Supporting Information for:

Volatility and Extractability of Strontium-85, Cesium-134, Cobalt-57, and Uranium After Heating Hardened Portland Cement Paste

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Gamma activity assay techniques - All radioisotopes were assayed using a well-type 3-in. diameter NaI (Tl-activated) gamma scintillation detector coupled with a multichannel analyzer (APHA 1995) controlling a Nuclear Data Model 5400 automatic changer for standard 25-mL scintillation vials. Although resolution of compound gamma-ray spectra, arising from multiple gamma-emitting isotopes within a single sample, is experimentally feasible using the available multichannel analyzer software employed (Accuspec©Canberra Nuclear Products Inc., Version 03), this technique would introduce inherent variability in detection limits and counting rate uncertainties among samples depending on the relative activities of the various isotopes. Thus, in the present study, the various spiked cement pastes and their resulting extracts contained only one isotope per sample. Three regions-of-interest (channel intervals) were established within the 2048 channels of the 0.05 to 2000 MeV gamma-ray energy range and were regularly collected covering various gamma-ray emission energies of each isotope including 0.05-0.39 MeV (region 1, channel numbers 47-350 for ^{57}Co and U), 0.42-0.65 MeV (region 2, channel numbers 384-594 for ^{85}Sr), and 0.43-1.62 MeV (region 3, channel numbers 384-1477 for ^{134}Cs). For ^{85}Sr, ^{57}Co, and ^{134}Cs, counting intervals were typically 10 minutes, which, in most cases, were adequate to

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attain an uncertainty in the gross count rate of <0.1% (± 1σ). Because of uranium's lower specific activity, counting intervals of 1-4 h were generally used to attain uncertainty in the counting rate of <1%. The channel numbers of each isotope's characteristic gamma ray(s) were checked daily for possible drift in their position but none was observed during the study interval (12 months). For ^{85}Sr (region 2), ^{57}Co (region 1), ^{134}Cs (region 3), and U (also region 1), initial gross counting rates of 20,000, 80,000, 100,000, and 2,300 counts per minute (cpm) were typical. Background counting rates in the same regions-of-interest were collected from unspiked but otherwise identical cement paste powders in crucibles and were subtracted from the gross counting rates in spiked samples; for the regions of interest for ^{85}Sr, ^{57}Co, ^{134}Cs, and U, background counting rates were typically 194, 1640, 450, and 1640 cpm, respectively, varying only slightly daily due to activities of other proximate samples within the sample changer queue. Background counting rates in all three regions-of-interest for samples containing either crucibles-plus-cement pastes, 20-mL of water, or empty scintillation vials were indistinguishable indicating that no experimental materials or containers introduced detectable gamma emissions. For extracts of cement powders, 20-mL volumes of solution in scintillation vials were employed for the same gamma spectrum regions-of-interest and the same counting intervals; typically, for all four isotopes, counting rates in 20-mL of aqueous solution in a scintillation vial were approximately 94% of the same activity contained in 1-g of cement paste within an alumina crucible in an identical scintillation vial. For both pulverized cement paste in crucibles and 20-mL liquid volumes, standard untreated spiked samples were prepared in triplicate for each radioisotope; these functioned as relative controls (containing maximal starting activity spiked material) to which subsequently heated or extracted samples could be compared (as fractions of

this starting spiked activity) using the ratio of total net counts in the region-of-interest. Under the identical detection geometries employed (either 1-g of cement paste within a crucible in a scintillation vial or 20-mL of aqueous liquid within a scintillation vial) and the daily use of sealed single-isotope standards, containing identical starting activities, to compute ratios, decay corrections within a given daily group of samples were unnecessary. Computer files of complete gamma-ray spectra were saved in ASCII format for every sample and standard counted, as backup in case inconsistencies, ambiguities were observed in regions-of-interest summary data output; routinely, only regions-of-interest gross count totals were processed along with sample identification and date and time of counting.

APHA (American Public Health Association). 1995. 7120 B. Gamma Spectroscopic Method. pp. 7-18-7-20, In, Standard Methods for the Examination of Water and Wastewater. American Public Health Association, Washington, D.C.

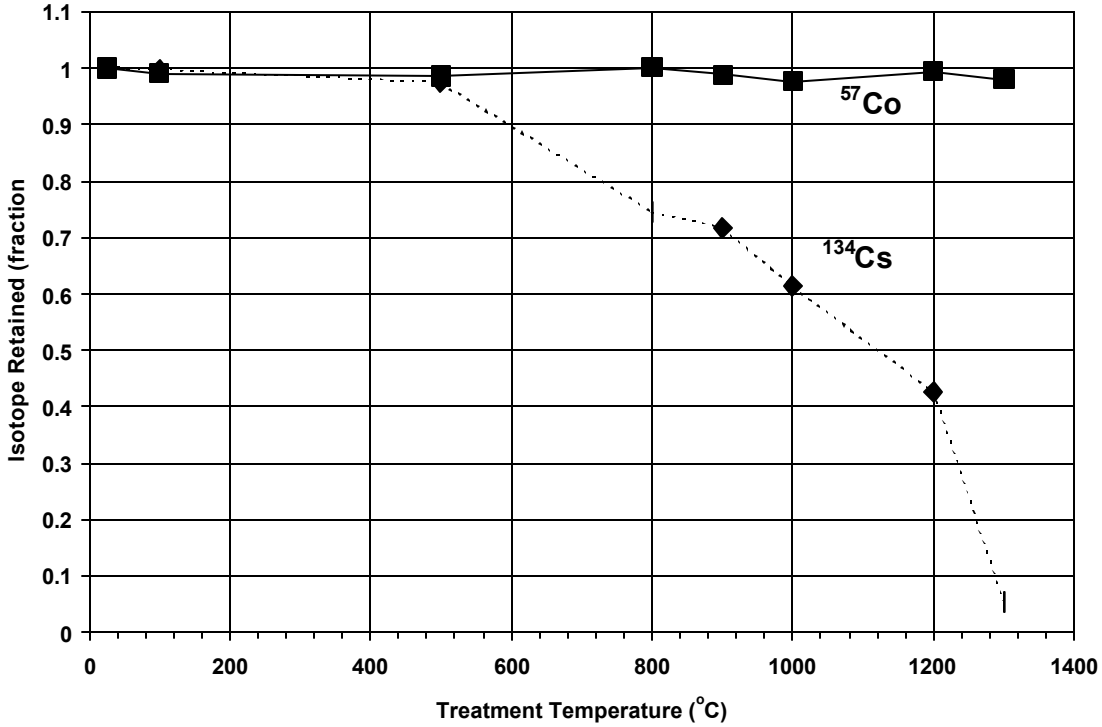

Figure S1. Retention of radioisotopes by cohesive cement paste plugs when heated sequentially to various final temperatures.

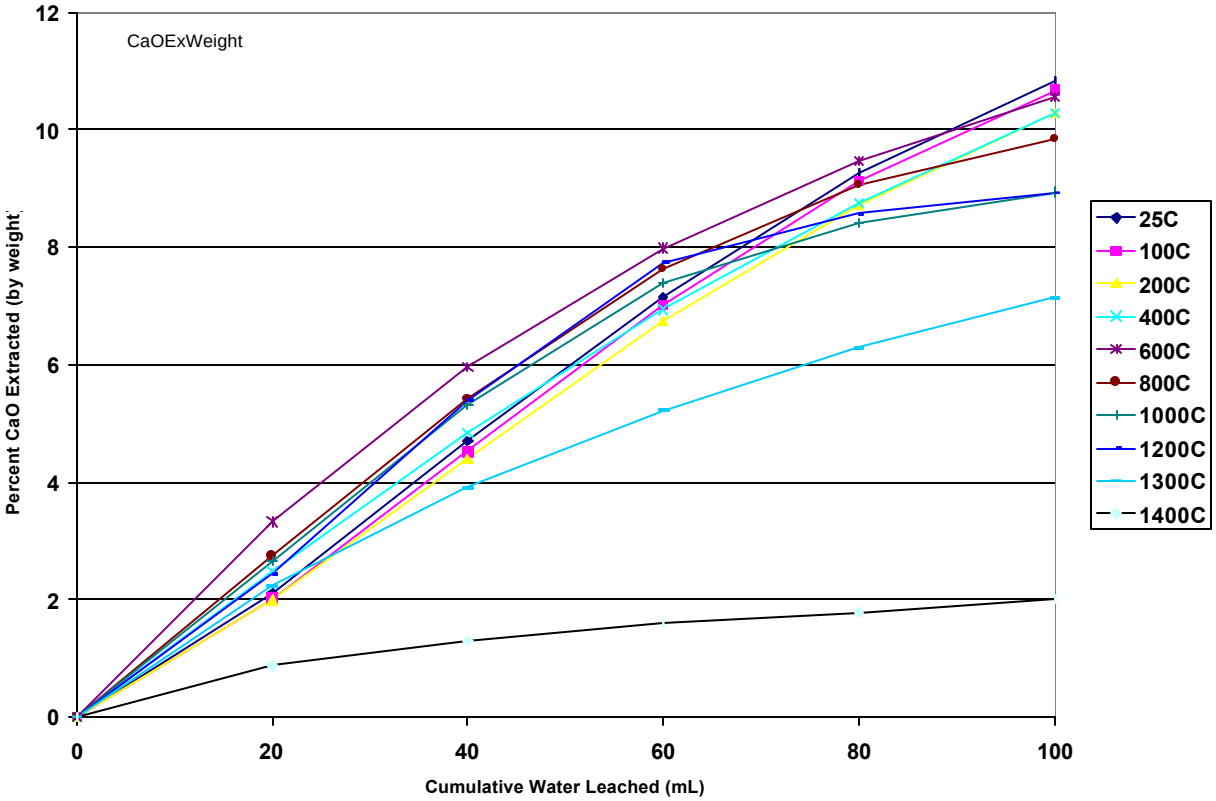

Figure S2. Extraction of CaO weight equivalent by water from pulverized Portland cement paste after heating to various temperatures.

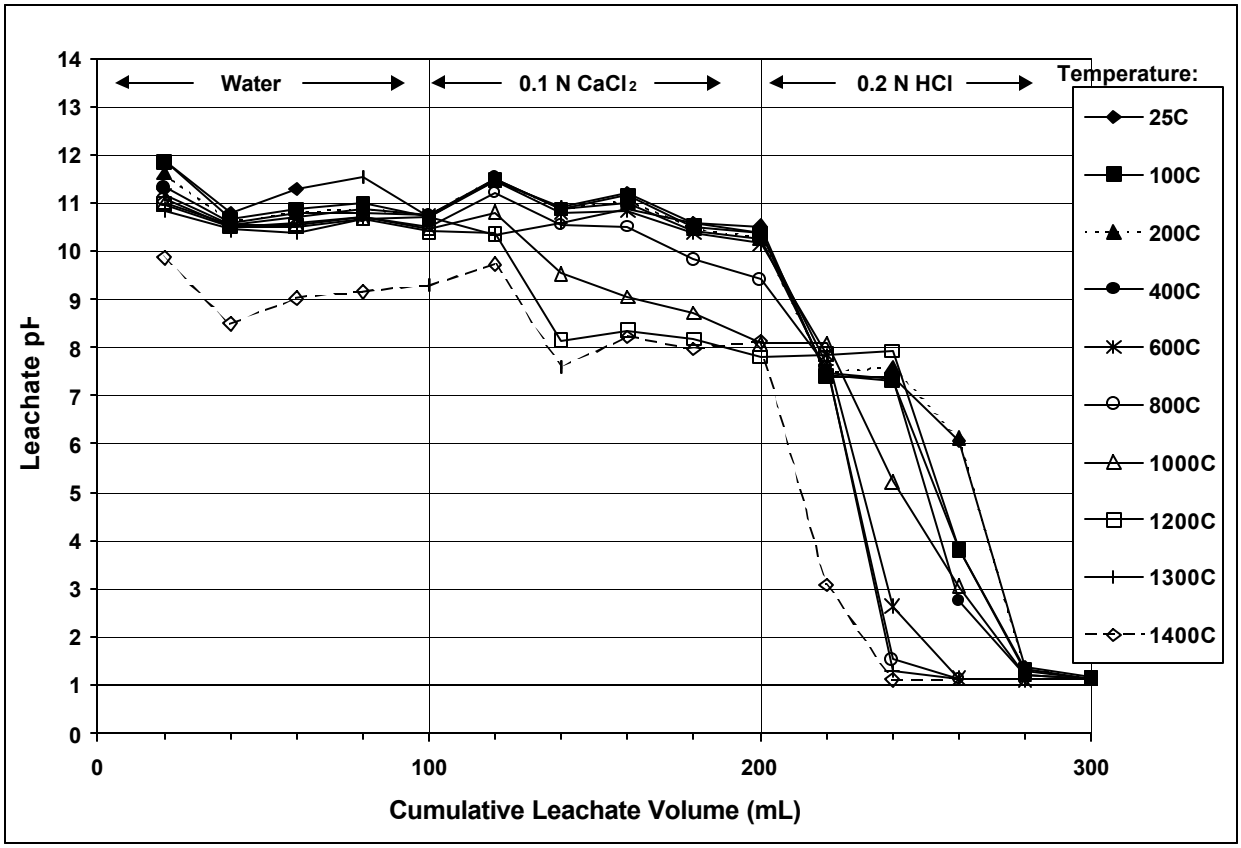

Figure S3. The average pH of sequential extracts of pulverized Portland cement pastes after heating to various temperatures.

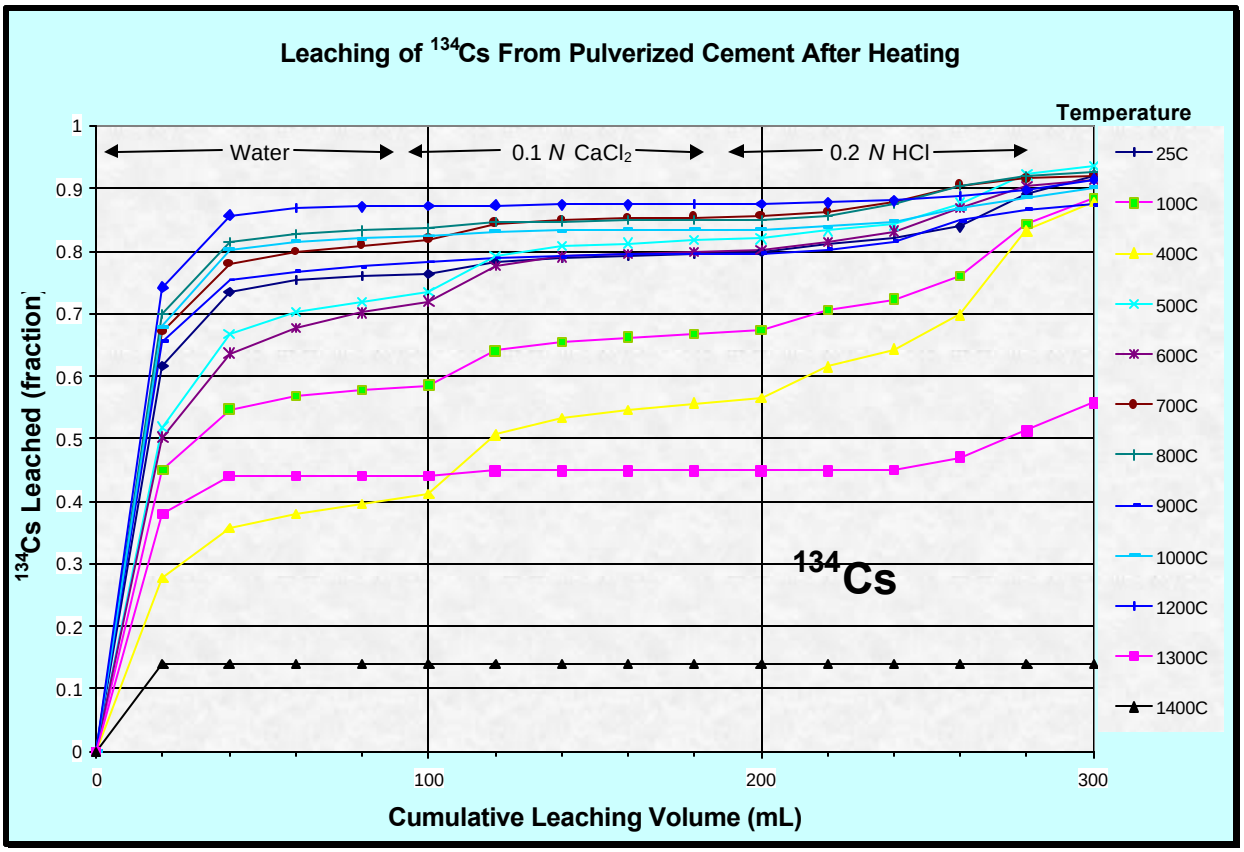

Figure S4. Sequential extraction profiles of ^{134}Cs from pulverized Portland cement paste following heating to various temperatures.

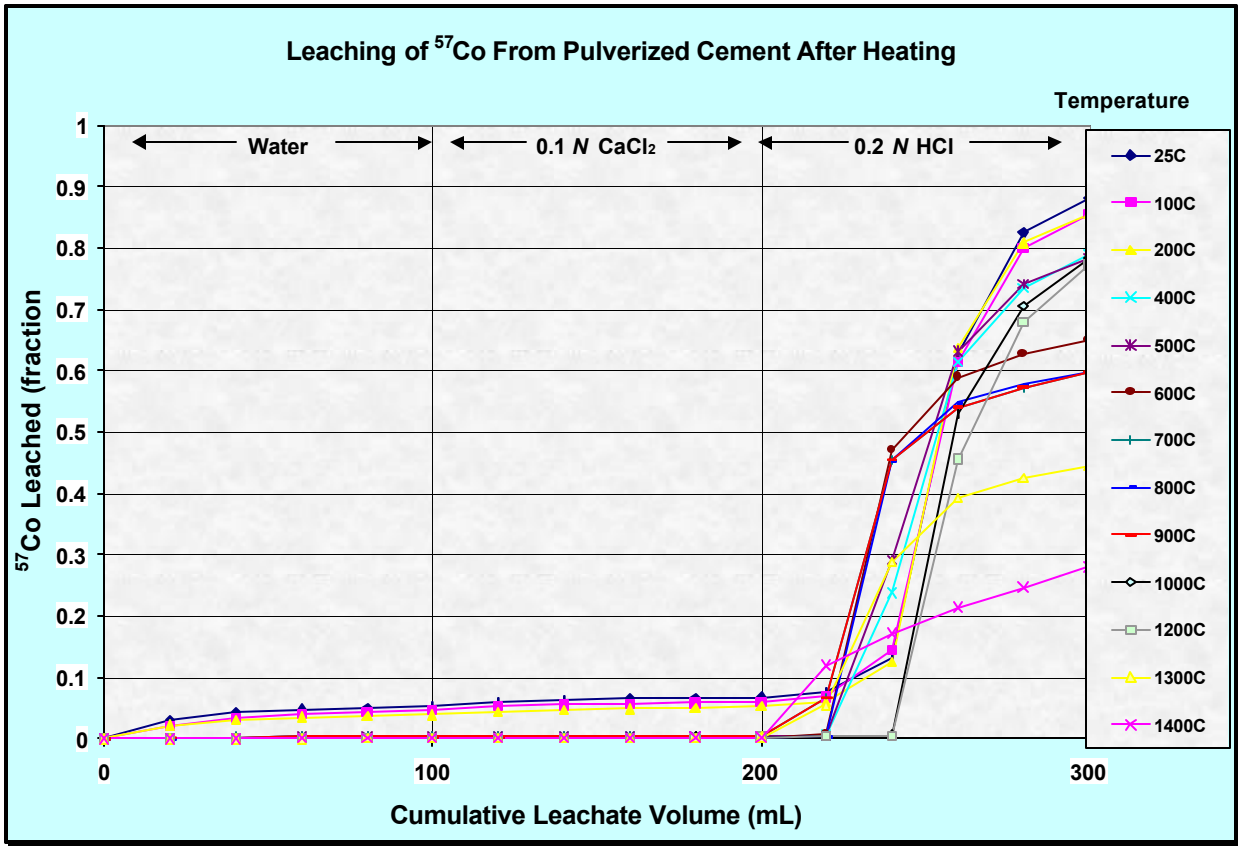

Figure S5. Sequential extraction profiles for ^{57}Co from pulverized Portland cement paste following heating to various temperatures.

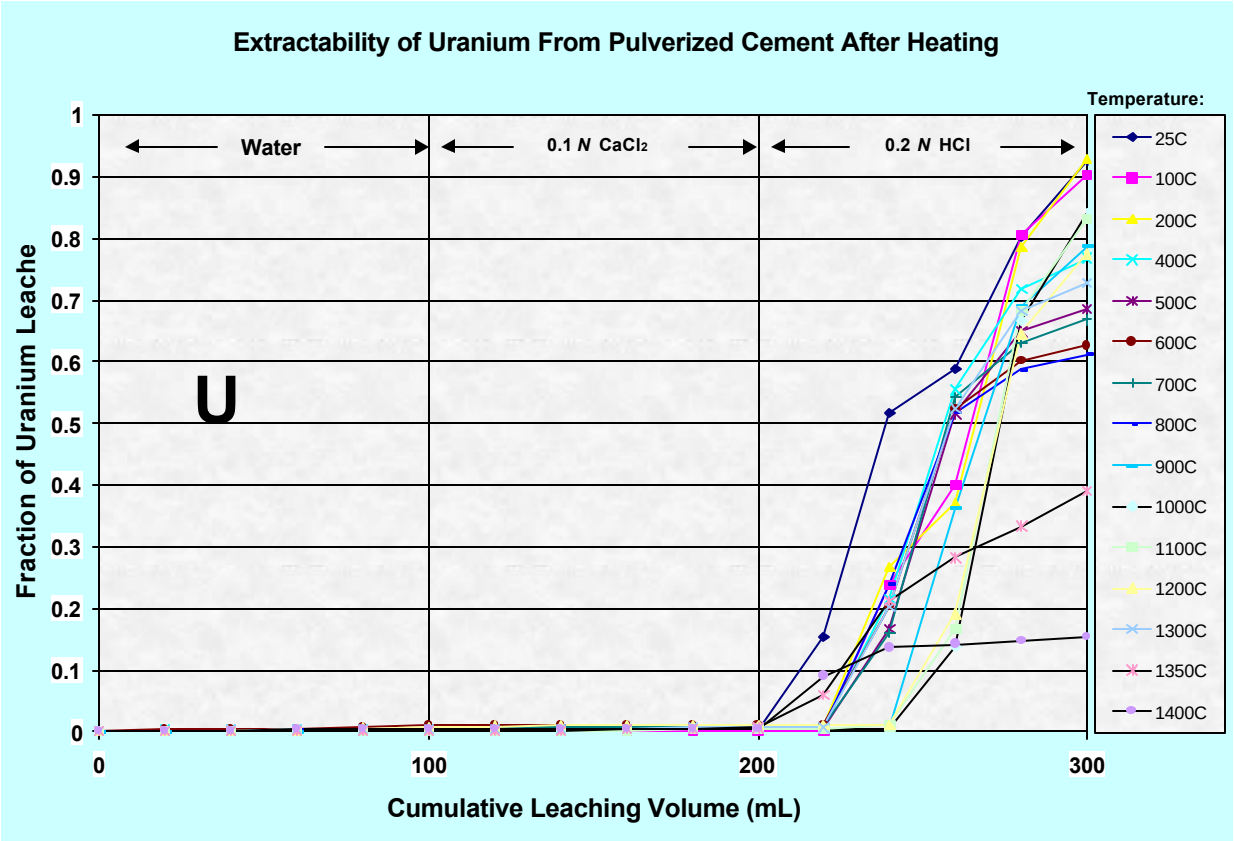

Figure S6. Sequential extraction profiles for uranium (total concentration = 9,977 µg/g) from pulverized Portland cement paste following heating to various temperatures.

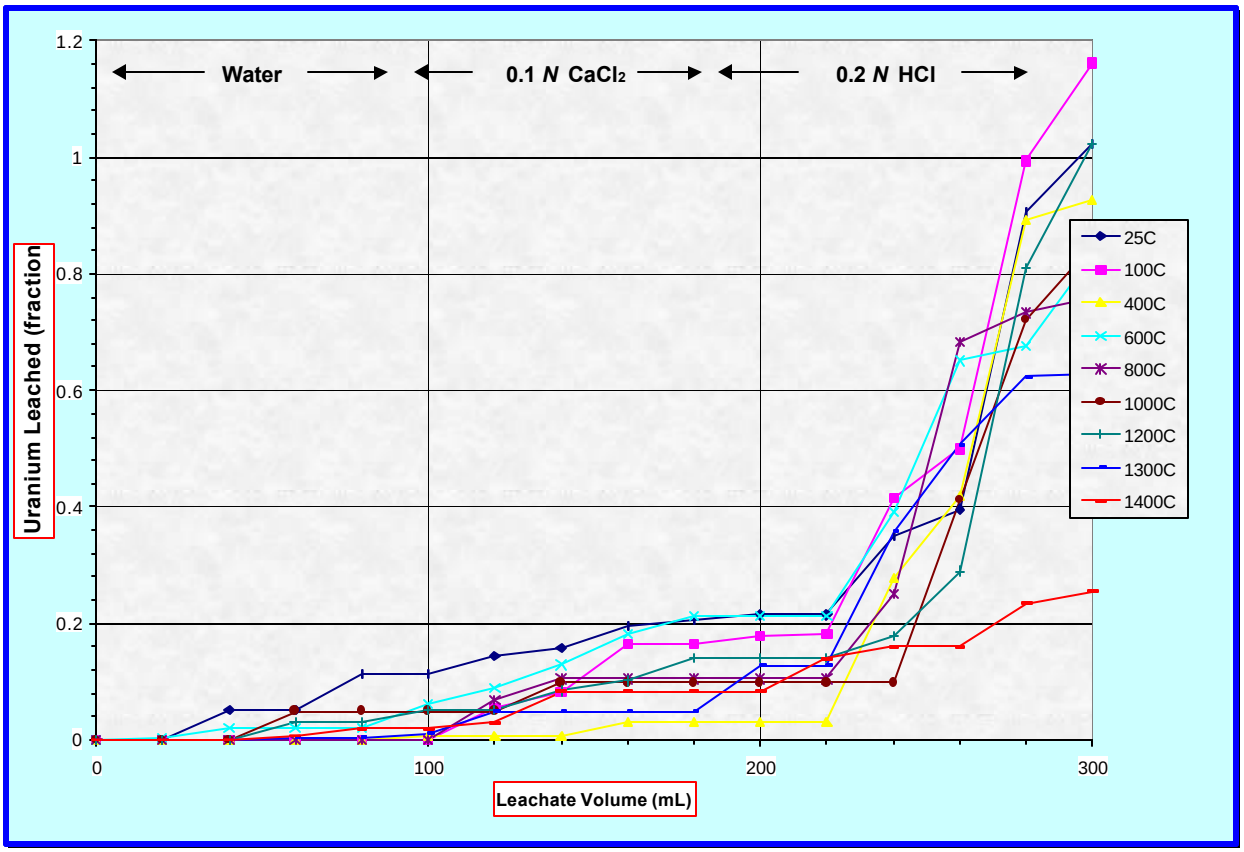

Figure S7. Sequential extraction profiles for uranium (total concentration = 997 µg/g) from pulverized Portland cement paste following heating to various temperatures.

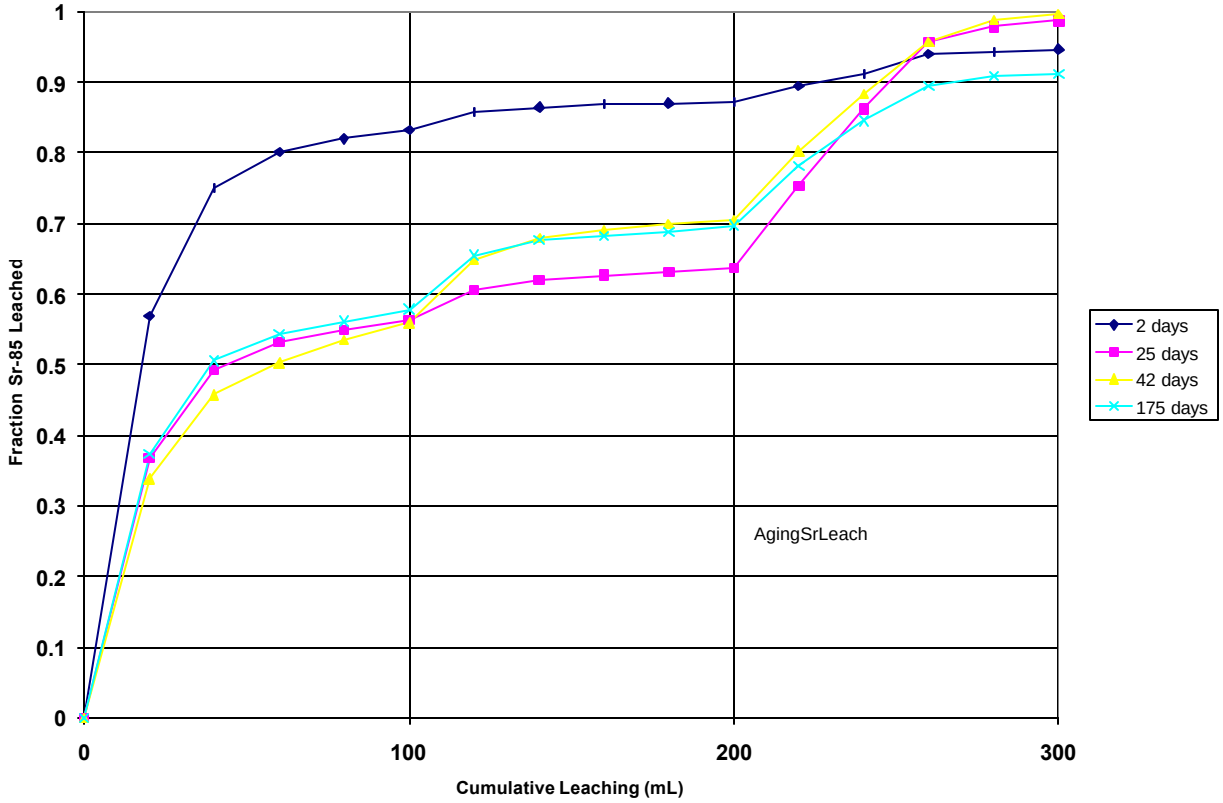

Figure S8. Sequential extraction profiles for ^{85}Sr from cured but unheated Portland cement paste after contact periods of 2, 25, 42, and 175 days.

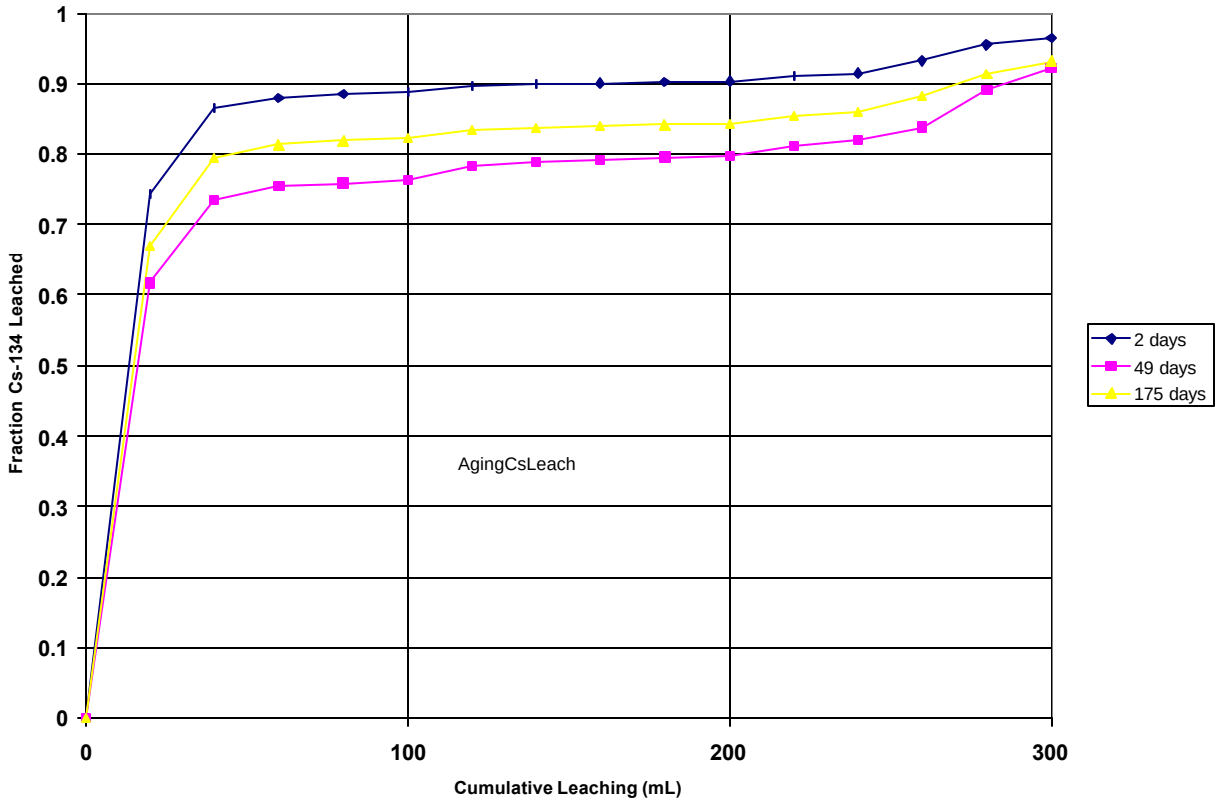

Figure S9. Sequential extraction profiles for ^{134}Cs from cured but unheated pulverized Portland cement paste after contact periods of 2, 49, and 175 days.

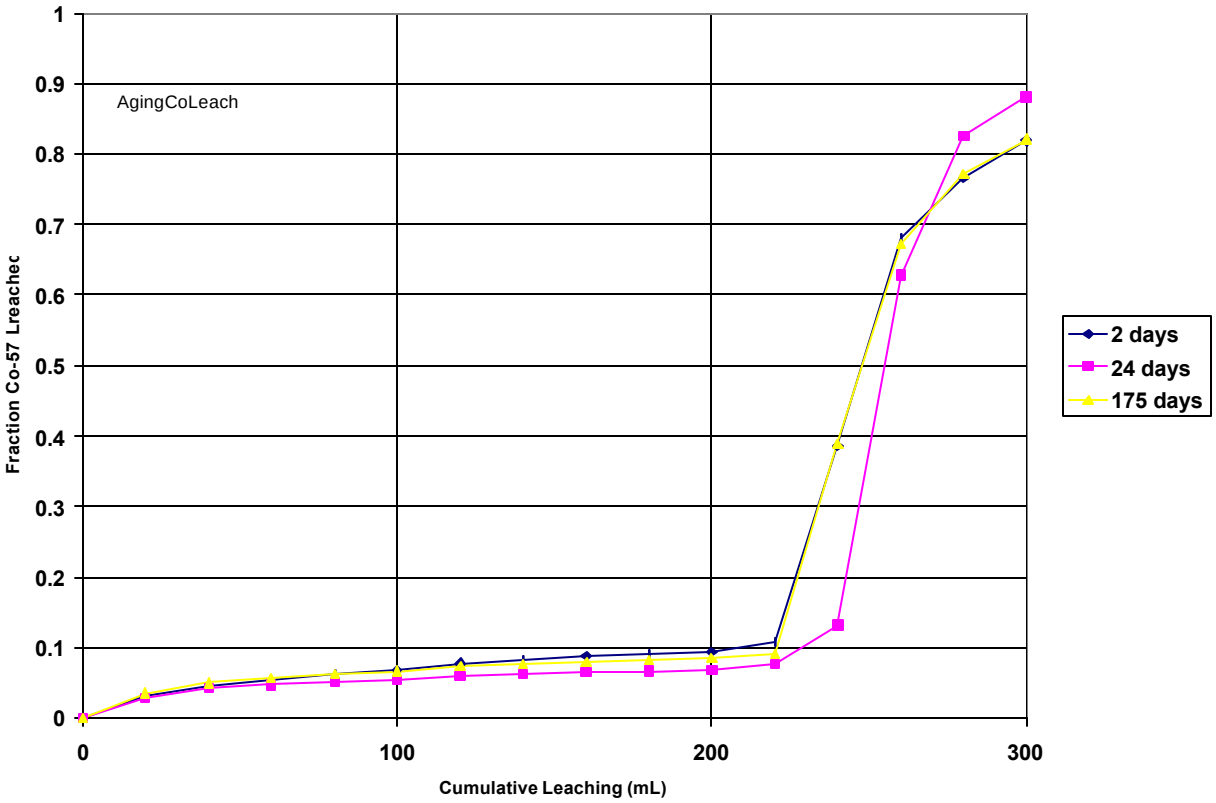

Figure S10. Sequential extraction profiles of ^{57}Co from cured but unheated pulverized Portland cement paste after contact periods of 2, 24, and 175 days.

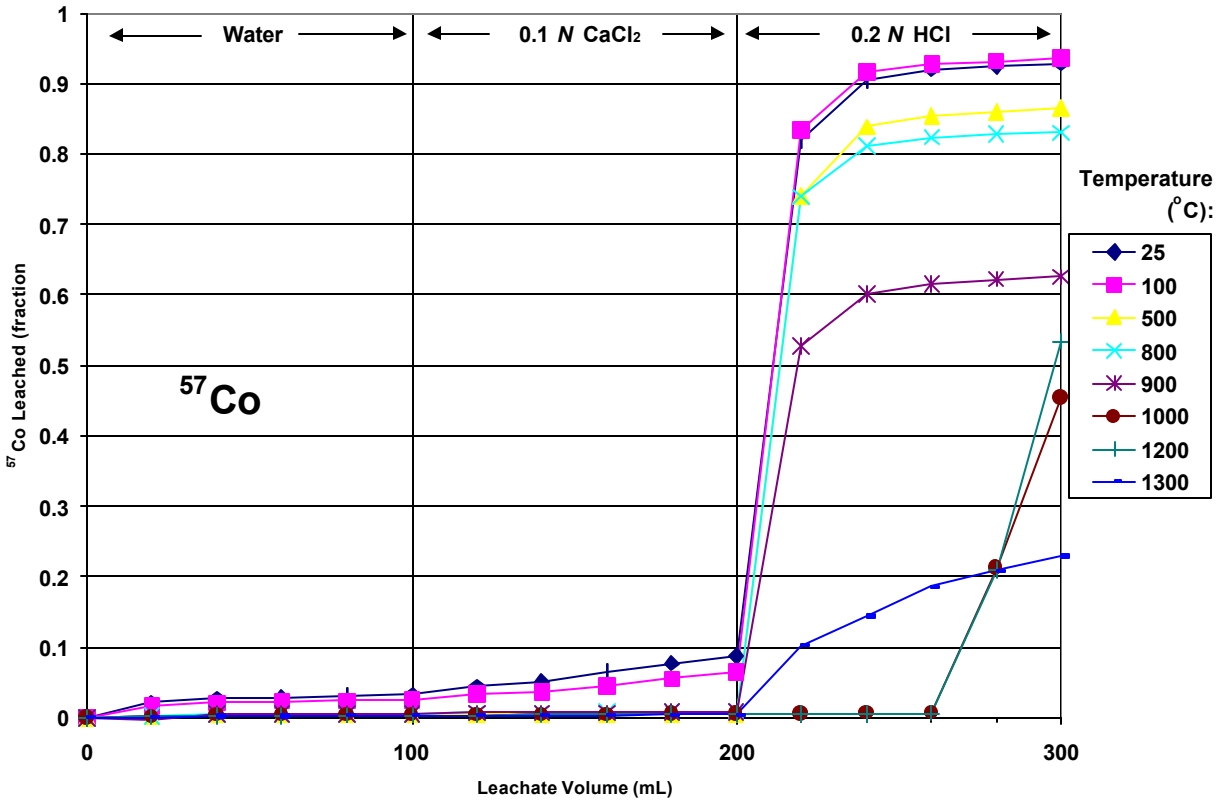

Figure S11. Sequential extraction profiles of ^{57}Co from the surface of cured Portland cement paste plugs following heating to various temperatures.

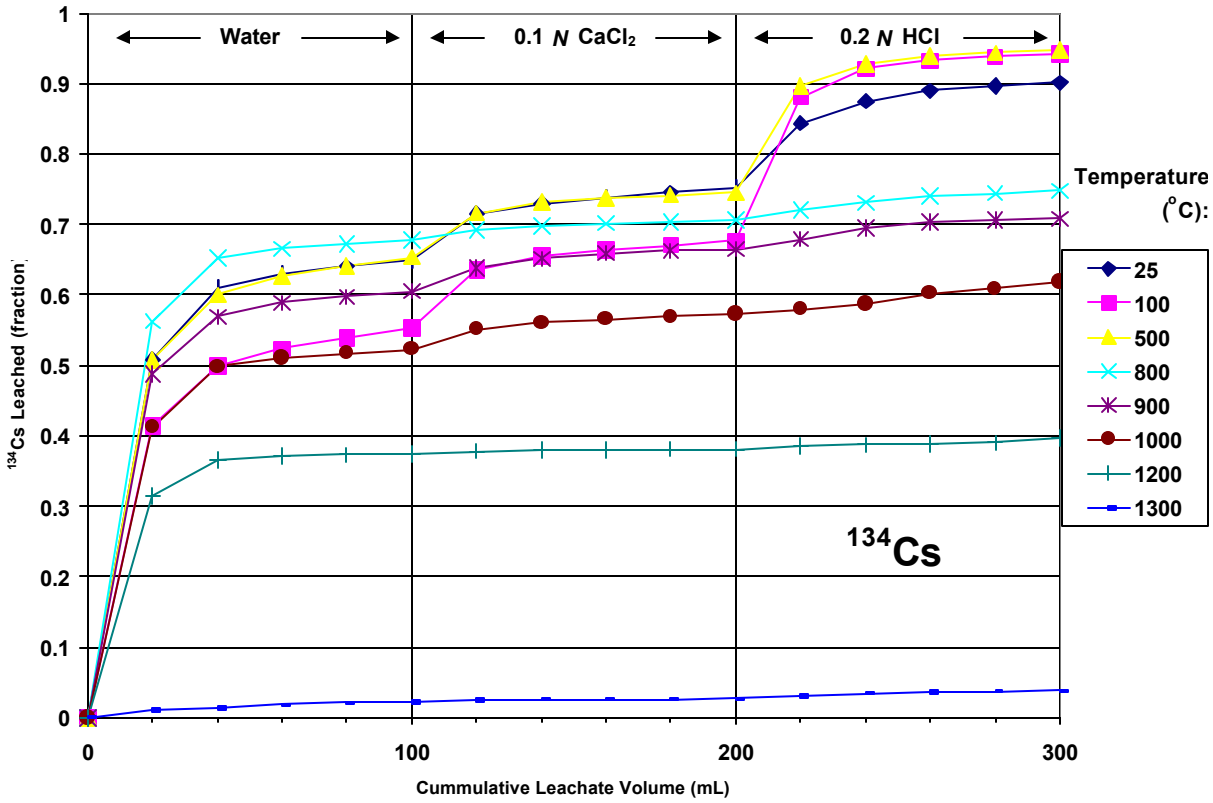

Figure S12. Sequential extraction profiles of ^{134}Cs from surface of cured Portland cement paste plugs following heating to various temperatures.

Table S 1. Average Elemental Analyses of Bulk Pulverized Hydrated Portland Cement Paste via Inductively Coupled Plasma Mass and Atomic Adsorption (for Na and K) Spectroscopy.

Elemental Oxide:	(Percent)
Al_2O_3	3.35
CaO	44.79
Fe_2O_3	2.66
K_2O	0.49
MgO	2.01
MnO_2	0.06
Na_2O	0.14
SiO_2	16.65
SrO	0.04
TiO_2	0.21
Loss-on-ignition (1400°C)	28.53
Sum of Oxides	98.95
Minor Elements:	
Ag	<0.04
As	<0.38
Ba	0.01
Be	<0.002
Cd	<0.02
Co	<0.04
Cr	<0.04
Cu	<0.04
Mo	<0.01
Ni	<0.09
P	<0.91
Pb	<0.19
Sb	<0.38
Se	<0.38
V	<0.004
Zn	<0.09
Zr	<0.38