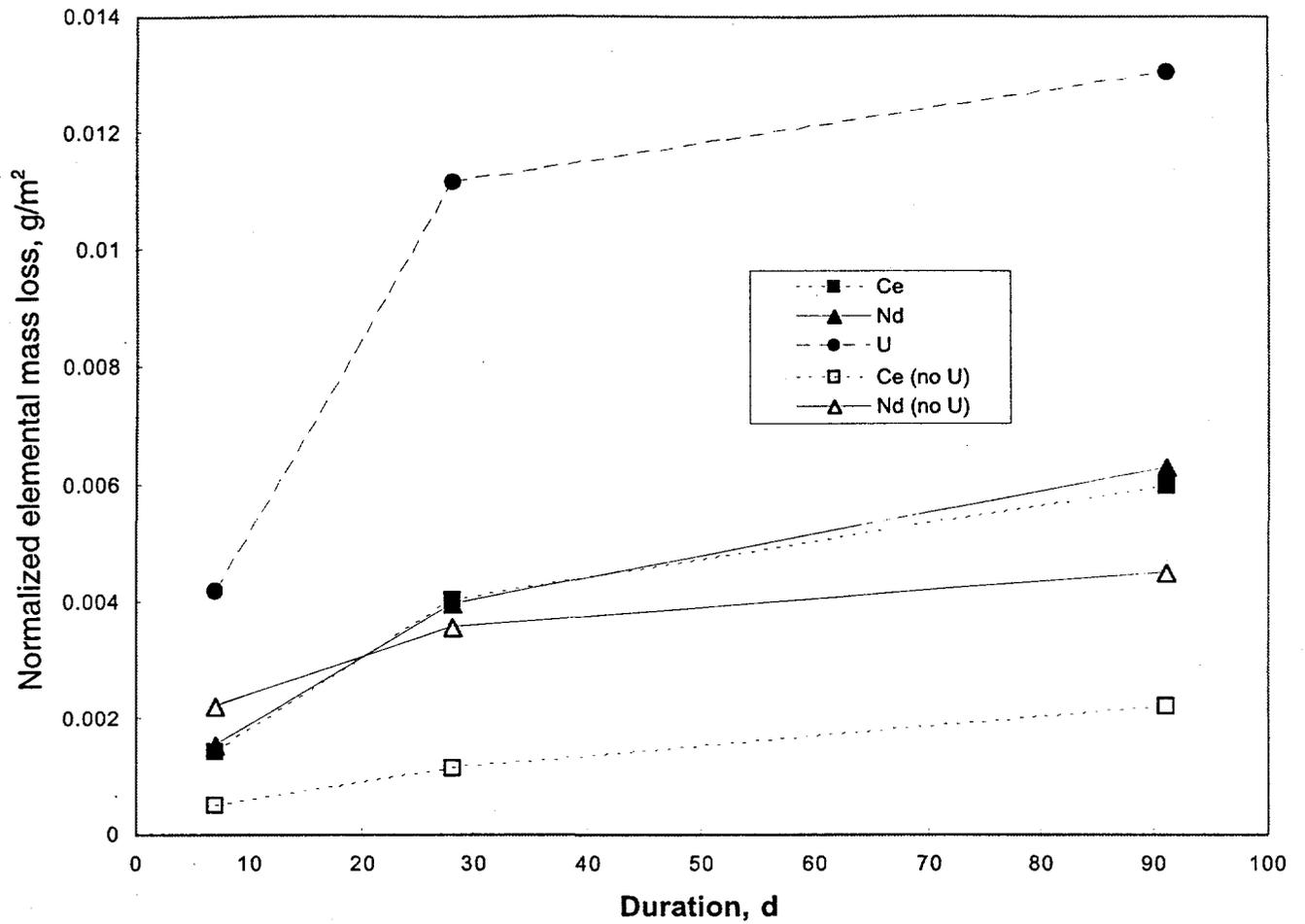


Fig. 4. Corrosion tests with crushed glass-bonded sodalite at 90°C with and without uranium. Note that cerium release is lower than release of other rare earths in absence of uranium.