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Time, Temperature, and Compositional Study of Am/Cm Target Glass Durability (U)

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Vitrification has been identified as a viable choice in the disposition of actinide materials such as americium (Am), curium (Cm), neptunium (Np), and plutonium (Pu). At the Westinghouse Savannah River Company near Aiken, South Carolina, a process is being developed to safely vitrify all of the highly radioactive americium/curium material and a portion of the other fissile actinide materials stored on site. This vitrification will allow safe transportation of the Am/Cm as well as easy storage at their final destination of Oak Ridge National Laboratory. This Am/Cm glass has been designed to be extremely durable in aqueous environments and can be selectively attacked by nitric acid to allow recovery of the valuable Am and Cm isotopes. A similar glass composition could allow for temporary or permanent storage of surplus plutonium. This paper will present results from a durability study on the actinide glass, Am/Cm Target, that will be used to vitrify the americium/curium material. The time, temperature, and compositional dependence of the Am/Cm Target durability will be discussed. All results show that the Am/Cm Target Glass is extremely durable and stable in aqueous systems, which is quite suitable for vitrification of americium/curium and possibly other actinides.

Introduction

Several time and temperature experiments had been performed to determine the rate and activation energy of dissolution of an actinide glass known as Am/Cm Target. These first studies, whose results are contained in an earlier report[1], substantiated a power law relation for the dissolution rate of the actinide glass. However, these earlier experiments raised questions about the time and temperature dependence of the dissolution rate. So another series of tests with both time and temperature variation were performed to supplement the earlier data. The new and old data were combined and then analyzed to help understand both the time and temperature effects on the actinide glass durability. In addition, there was interest in knowing how the Am/Cm Target glass durability varied with respect to the actinide loading in the glass. The concern over actinide loading was based on Oak Ridge National Laboratory's processing ability to recover the Am/Cm isotopes from the glass once it arrived at their facilities. In response to our customer's needs, a series of experiments were performed to investigate the dependence of the Am/Cm Target glass durability on different actinide loading. All the experiments were performed using the Product Consistency Test developed at WSRC and discussed in the next section.

Product Consistency Test Description

Product Consistency Tests (PCT Test Method B) were performed on Am/Cm Target and Approved Reference Material (ARM) glasses at WSRC Savannah River Technology Center (SRTC). The PCT's were done according to the requirements in the American Society for Testing and Materials (ASTM) Test Method C1285-94 *Determining Chemical Durability of Nuclear Waste Glasses: The Product Consistency Test (PCT)*[2].

Tests were performed on the Am/Cm Target and the Approved Reference Material (ARM) glasses in triplicate for various time periods and temperatures. Duplicate water blanks were run for each time and temperature. Table I lists the s ilica (Si) and boron (B) leachate concentrations zeroed against their corresponding blank samples. Table II lists the oxide compositions of the Am/Cm Target (ACT) and ARM-1 glasses.

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1 Daniel, W. E., D. R. Best, W. G. Ramsey, and T. F. Meaker, *Determining the Dissolution Rates of Actinide Glasses: A Time And Temperature Product Consistency Test Study*, Savannah River Technology Section, Westinghouse Savannah River Company, Aiken, SC 29808, presented at the I&EC Special Symposium for American Chemical Society in Atlanta, GA on September 17, 1995.

2 ASTM C 1285-94, Annual Book of ASTM Standards, American Society for Testing and Materials, Committee C-26 on Nuclear Fuel Cycle, Subcommittee C26.13 on Repository Waste Package Materials Testing, Volume 12.01, 1994.

Table I. PCT Leachate Concentrations for Am/Cm Target (ACT) and ARM Glasses

Temp °C	Time day	ACT-Si ppm	ARM-Si ppm	ACT-B ppm	ARM-B ppm
60	7	0.852	33.950	0.280	11.890
60	7	0.901	32.350	0.332	10.590
60	7	0.880	36.150	0.279	12.990
90	1	0.835	40.039	0.408	12.422
90	1	0.996	39.948	0.416	12.311
90	1	1.057	44.792	0.226	14.859
90	3	1.826	63.168	0.441	18.670
90	3	1.192	60.606	0.159	17.539
90	3	1.403	59.632	0.159	17.013
90	5	1.151	67.423	0.640	21.323
90	5	1.104	68.542	0.555	22.466
90	5	1.182	65.523	0.519	21.914
90	7	1.776	69.946	0.450	23.490
90	7	1.856	75.646	0.471	28.590
90	7	1.716	73.546	0.427	26.790
90	7	2.100	*	0.537	*
90	7	2.050	*	0.514	*
90	7	2.120	*	0.499	*
90	14	2.340	*	0.535	*
90	14	2.410	*	0.545	*
90	14	2.330	*	0.515	*
90	28	3.040	*	0.613	*
90	28	2.940	*	0.593	*
90	28	3.860	*	0.729	*
90	56	3.586	*	0.715	*
90	56	3.406	*	0.694	*
90	56	3.336	*	0.694	*
120	1	1.839	88.826	0.955	24.460
120	1	1.783	89.655	0.786	24.901
120	1	1.722	94.544	0.709	26.441
120	3	3.632	110.208	1.083	33.578
120	3	3.386	110.486	1.004	33.719
120	3	3.517	112.037	0.955	34.899
120	5	3.624	106.818	1.295	36.935
120	5	3.450	108.665	1.155	37.663
120	5	3.565	107.787	1.123	37.433
120	7	3.873	121.633	1.710	34.885
120	7	3.909	119.525	1.256	35.108
120	7	3.803	124.098	1.159	37.246
120	7	4.025	113.525	1.080	35.520
120	7	3.995	114.525	1.060	36.120
120	7	3.925	124.525	1.010	37.720

*not available

Table I (Continued). PCT Leachate Concentrations

Temp °C	Time day	ACT-Si ppm	ARM-Si ppm	ACT-B ppm	ARM-B ppm
120	14	4.703	134.880	1.777	43.360
120	14	4.155	133.402	1.020	43.436
120	14	5.540	136.732	1.171	44.335
120	28	6.219	154.012	1.761	59.542
120	28	8.075	145.899	2.224	56.016
120	28	7.314	152.843	2.037	57.523
150	7	6.653	202.903	1.534	80.534
150	7	6.923	199.903	1.574	78.234
150	7	6.283	185.903	1.434	74.234
180	1	3.834	200.026	1.384	70.448
180	1	3.906	199.127	1.154	71.132
180	1	4.725	202.187	1.313	72.796
180	3	5.251	279.641	1.935	107.454
180	3	5.830	263.838	1.809	100.380
180	3	5.543	266.564	1.522	101.703
180	5	5.537	272.196	1.122	107.713
180	5	6.761	257.117	1.235	102.191
180	5	6.320	274.089	1.172	109.615
180	7	6.090	287.770	1.401	112.971
180	7	8.470	286.770	1.811	111.971
180	7	6.910	302.770	1.611	118.971
180	7	4.900	279.722	1.997	120.259
180	7	4.359	275.647	1.574	120.703
180	7	4.724	258.517	1.501	111.590
180	14	5.438	373.547	1.818	162.292
180	14	5.595	342.711	1.393	143.897
180	14	7.319	340.284	1.752	145.249
180	28	7.297	1051.5345	1.786	2071.795
180	28	5.891	1162.8905	1.526	2199.871
180	28	9.8725	912.9505	2.492	1672.386
210	7	6.1965	894.7865	1.872	1099.982
210	7	6.5065	953.7865	1.912	1499.982
210	7	6.4565	887.7865	2.072	1199.982

Table II. Am/Cm Target and ARM-1 Glass Compositions

Oxide	ARM-1 Wt %	Am/Cm Target Wt %
Al_2O_3	5.59	6.24
B_2O_3	11.3	6.19
BaO	*	2.25
CaO	2.24	*
CeO_2	1.51	*
Ce_2O_3	*	8.22
Cs_2O	1.17	*
Eu_2O_3	*	0.72
Fe_2O_3	*	*
FeO	*	*
K_2O	*	*
La_2O_3	*	23.3
Li_2O	5.08	*
MgO	*	*
MnO	*	*
MnO_2	*	*
MoO_3	1.66	*
Na_2O	9.66	*
Nd_2O_3	5.96	11.62
PbO	*	13.48
SiO_2	46.5	27.97
TiO_2	3.21	*
ZnO	1.46	*
ZrO_2	1.8	*

*not applicable

Each glass was ground using a Tekmar grinder with tungsten carbide blades. The ground glass was then sieved and a 100-200 mesh size collected in a beaker. The glasses were then washed by forcibly adding 15-20 ml of de-ionized water to the beaker and then decanting. This process was done three times. The glasses were also washed twice by forcibly adding 15-20 ml of de-ionized water, placing the beaker in an ultrasonic water bath for two minutes and then decanting. The process was repeated with ethyl alcohol. The glasses were then placed in a convection oven over night to dry.

Teflon® vessels were then prepared to receive the dried samples. First, the vessels and lids were cleaned by soaking them in 0.16M Nitric Acid (HNO_3) at 90°C ± 10°C for approximately one hour on a hot plate. These items were then rinsed with de-ionized water. The vessels were then soaked in fresh de-ionized water at 90°C ± 10°C for about an hour on a hot plate. The vessels were filled 80% full with de-ionized water (pH 5.0- 7.0), capped, and placed in a convection oven at 90°C ± 2°C for 16 hours. The pH values of the water after the 16 hour period were still between the pH 5.0 - 7.0 range. Approximately 3.5 grams of each glass were then added to the cleaned Teflon® vessels and their weights recorded.

An Orion pH meter was calibrated using 4, 7, and 10 pH buffers. De-ionized water was collected at an electrical resistivity of 18 megaohms•cm and then a pH measurement was taken. The initial pH of the ASTM Type 1 water was 6.6. Approximately 35 grams of the de-ionized water was then added to each vessel and a total weight of the vessel, water, and glass was recorded. The vessels were capped using a CEM® capping station and then placed in a Blue M convection oven at the desired temperature within +/- 2°C. The following day, the vessels were removed and checked to make sure that they were still tightly sealed. The oven temperature was monitored at half an hour intervals during the course of the study, using an Omega Thermocouple Thermometer.

After each test period was complete, the appropriate vessels were taken out of the convection oven and allowed to cool. The vessels were weighed and the weight recorded. All of the samples had a weight loss of less than 1% over the course of the study. Each vessel was then uncapped and a pH taken. The final pH of the leachate solutions ranged from 5.0-7.5. The final pH levels of the ARM glass standards were 10.22, 10.26, and 10.27.

Sterilized syringes and filters were used to filter the leachate into pre-sterilized vials. A total of 20 ml was filtered into each vial and then 200 ml of ultrapure nitric acid was added. The samples were then submitted for elemental analyses on an Inductively Coupled Plasma-Atomic Emission Spectrometer (ICP-AES). Elements analyzed in the blanks, ARM glass standard, and Am/Cm glass leachates included silicon (Si), lead (Pb), boron (B), aluminum (Al), neodymium (Nd), barium (Ba), europium (Eu), lanthanum (La), and cerium (Ce). Silica and boron leachate analyses base-lined to the blank samples are shown in Table I. In the next section, dissolution rates are calculated from the data shown in Table I.

Dissolution Rate Calculation

Data obtained from the PCT Tests were analyzed to determine the dissolution rates of the Am/Cm Target and ARM Glasses. The dissolution rate equation can be written as:

(1)
$$\frac{dC}{dt} = C_0 \bullet \frac{G}{t} \bullet \left[\frac{t}{t_0}\right]^G$$

where C is the concentration at time t, C_0 is the concentration at t_0, t is the time in days, t_0 represents 7 days, and G is the logarithmic release rate.[3] This rate model is based on the doctorate research of W. G. Ramsey and is supported by similar models in the literature.[4] The term G gets its name from its derivation by plotting the log of concentration ratios versus time ratios. More specifically, G is found by plotting $\ln(C/C_0)$ versus $\ln(t/t_0)$. In these studies, the concentrations were represented by the AES-ICP ppm analyses. To compare the Am/Cm Target glass against the ARM glass, the concentrations of silica and boron were examined. These elements are good indicators of glass durability. The log-log plots for the Am/Cm Target and

[3]Ramsey, W. G., *Glass Dissolution Chemistry of the System Na_2O-B_2O_3-SiO_2-Al_2O_3-Fe_2O_3-CaO*, Ph.D. Thesis, Clemson University, 1994.

[4]Jantzen, C. M., "Prediction of Glass Durability as a Function of Glass Composition and Test Conditions: Thermodynamics and Kinetics," Proceedings of the First International Conference on Advances in the Fusion of Glass, American Ceramic Society, Westerville, OH, pp. 24.1-24.17, 1988.

ARM silica data for 90°C, 120°C, and 180°C are shown in Figures 1 through 6. The log-log plots for the Am/Cm Target and ARM boron data for 90°C, 120°C, and 180°C are shown in Figures 7 through 12. Please note that t_0 represents 7 days in these figures.

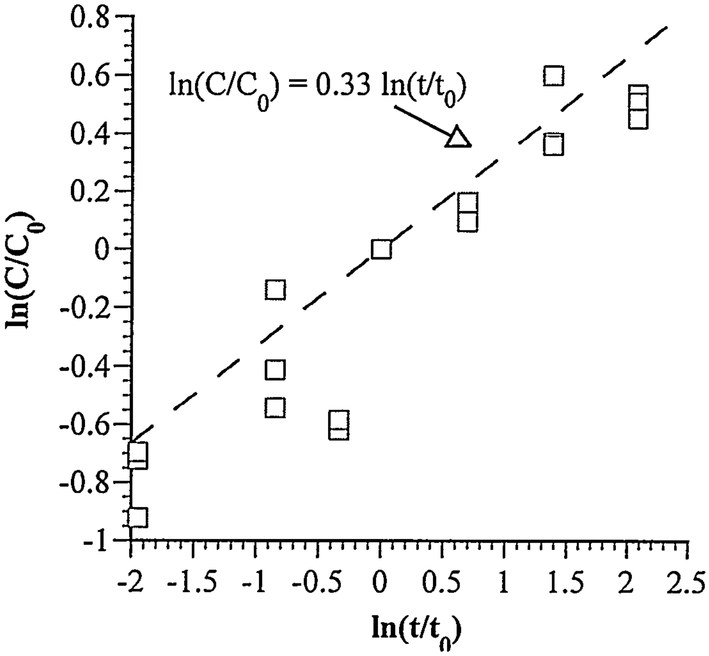

Figure 1. PCT Am/Cm Target silica data at 90°C.

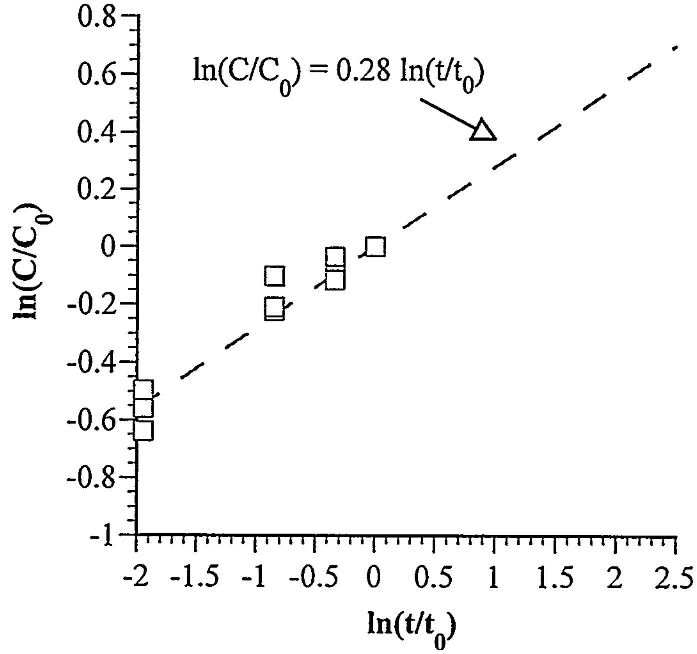

Figure 2. PCT ARM silica data at 90°C.

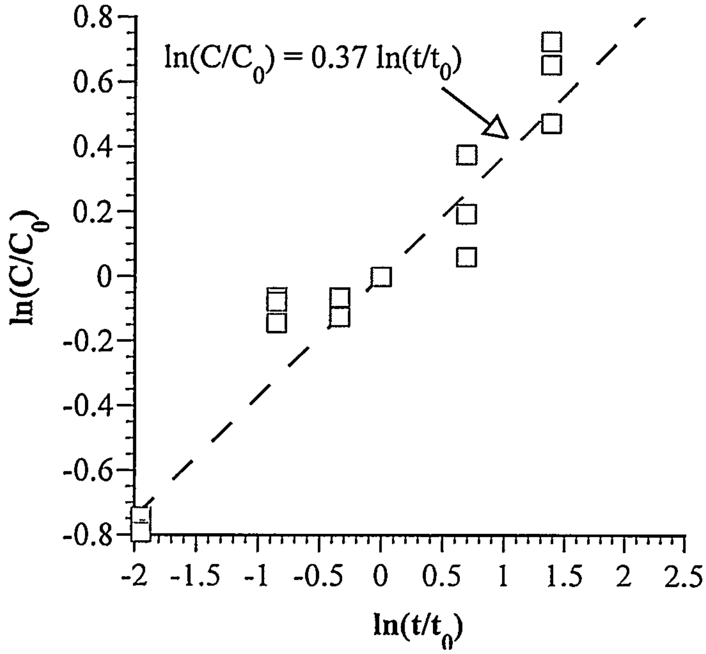

Figure 3. PCT Am/Cm Target silica data at 120°C.

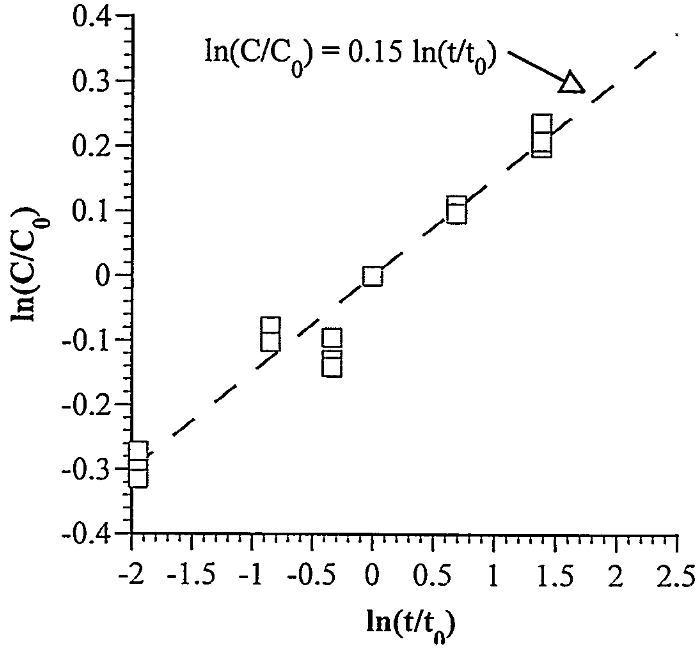

Figure 4. PCT ARM silica data at 120°C.

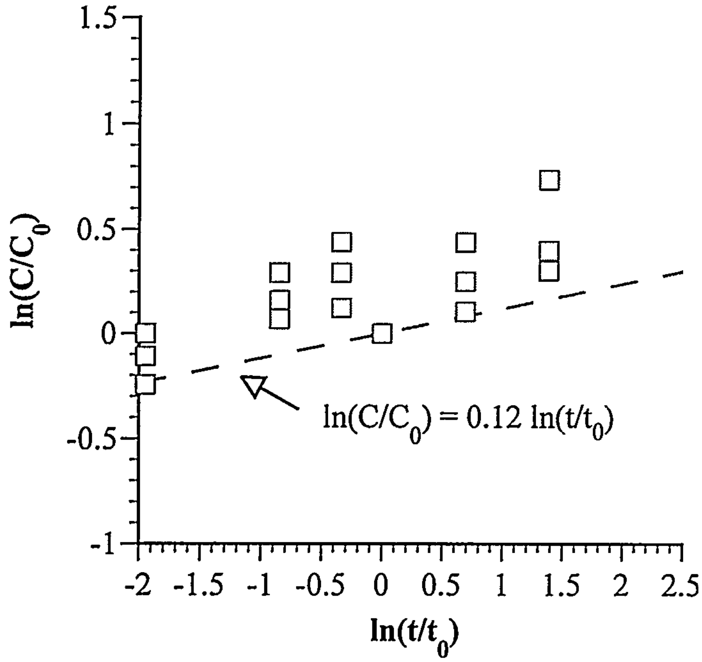

Figure 5. PCT Am/Cm Target silica data at 180°C.

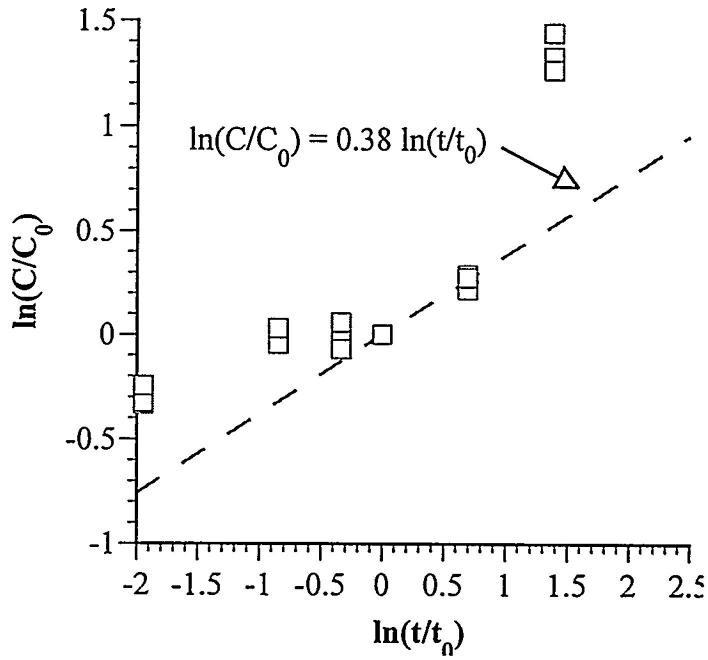

Figure 6. PCT ARM silica data at 180°C.

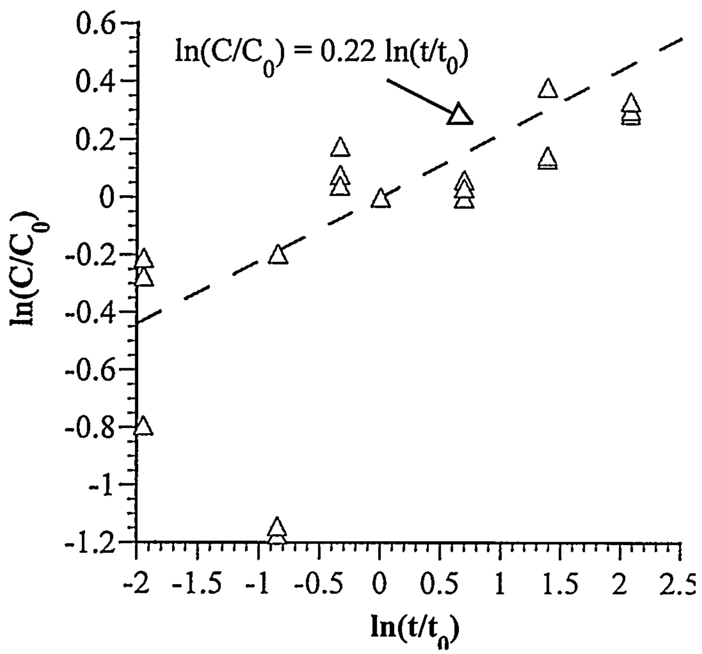

Figure 7. PCT Am/Cm Target boron data at 90°C.

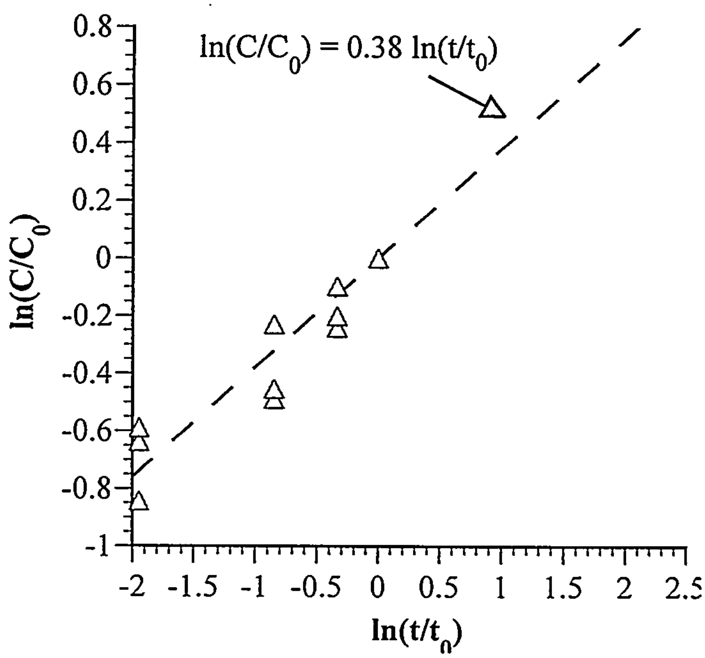

Figure 8. PCT ARM boron data at 90°C.

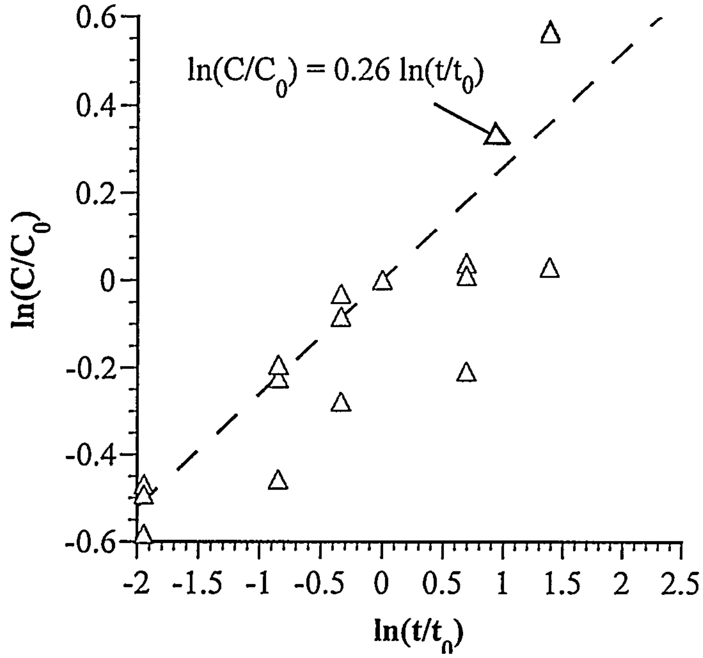

Figure 9. PCT Am/Cm Target boron data at 120°C.

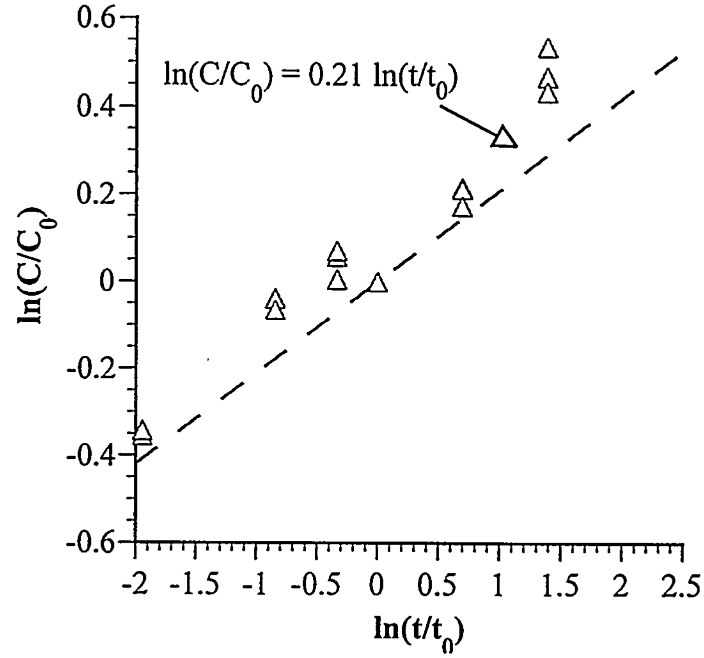

Figure 10. PCT ARM boron data at 120°C.

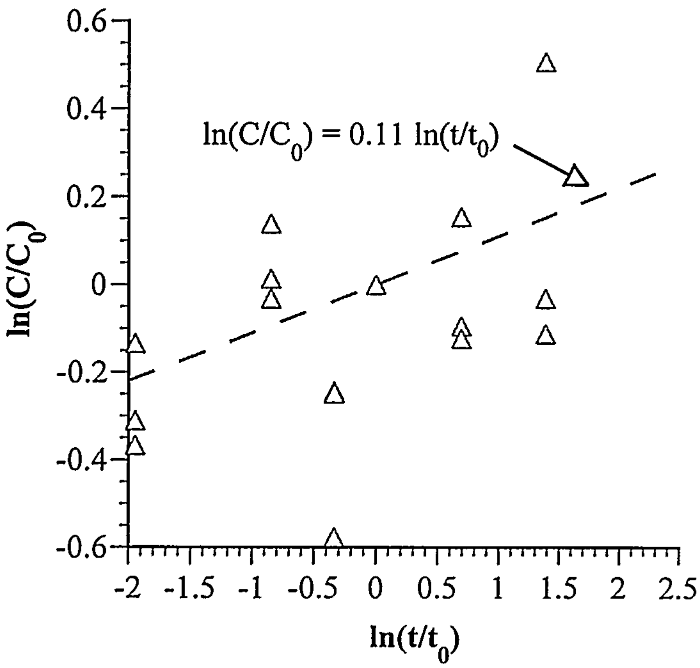

Figure 11. PCT Am/Cm Target boron data at 180°C.

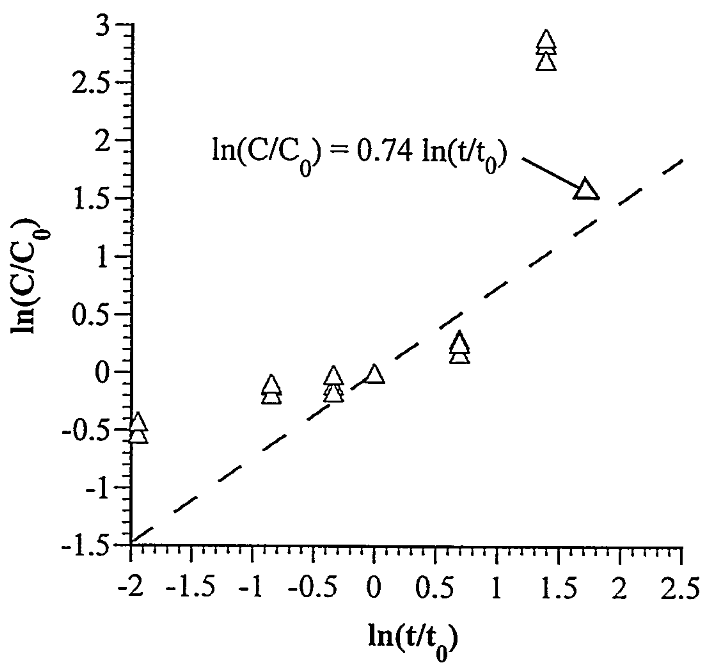

Figure 12. PCT ARM boron data at 180°C.

Figures 1 through 12 contain concentration data spanning 1 to 56 days. For each time and temperature point, three separate samples were maintained and calibrated against blank standards. The dashed lines in these figures represent the Least Squares (LS) linear fit through <u>all</u> the points. One may argue that some points could be dropped because they are statistically different from their duplicates. Since a least squares fit considers all the points, dropping the outliers and repeating the fit makes little difference. Another reason dropping the extreme points makes little difference is that the model in Equation 1 forces all fits through the point [$\ln(t_0/t_0)$,

$\ln(C_0/C_0)$] or [0,0]. The slopes of the fitted lines are the logarithmic rate terms (G). These rate terms plus or minus their 95% confidence limits are listed in Table III .

Table III. Logarithmic Release Rates (G) for Am/Cm Target (ACT) and ARM Glasses

Temperature °C	ACT-silica	ACT-boron	ARM-silica	ARM-boron
90	0.33 ±0.08	0.22 ±0.11	0.28 ±0.03	0.38 ±0.06
120	0.37 ±0.06	0.26 ±0.08	0.15 ±0.02	0.21 ±0.05
180	0.12 ±0.11	0.11 ±0.09	0.38 ±0.17	0.74 ±0.37

From these temperature studies, it appears that the dissolution rate changes in response to temperature. To incorporate this temperature effect, assume the logarithmic dissolution rates (G) found above are only a function of temperature. Now integrate the dissolution rate equation (1) over constant temperature to give:

(2) $$C(t) = C_0 \bullet \left[\frac{t}{t_0} \right]^{G(T)}$$

where C_0 is the concentration of an element at time t_0, G(T) is the logarithmic release rate for the element as a function of temperature, and t is the elapsed time. From the limited data points available, simple polynomial fits can be made to the logarithmic rate temperature data of Table III. These fits are shown in Figures 13 and 14 where the 95% confidence limits of each point are shown as error bars.

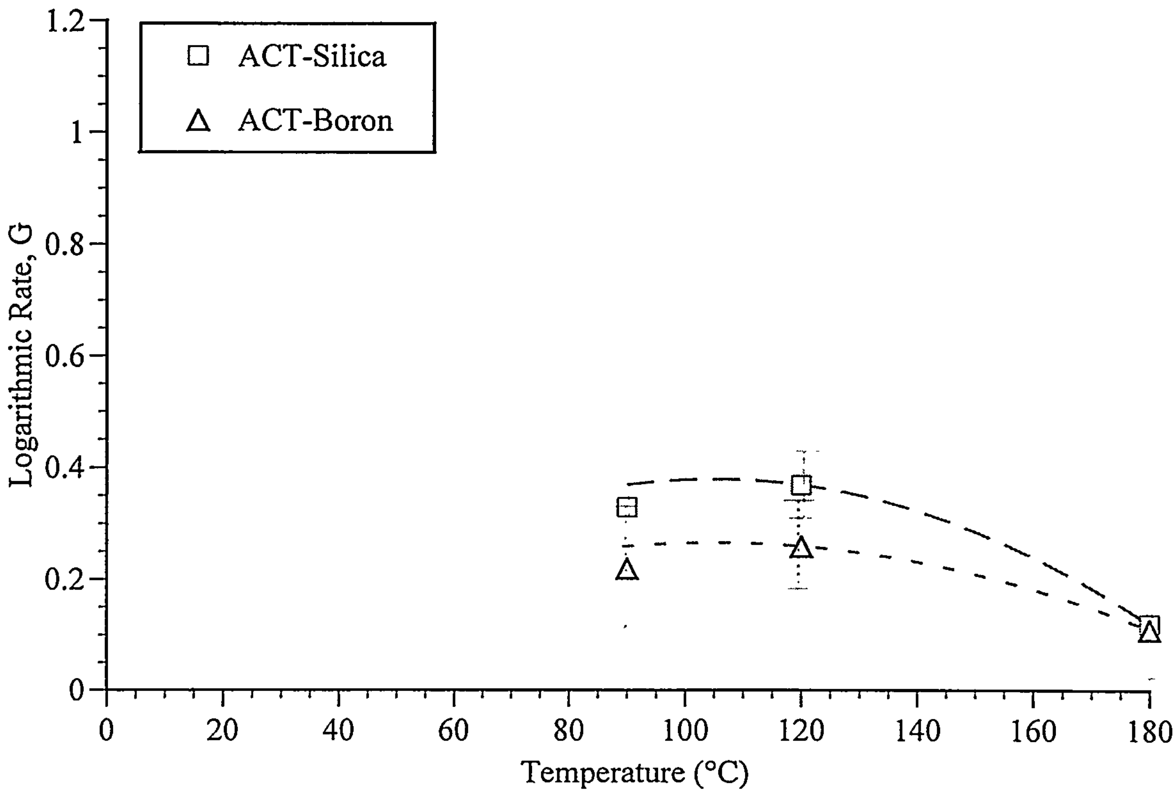

Figure 13. Logarithmic rate temperature dependence for Am/Cm Target (ACT) glass.

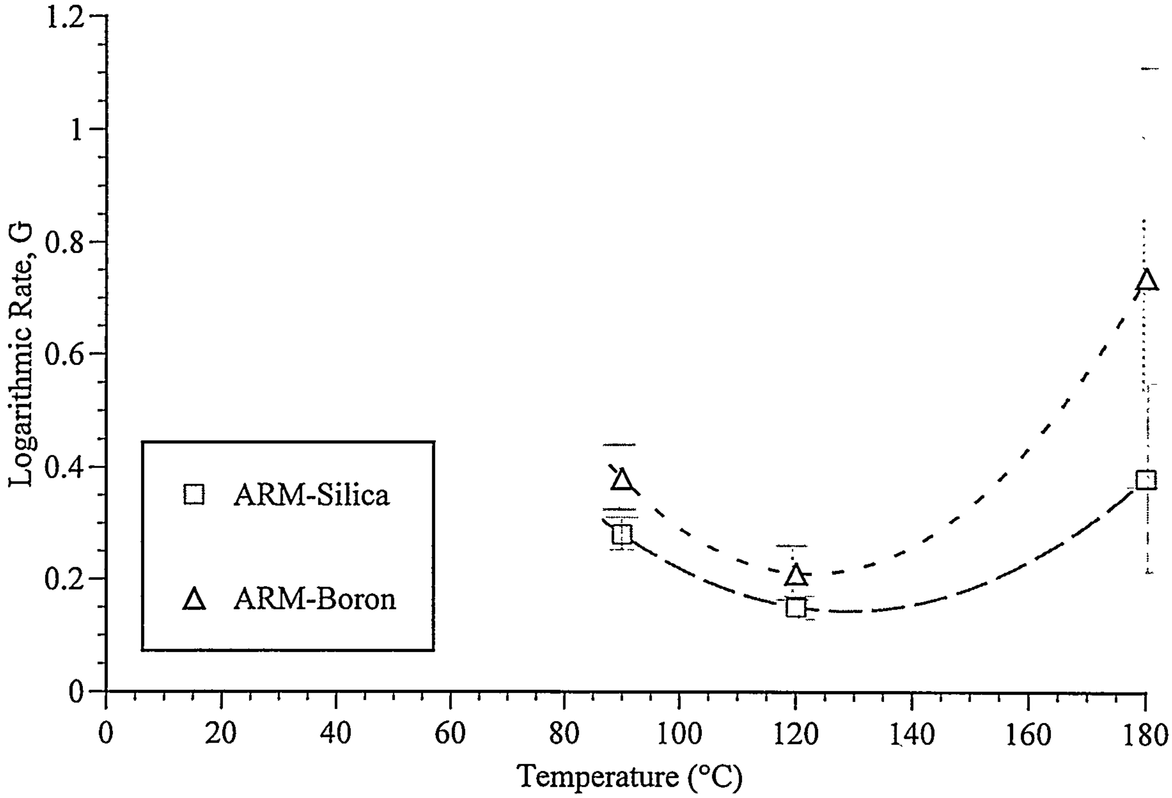

Figure 14. Logarithmic rate temperature dependence for ARM glass.

Based on the accuracy of the data, the following fits are assumed for the logarithmic rates between 90°C and 180°C:

(3) $G_{Lof\text{-}Si} = -5E\text{-}05\ T^2 + 0.0097\ T - 0.13$

(4) $G_{Lof\text{-}B} = -3E\text{-}05\ T^2 + 0.0058\ T - 0.04$

(5) $G_{ARM\text{-}Si} = 9E\text{-}05\ T^2 - 0.0234\ T + 1.65$

(6) $G_{ARM\text{-}B} = 2E\text{-}04\ T^2 - 0.0395\ T + 2.63$

Please note that these fits are based on a limited set of data and only approximate the observed behavior for this study. The logarithmic rate equations should not be applied outside the tested temperature range without performing more experiments. Other types of fits could be used but the quadratic equations most simply represent the observed behavior. The idea of the logarithmic rate (G) being a function of temperature is supported by the activation energy study conducted in an earlier experiment[1]. In that study the log of the PCT concentrations were plotted against the reciprocal of absolute temperature to determine activation energies for the dissolution process. The data collected in this study was added to that earlier data and these plots were re-drawn for silica and boron releases in Figures 15 and 16. These plots again show that at higher temperatures the Am/Cm Target dissolution rate levels off or its activation energy decreases while the ARM dissolution increases or its activation energy increases. This behavior is also evident from the plots of the logarithmic rates G in Figures 13 and 14. The logarithmic rates for the Am/Cm Target glass are concave or go through a maximum while the logarithmic rates for ARM are convex or go through a minimum.

Using Equation 2 with the logarithmic rates defined in Equations 3 through 6, 3-D surfaces of the silica and boron leachate concentrations in terms of temperature (°C) and time (days) were constructed for the Am/Cm Target and ARM glasses. Figures 17 and 18 show the silica leachate response for Am/Cm Target and ARM glass, respectively. The symbols in the figures indicate the real data while the surface is generated from the power law dissolution model. Figure 19 shows an overlap of the Am/Cm Target and ARM silica dissolution models. Figures 20 and 21 show the boron leachate response for Am/Cm Target and ARM glass, respectively. Figure 22 shows an overlap of the Am/Cm Target and ARM boron dissolution models. From the plots, one can see that the Am/Cm Target glass is more durable over a given temperature range and time period than ARM glass. In fact, the leachate concentration of boron and silica for Am/Cm Target goes through a maximum value whereas ARM goes through a minimum. The ARM glass leachate concentrations increase with both time and temperature at a faster rate than the Am/Cm Target glass. These results should not be extrapolated beyond the tested data range but certainly indicate that Am/Cm Target is a stable glass in terms of time and temperature response compared to ARM glass.

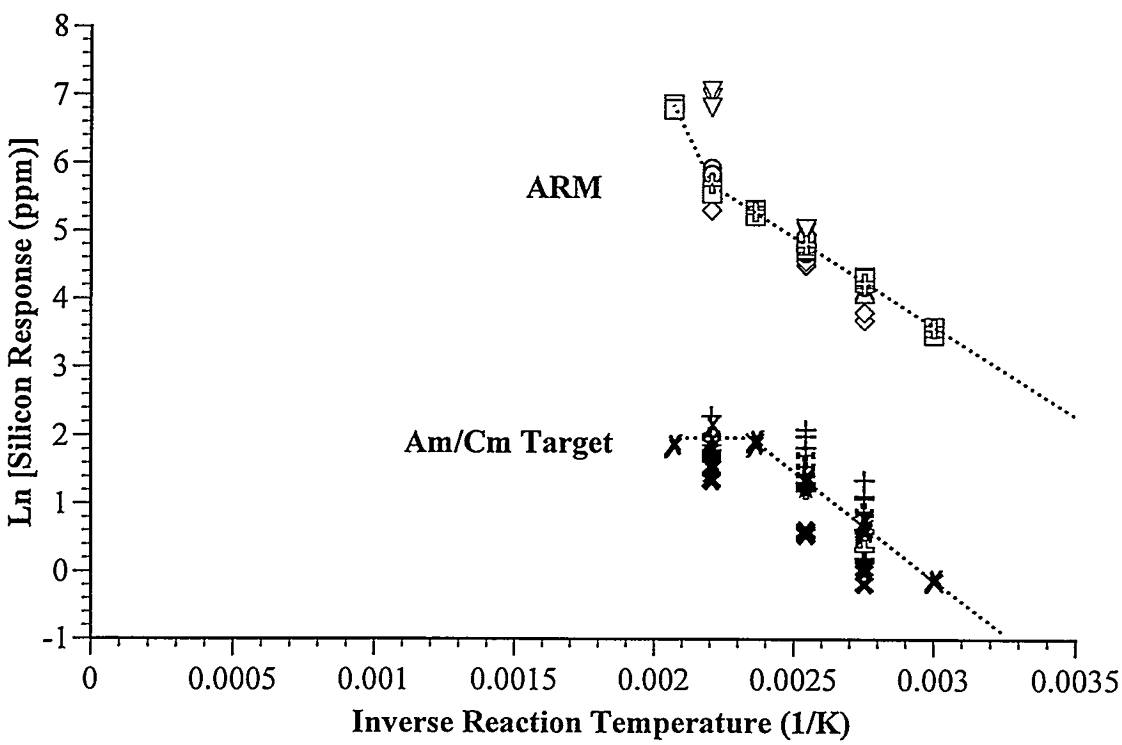

Figure 15. Natural logarithm of PCT silica leachate concentration versus reciprocal of absolute reaction temperature for 1 to 28 days.

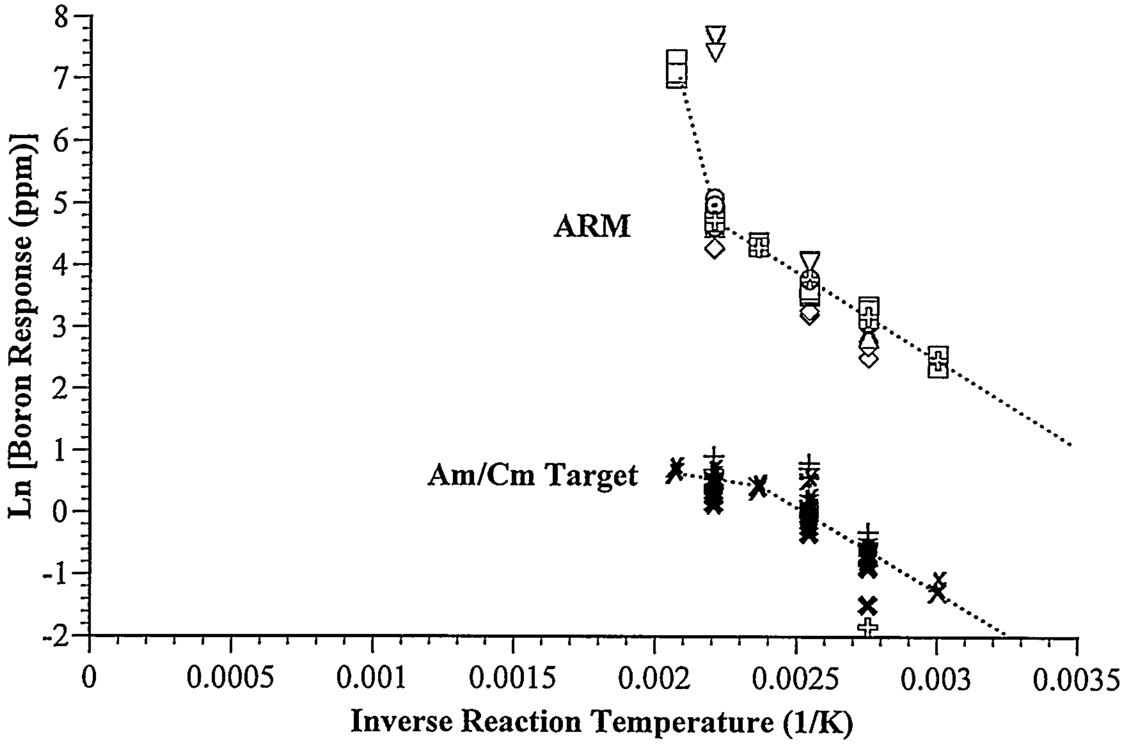

Figure 16. Natural logarithm of PCT boron leachate concentration versus reciprocal of absolute reaction temperature for 1 to 28 days.

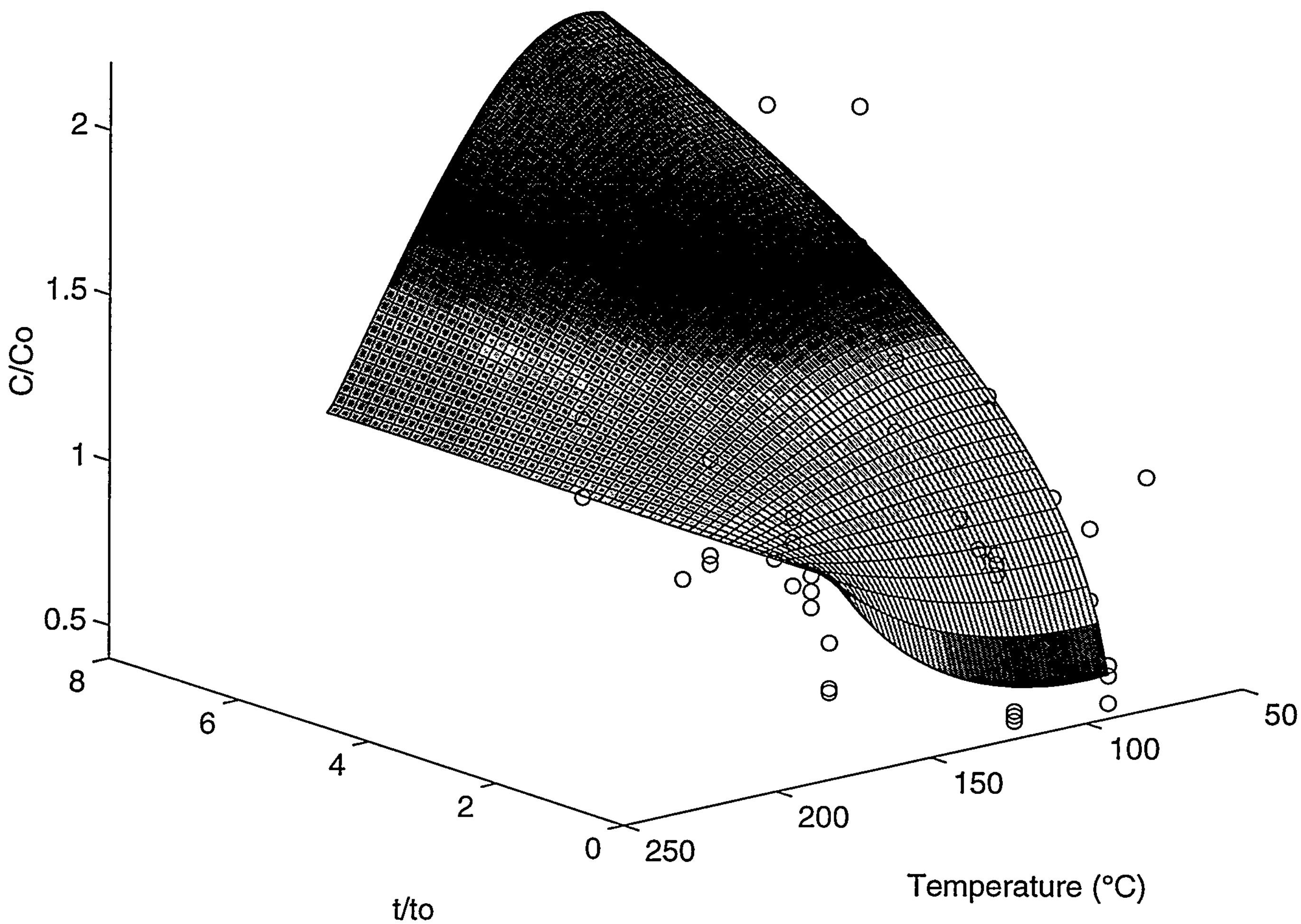

Figure 17. Silica leachate response for Am/Cm Target glass where t_0 is 7 days.

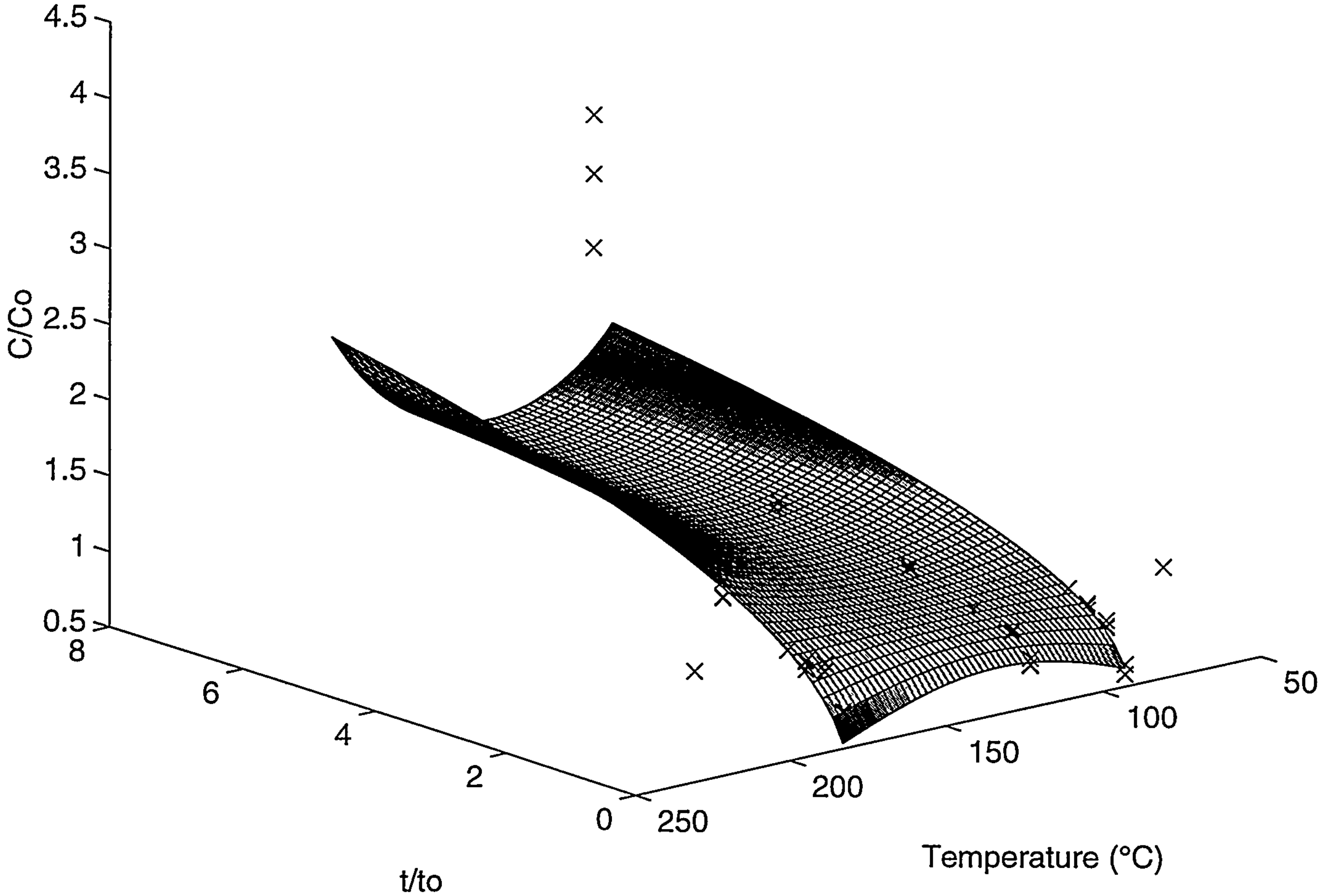

Figure 18. Silica leachate response for ARM glass where t_0 is 7 days.

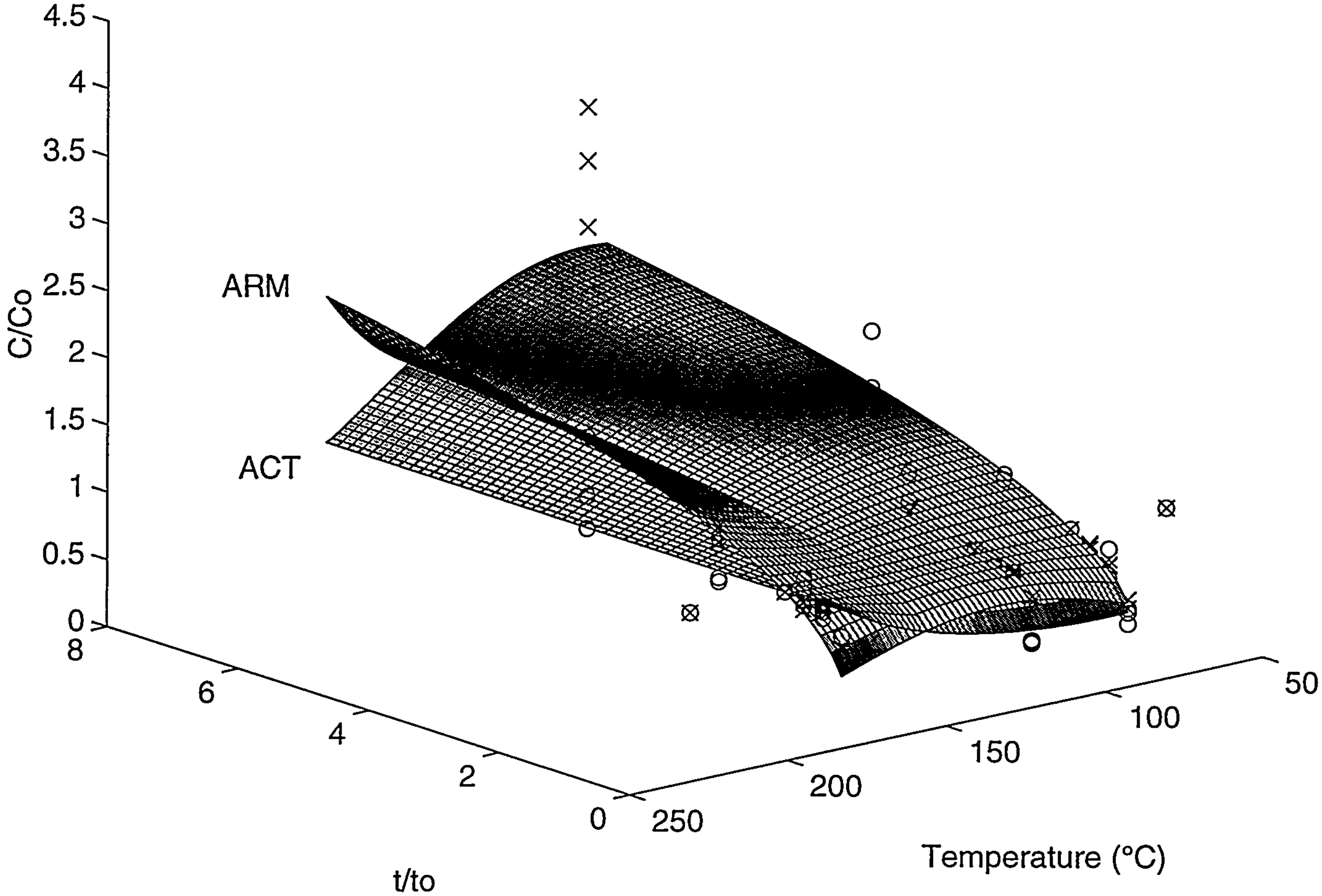

Figure 19. Silica leachate response for Am/Cm Target (ACT) and ARM glasses where t_0 is 7 days.

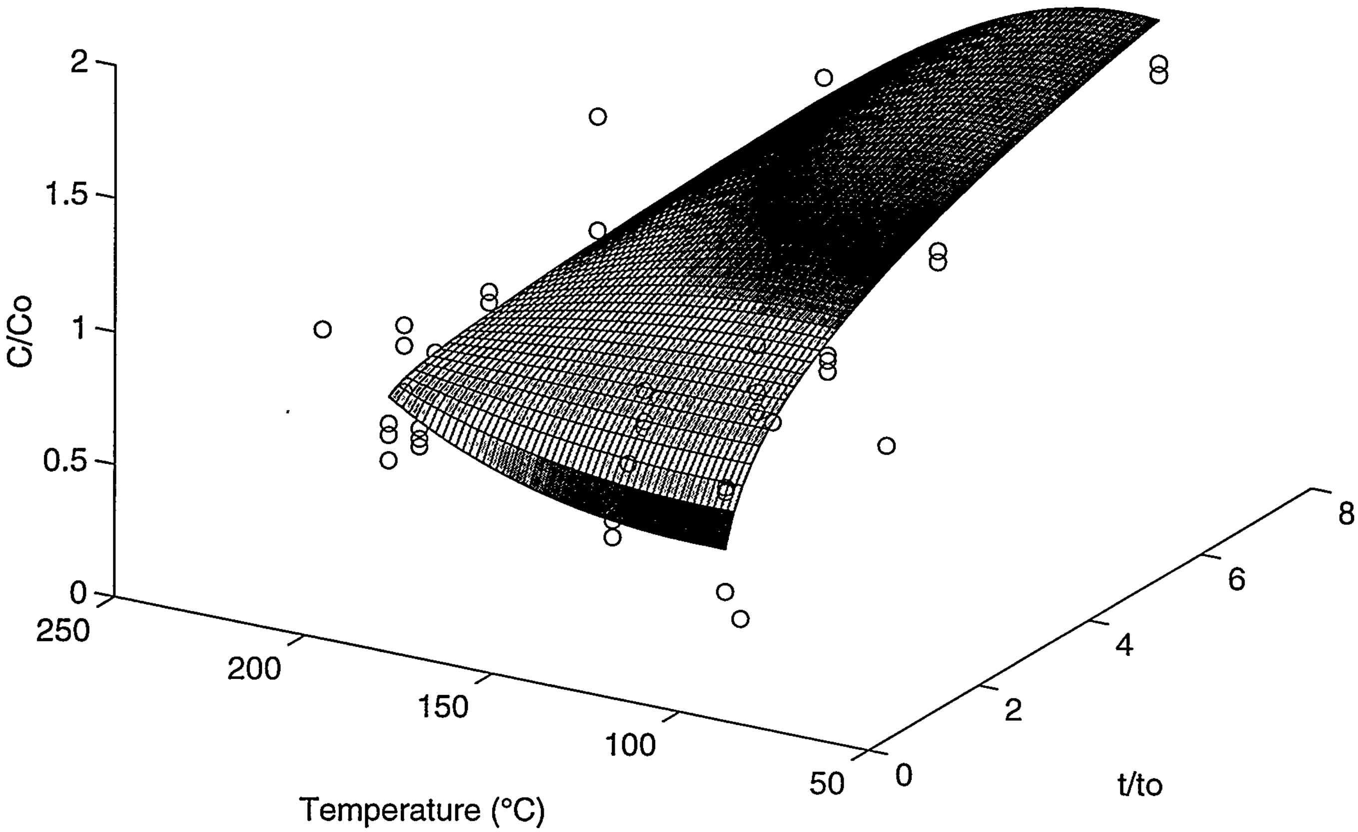

Figure 20. Boron leachate response for Am/Cm Target glass where t_0 is 7 days.

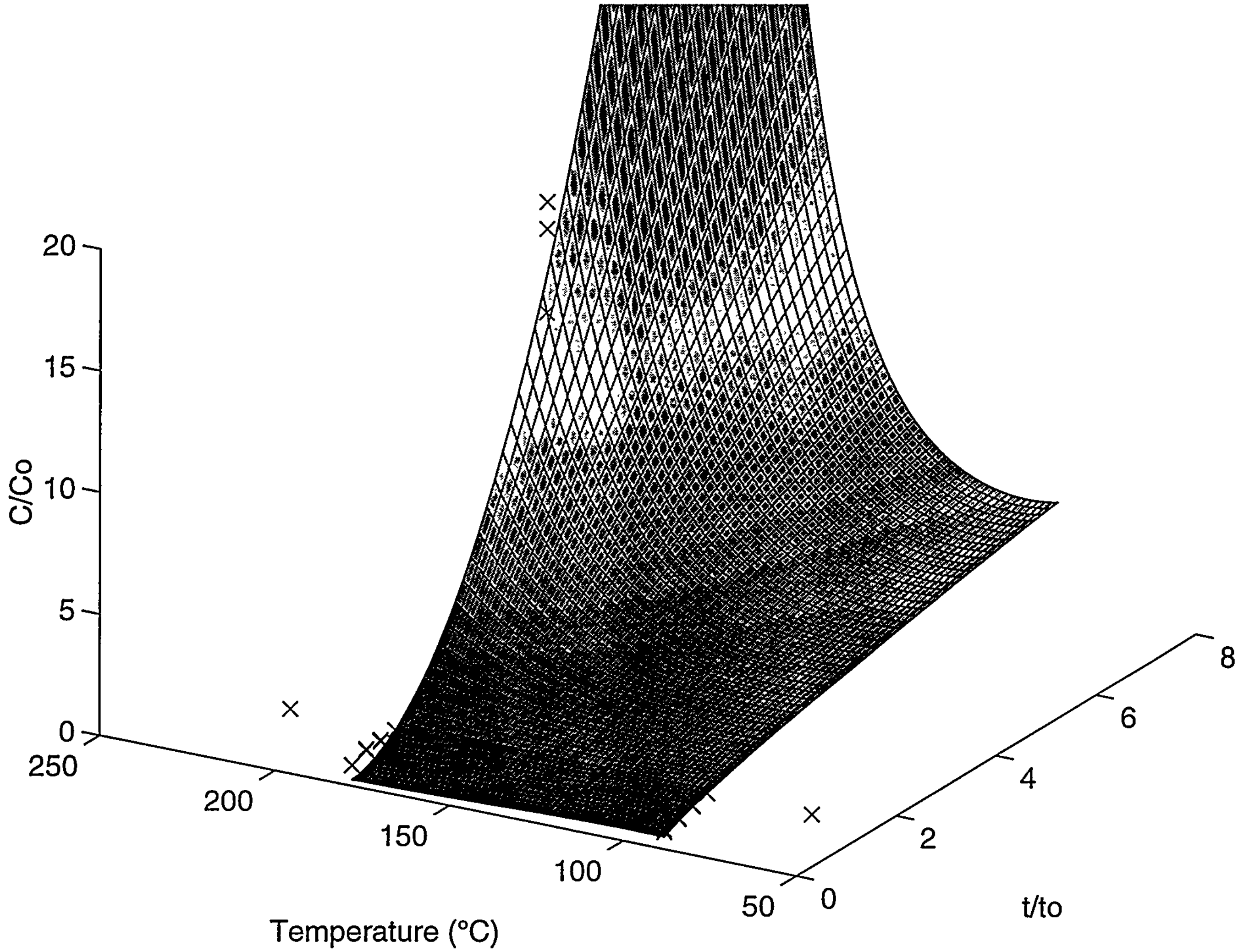

Figure 21. Boron leachate response for ARM glass where t_0 is 7 days.

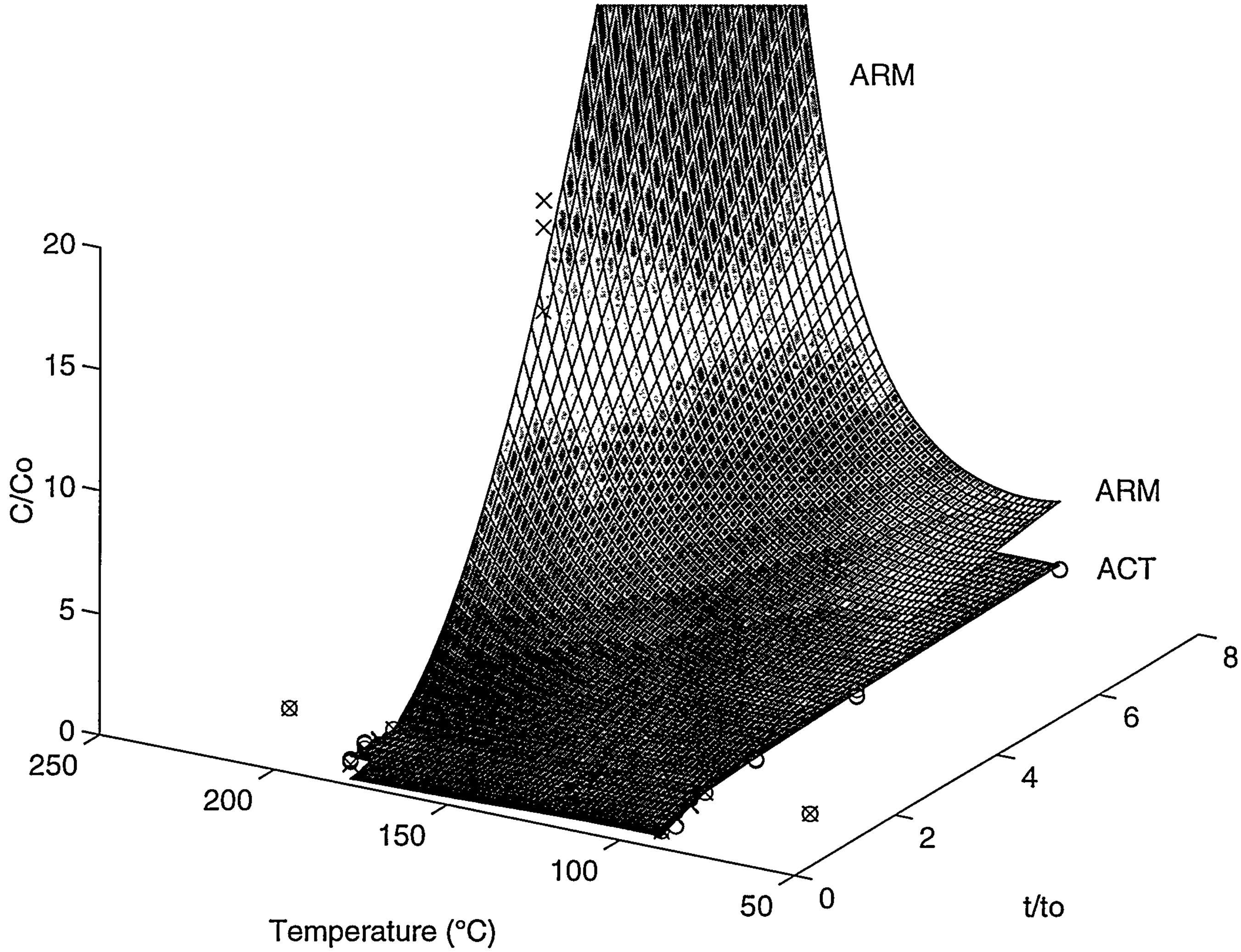

Figure 22. Boron leachate response for Am/Cm Target (ACT) and ARM glasses where t_0 is 7 days.

Durability Dependence on Lanthanide Loading

Surrogate samples were made up at SRTC labs to represent various lanthanide weight percent loading in the Am/Cm Target glass used to vitrify the americium/curium at WSRC. Previous studies on the durability and suitability of the Am/Cm Target glass for this project are given in an earlier report[1]. The surrogate samples were analyzed for elemental content using Inductively Coupled Plasma-Atomic Emission Spectrometer (ICP-AES). From the elemental analyses, the oxide content of the glass samples were then deduced. The compositional data along with other property data for the samples are shown at the end of the report in Table IV. Sample 0 represents a glass composition termed Frit 2000 that was mixed with various amounts of lanthanide to form the other glass samples 1 through 9. Frit 2000 was developed in cooperation with Ferro Corporation to be able to form glass with the current actinide and lanthanide contents of materials on site. At the bottom of Table IV are the weight percent of lanthanide oxides (LnOx) that were added to the frit to form each glass and the total lanthanide oxides present in each glass. Density measurements were performed on these same glass samples to determine the relationship between density and lanthanide loading and are shown in Table IV. These glass samples were also subjected to the ASTM C 1285 Product Consistency Test (PCT) to determine the effect of lanthanide loading on durability[5]. This procedure is described in detail earlier in this report. To summarize, PCT basically consists of crushing and sieving the glass to -100 to 200 mesh, adding ASTM type 1 water, and placing the sample in a sealed container in a oven at a desired temperature for a certain length of time. After the PCT, the samples were submitted to SRTC labs for elemental analyses on an Inductively Coupled Plasma-Atomic Emission Spectrometer (ICP-AES). Figures 23 and 24 show the silica and boron releases as a function of lanthanide oxide content of the Am/Cm Target samples. The average release value for ARM glass is also shown on these figures as a horizontal dashed line but its value is at a fixed lanthanide composition which defines ARM glass. These elements are the most soluble and are generally used as indicators of a glass's durability. To compare glasses on a consistent basis, normalized releases are used which take into account the different weight loading of each element. These values are plotted in Figures 25 and 26. Since there were three vials taken per glass sample, there are generally three values shown per lanthanide oxide loading in the figures and Table III. From this data it can be seen that the Am/Cm Target glass normalized releases are orders of magnitude less than the Approved Reference Material (ARM) boron values. More importantly, the durability of the Am/Cm Target glass is unaffected by the lanthanide loading and can be removed from the set of control variables.

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[5] *ASTM C 1285-94*, Annual Book of ASTM Standards, American Society for Testing and Materials, Committee c-26 on Nuclear Fuel Cycle, Subcommittee c26.13 on Repository Waste Package Materials Testing, Volume 12.01, 1994.

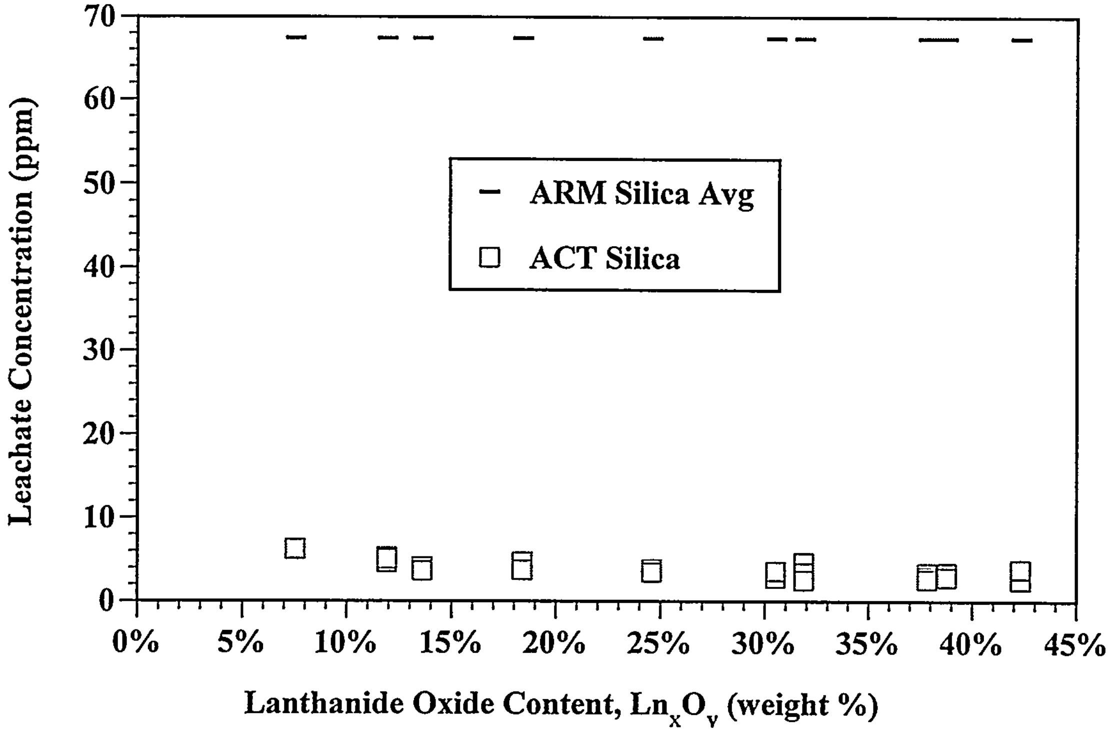

Figure 23. ASTM C-1285 leachate silica concentrations for Am/Cm Target (ACT) and Approved Reference Material (ARM) glasses.

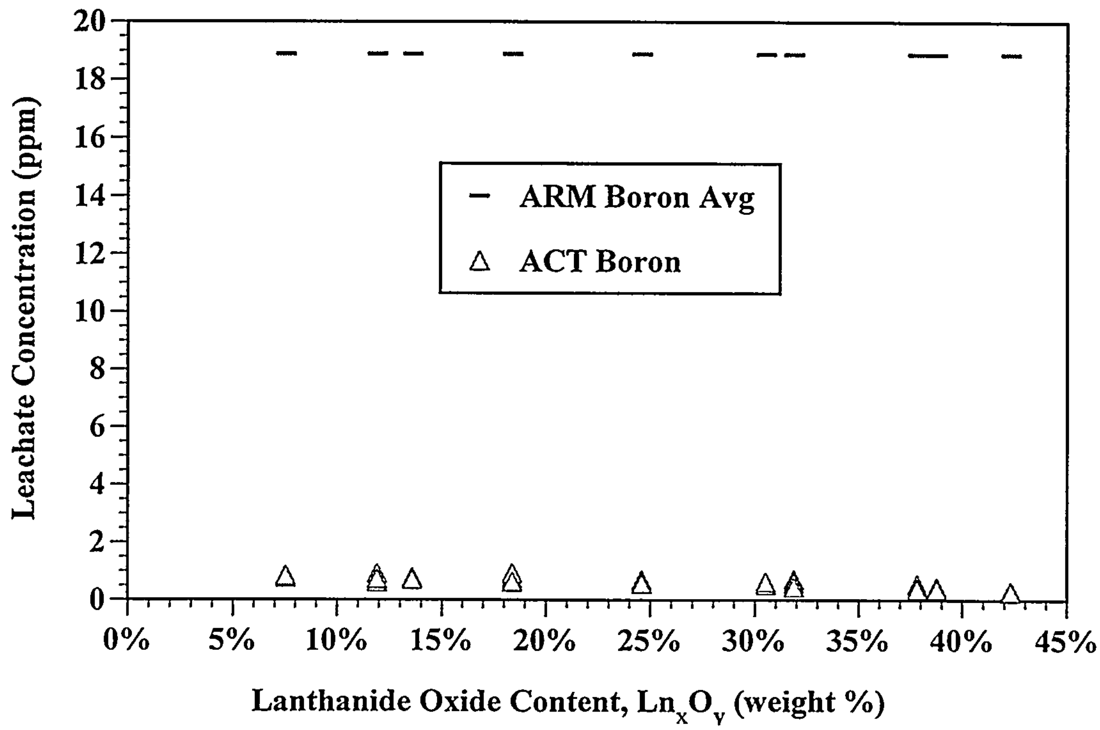

Figure 24. ASTM C-1285 leachate boron concentrations for Am/Cm Target (ACT) and Approved Reference Material (ARM) glasses.

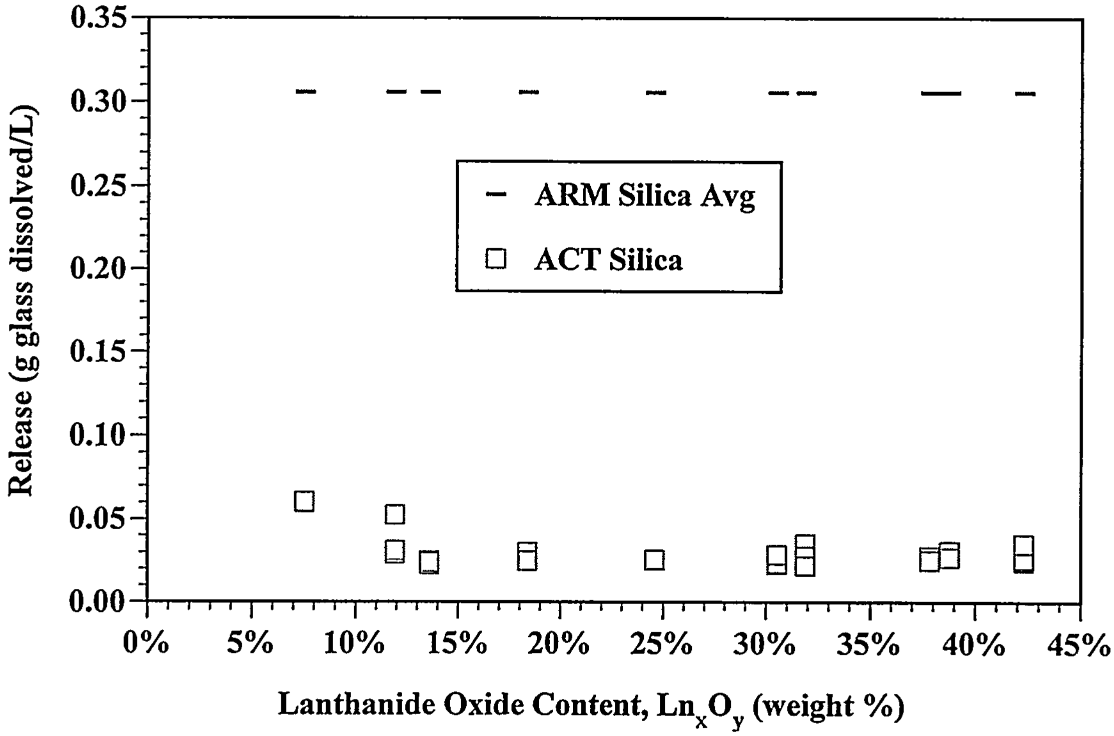

Figure 25. Normalized silica releases for Am/Cm Target (ACT) and Approved Reference Material (ARM) glasses based on ASTM C-1285 procedure.

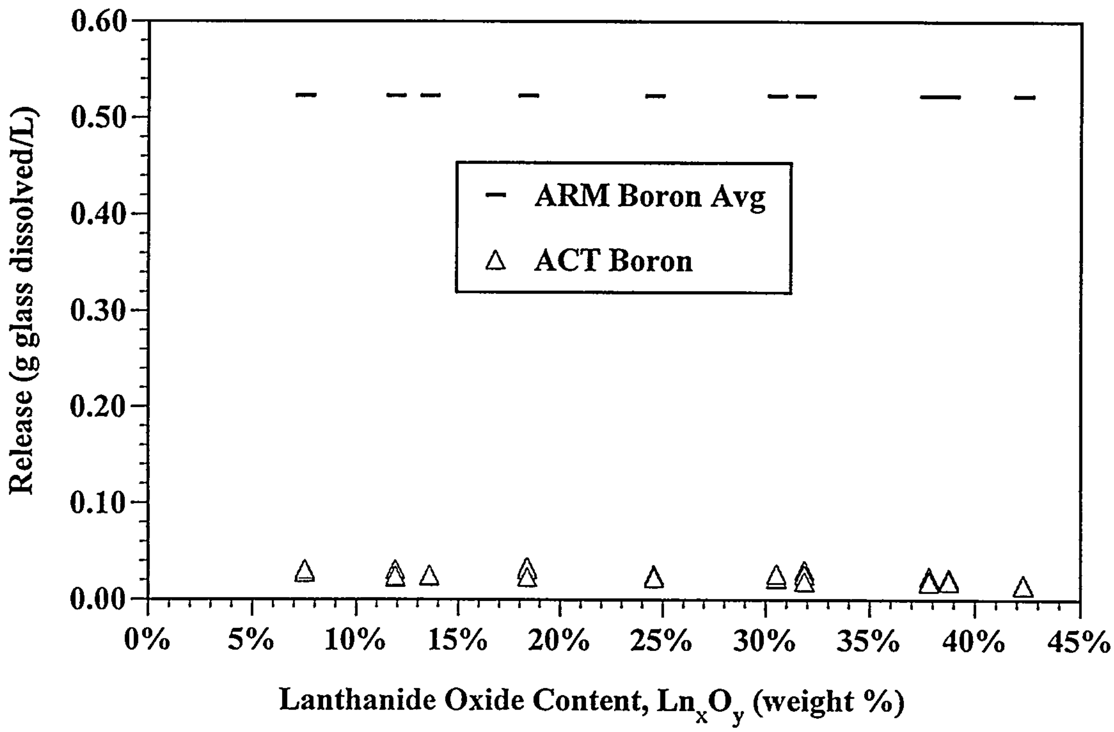

Figure 26. Normalized boron releases for Am/Cm Target (ACT) and Approved Reference Material (ARM) glasses based on ASTM C-1285 procedure.

Table IV. Am/Cm Target Glass Sample Property Data

	Sample 0	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5	Sample 6	Sample 7	Sample 8	Sample 9
SiO_2	43.77%	41.60%	41.82%	39.67%	36.69%	33.75%	33.11%	30.15%	30.07%	28.87%
B_2O_3	8.79%	9.57%	9.16%	8.59%	7.95%	7.40%	7.21%	6.73%	6.46%	5.93%
BaO	9.69%	8.64%	8.37%	7.89%	7.31%	6.74%	6.62%	6.17%	6.15%	5.92%
Al_2O_3	6.01%	5.82%	5.59%	5.17%	4.68%	4.17%	4.10%	3.61%	3.62%	3.72%
PbO	24.15%	22.45%	21.47%	20.29%	18.79%	17.42%	17.09%	15.50%	14.93%	13.26%
La_2O_3	7.53%	7.58%	7.66%	8.10%	8.69%	9.18%	9.34%	9.91%	10.00%	10.43%
Eu_2O_3	0.00%	0.08%	0.12%	0.21%	0.33%	0.44%	0.47%	0.57%	0.58%	0.64%
Sm_2O_3	0.00%	0.53%	0.73%	1.25%	1.93%	2.58%	2.74%	3.34%	3.43%	3.77%
Pr_2O_3	0.00%	0.84%	1.17%	2.03%	3.13%	4.20%	4.43%	5.43%	5.64%	6.25%
Gd_2O_3	0.00%	0.21%	0.29%	0.51%	0.79%	1.07%	1.13%	1.37%	1.39%	1.53%
Nd_2O_3	0.00%	1.81%	2.44%	4.21%	6.49%	8.72%	9.24%	11.61%	11.92%	13.21%
CeO_2	0.00%	0.85%	1.18%	2.06%	3.21%	4.32%	4.52%	5.59%	5.81%	6.47%
LnOx Added	0.00%	4.75%	6.80%	11.98%	18.55%	24.87%	26.60%	32.93%	34.25%	38.16%
LnOx Total	7.53%	11.91%	13.58%	18.38%	24.58%	30.52%	31.87%	37.83%	38.77%	42.30%
Density (g/cm^3)	3.13	3.33	3.39	3.52	3.72	3.78	3.91	4.08	4.18	4.26
Si Release (ppm)	6.183 6.233 5.293	4.643 5.032 3.826	4.027 3.610 3.889	4.678 3.728 3.824	* 3.463 2.987	2.797 3.580 4.611	3.410 2.500 3.382	2.933 2.603 3.023	3.423 2.853 2.513	2.543 3.813 2.323
B Release (ppm)	0.771 0.843 0.620	0.917 0.734 0.719	0.795 0.706 0.752	0.921 0.634 0.699	0.592 0.571 0.544	0.530 0.623 0.718	0.597 0.441 0.554	0.418 0.382 0.453	0.458 0.385 0.325	0.309 0.276 0.319
Si Normalized Release (g glass/L)	0.0600 0.0604 0.0526	0.0289 0.0309 0.0247	0.0225 0.0239 0.0278	0.0300 0.0249 0.0255	* 0.0254 0.0227	0.0237 0.0285 0.0350	0.0277 0.0219 0.0275	0.0274 0.0251 0.0280	0.0308 0.0269 0.0245	0.0258 0.0350 0.0242
B Normalized Release (g glass/L)	0.0282 0.0309 0.0227	0.0308 0.0246 0.0241	0.0238 0.0253 0.0310	0.0339 0.0233 0.0257	0.0235 0.0227 0.0216	0.0227 0.0267 0.0308	0.0260 0.0192 0.0241	0.0197 0.0180 0.0214	0.0225 0.0189 0.0159	0.0165 0.0148 0.0171

*not applicable

Conclusions

The experimental studies on Am/Cm Target glass have shown that the concentration of PCT leachate species is both time and temperature dependent. This concentration dependence can be categorized as the following power-law relation:

(2) $$C(t) = C_0 \bullet \left[\frac{t}{t_0} \right]^{G(T)}$$

where C is the concentration at time t, C_0 is the concentration at t_0, t is the time in days, t_0 represents 7 days, and G(T) is the logarithmic release rate, which is temperature dependent. From PCT data collected and analyzed for a particular glass at various times and temperatures, the logarithmic rate expressions G(T) can be derived. Using this leachate concentration model, 3-D surfaces in terms of time and temperature were constructed for Am/Cm Target and ARM glasses. From these plots, it was seen that the Am/Cm Target glass is more durable over a given time and temperature range than the ARM glass. In fact, the leachate concentration of the ARM glass increases exponentially with time and temperature. In contrast, the Am/Cm Target glass leachate concentrations level off as time and temperature increase. This type of behavior was evidenced in earlier experiments as well. Not only is the Am/Cm Target durability robust in terms of time and temperature, but experiments were conducted that showed the durability was essentially independent of the lanthanide oxide loading in the glass. All these findings were important in that the durability of the Am/Cm Target glass would not be a concern in the operation and control of the Am/Cm vitrification process. Other experiments are being conducted to examine other properties of the Am/Cm Target glass, including liquidus temperature, density, and viscosity. The results from these studies will be used in upcoming projects at WSRC involving the vitrification of actinide and lanthanide materials.