Decontamination of Radionuclides from Concrete During and After Thermal Treatment

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DOE Concrete Decontamination Needs & Current Technology

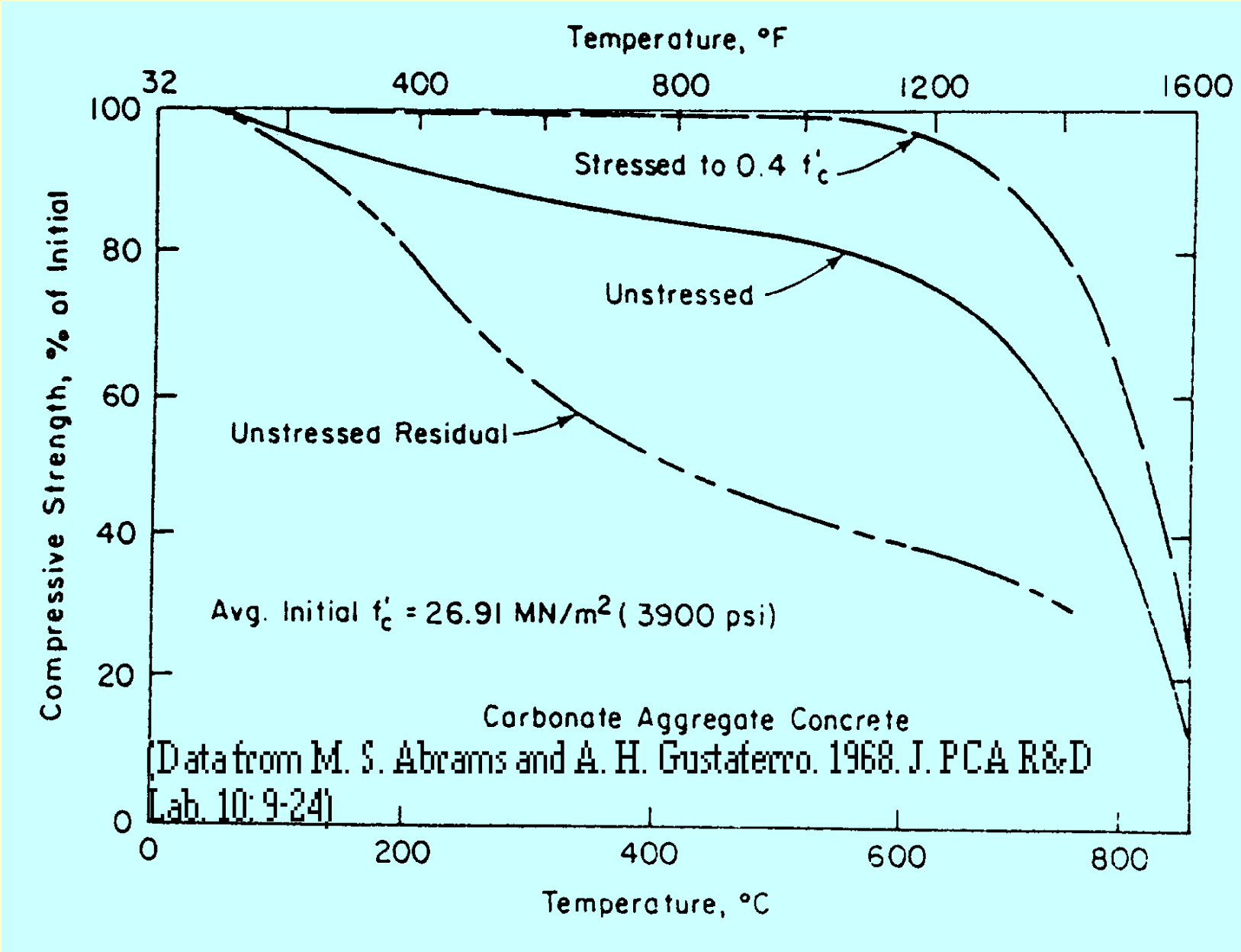
Problem Status

- 18,000 acres of contaminated concrete surfaces in DOE facilities.
- Major radionuclides are ^{137}Cs, ^{90}Sr, U, ^{60}Co, & ^{3}H.
- Thermal effects on concrete properties are generally known.
- But thermal behavior of radionuclides in concrete have not been explored.

Current Decontamination Technologies

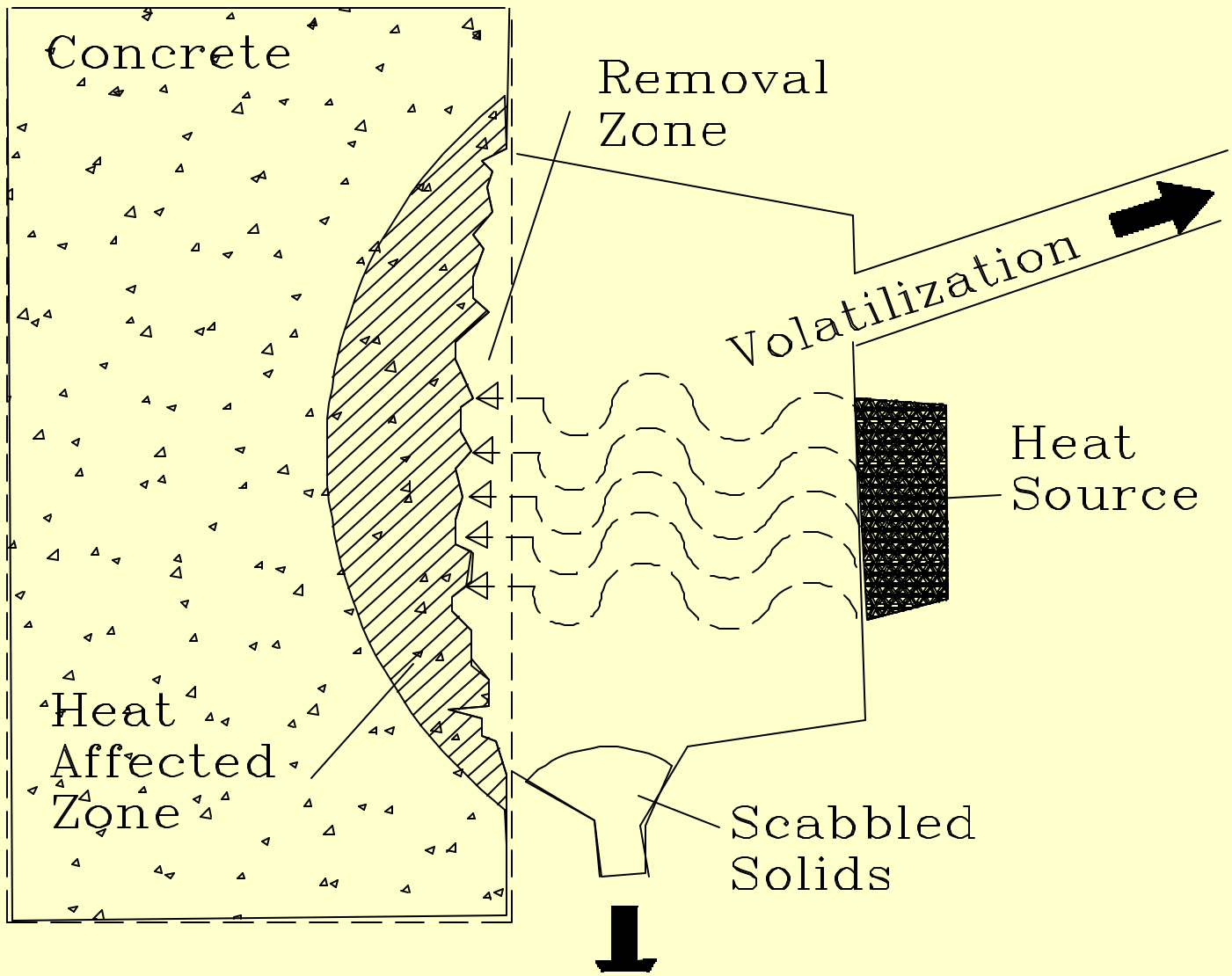
- abrasion techniques (hammer & chisel, electrohydraulic scabbling, dry-ice blasting)
- heating techniques (microwaves, plasma torch, laser ablation, high-density infrared radiation)
- chemical extraction (chelators, complexing agents, acidic dissolvers, electrokinetics)

Heating Decreases Cement Strength



Research Objectives/Questions for Thermal Treatment of Concrete

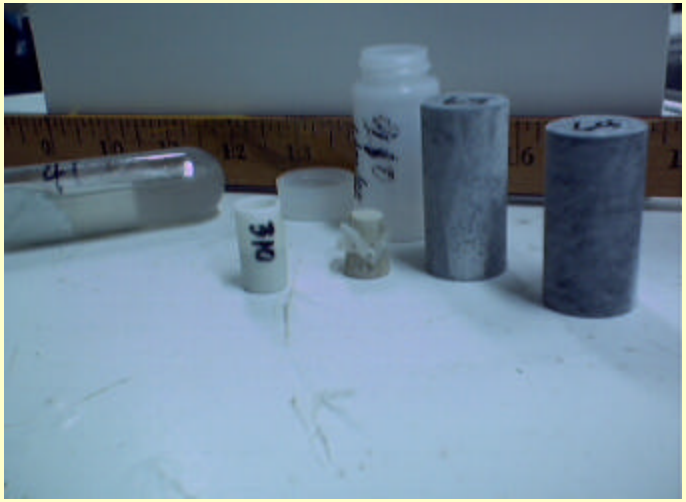
- To what degree do the major DOE radionuclide contaminants volatilize when cement is heating through its range of strength deterioration up to its vitrification?
- How does the extractability or leachability of the major DOE radionuclide contaminants change after cement is heated through its range of strength deterioration up to its vitrification?
- Can technologies for the decontamination of concrete surfaces be augmented by concurrent or prior thermal treatments?



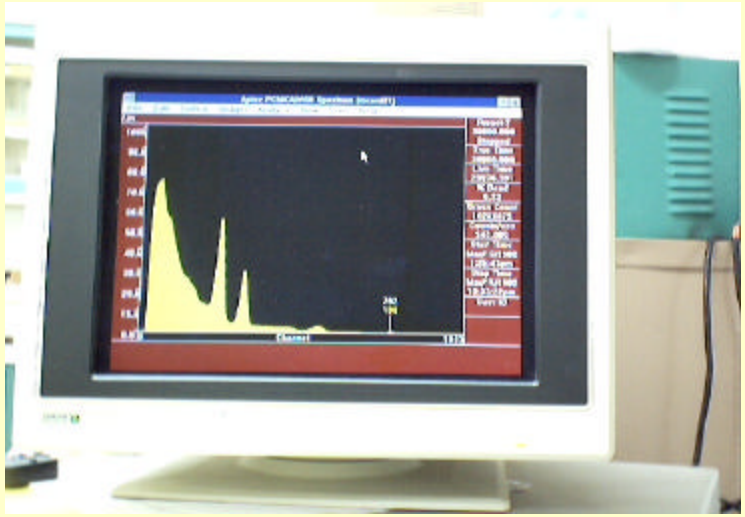
Conceptual Processes During Heating of Concrete

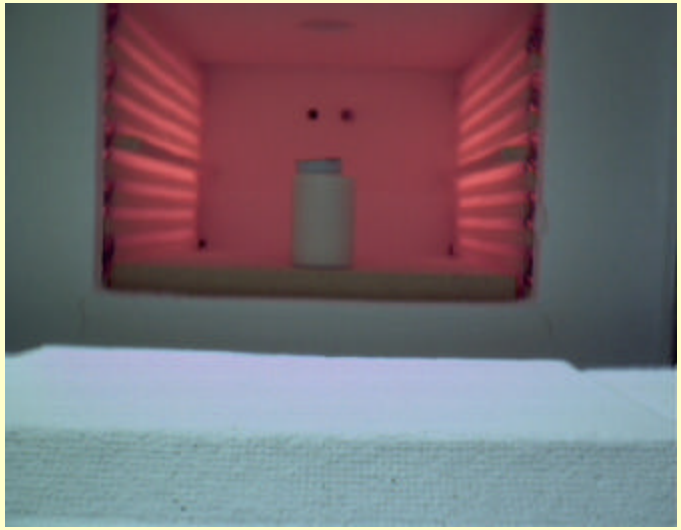
Heating and Decontaminating Cement - The Tools

Crucibles, Extraction Tubes, & Cement Plugs



Gamma Spectroscopy





High-Temperature Furnace

Compression Testing

Methods for Investigating Radionuclide Contaminant Behavior:

1. Portland Cement + Water Þ Hydrated Portland Cement (Cured)
2. Hydrated Portland Cement Þ Fine (100-200 mesh) Cement Powder
3. Cement Powder + Radioactive Tracer (^{134}Cs, ^{85}Sr, ^{57}Co, or U) Þ Contaminated Cement Powder
4. Contaminated Cement Powder + Time Þ Aged Contaminated Cement

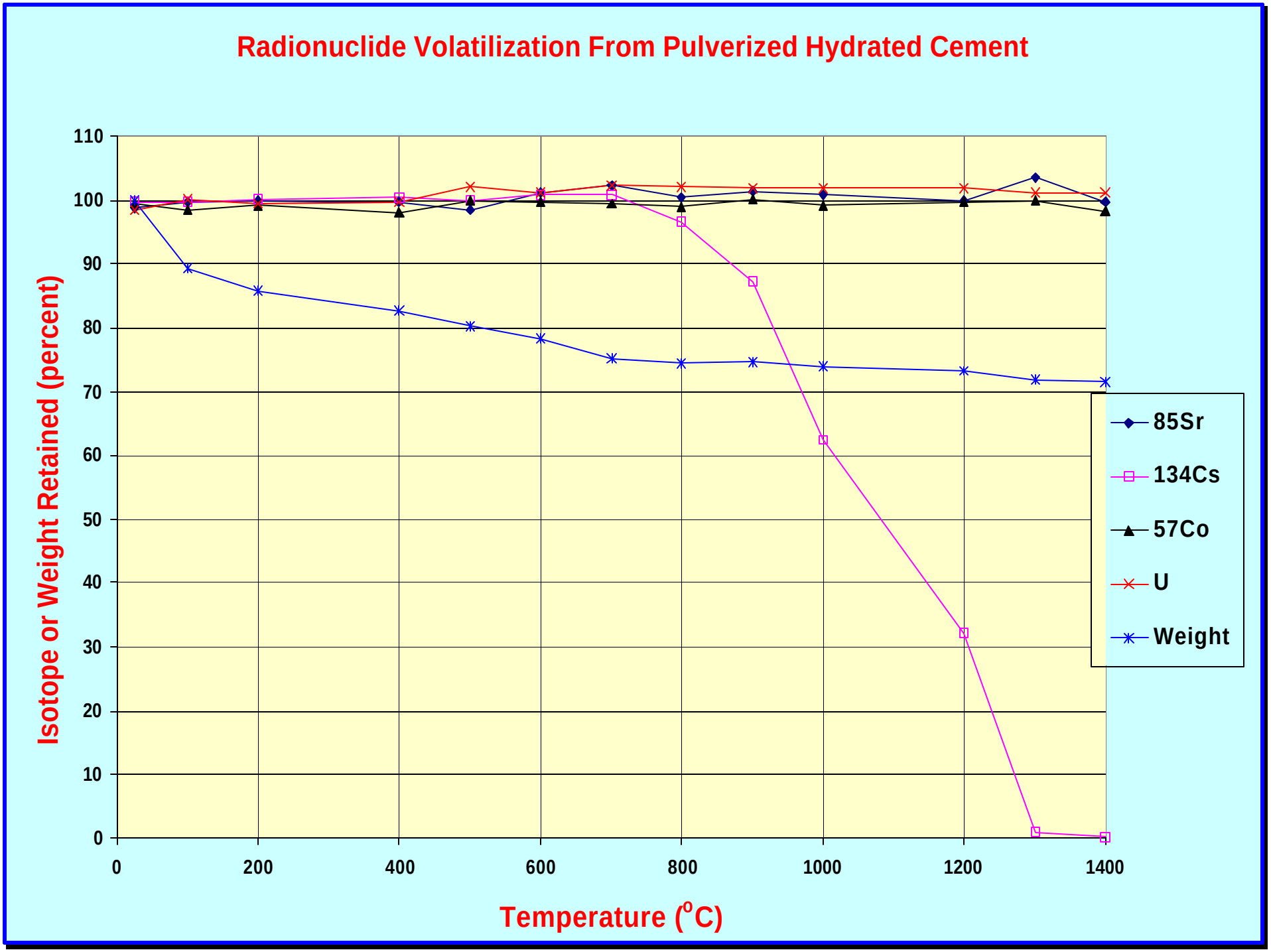
5.a. Aged Contaminated Cement + Heat (100-1400°C) Þ Volatilized Radionuclides

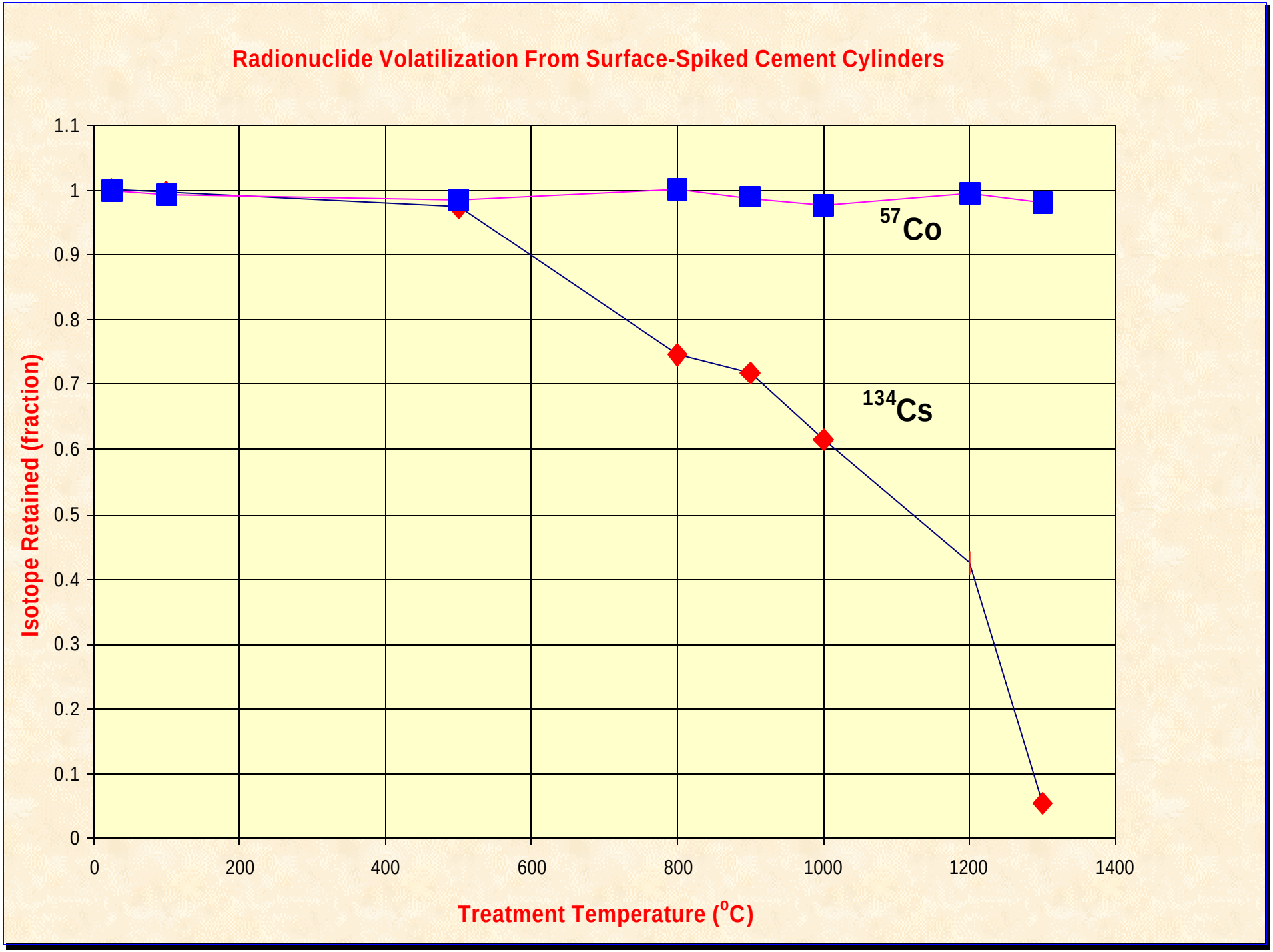
5.b. Aged Contaminated Cement + Heat (100-1400°C) Þ Thermally-Altered Cement

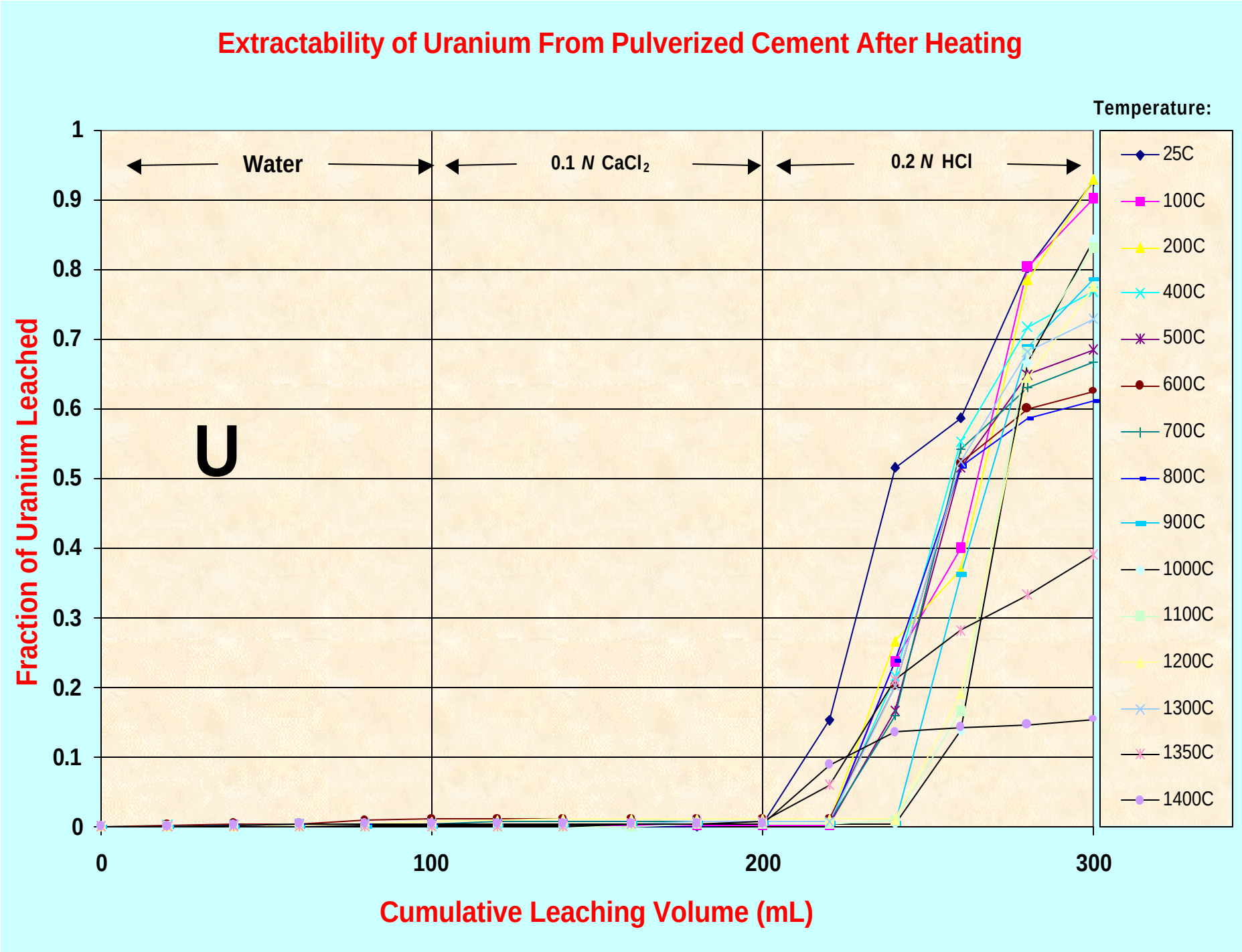
6. Thermally-Altered Cement + Extractants Þ Soluble Contaminants + Residual Cement Solids

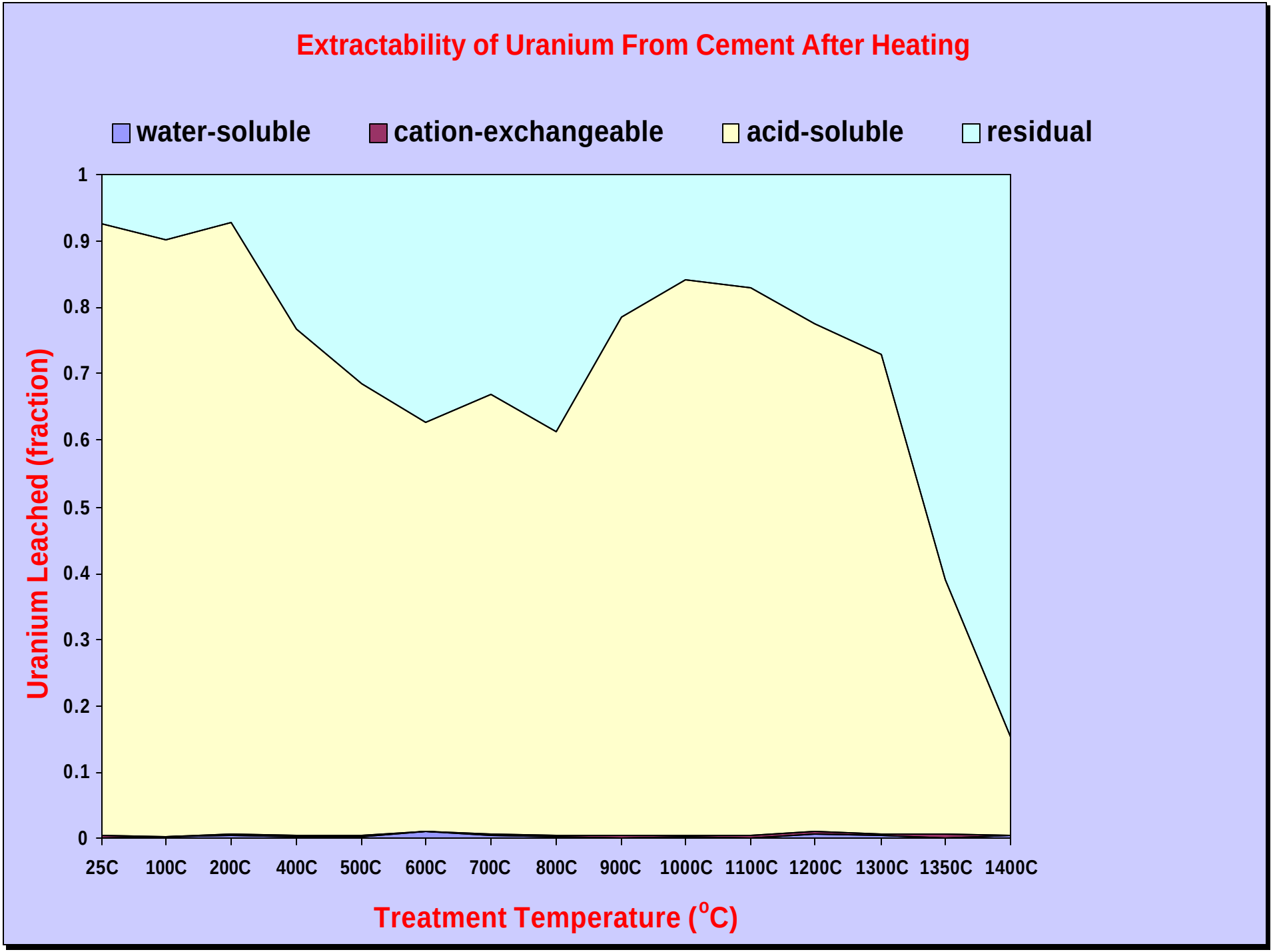
Sequential Extractant Scheme:

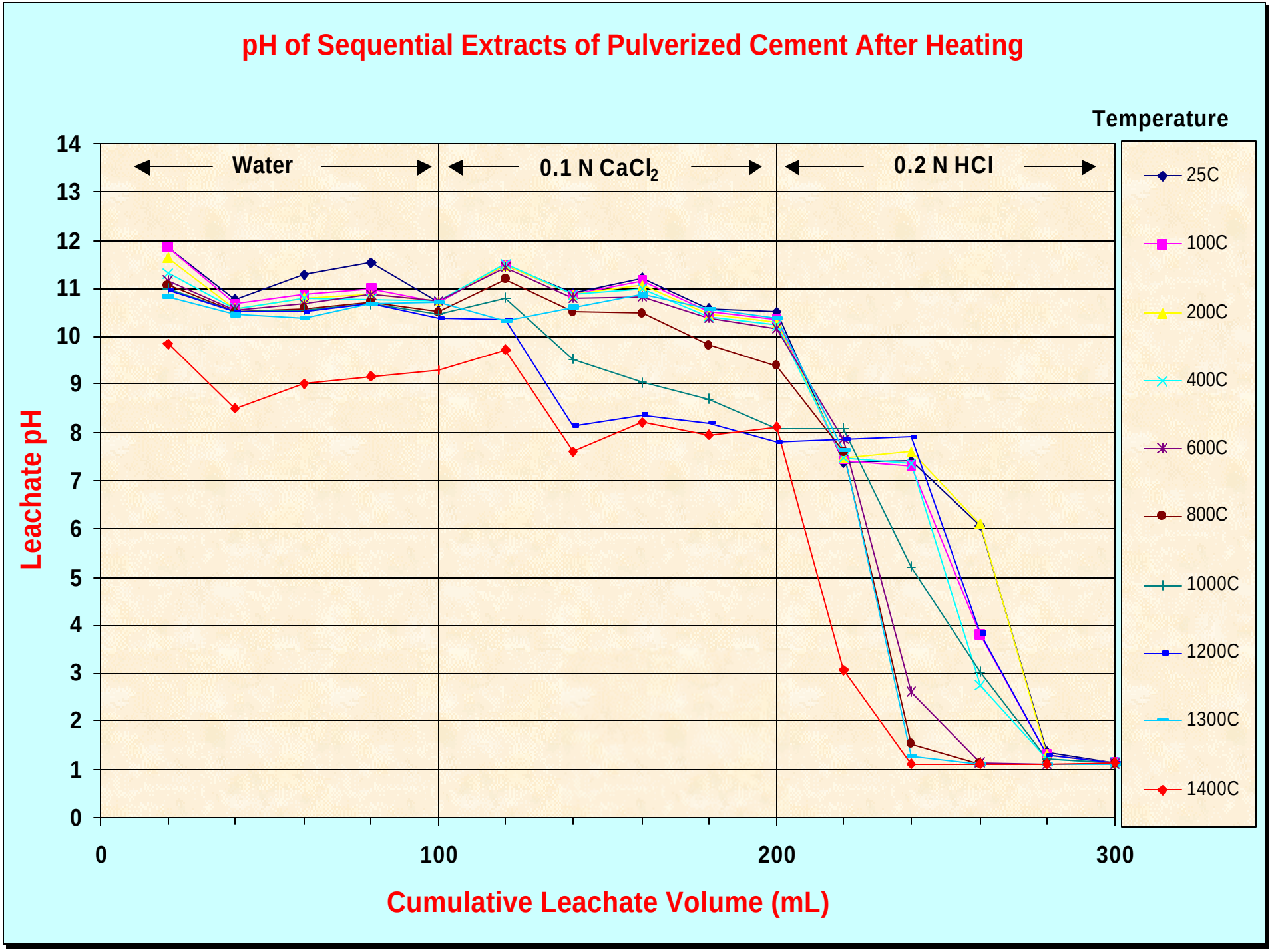
A. Water - Soluble Fraction
B. Dilute $CaCl_2$ - Cation Exchangeable Fraction
C. Dilute HCl - Surface Dissolvable Fraction

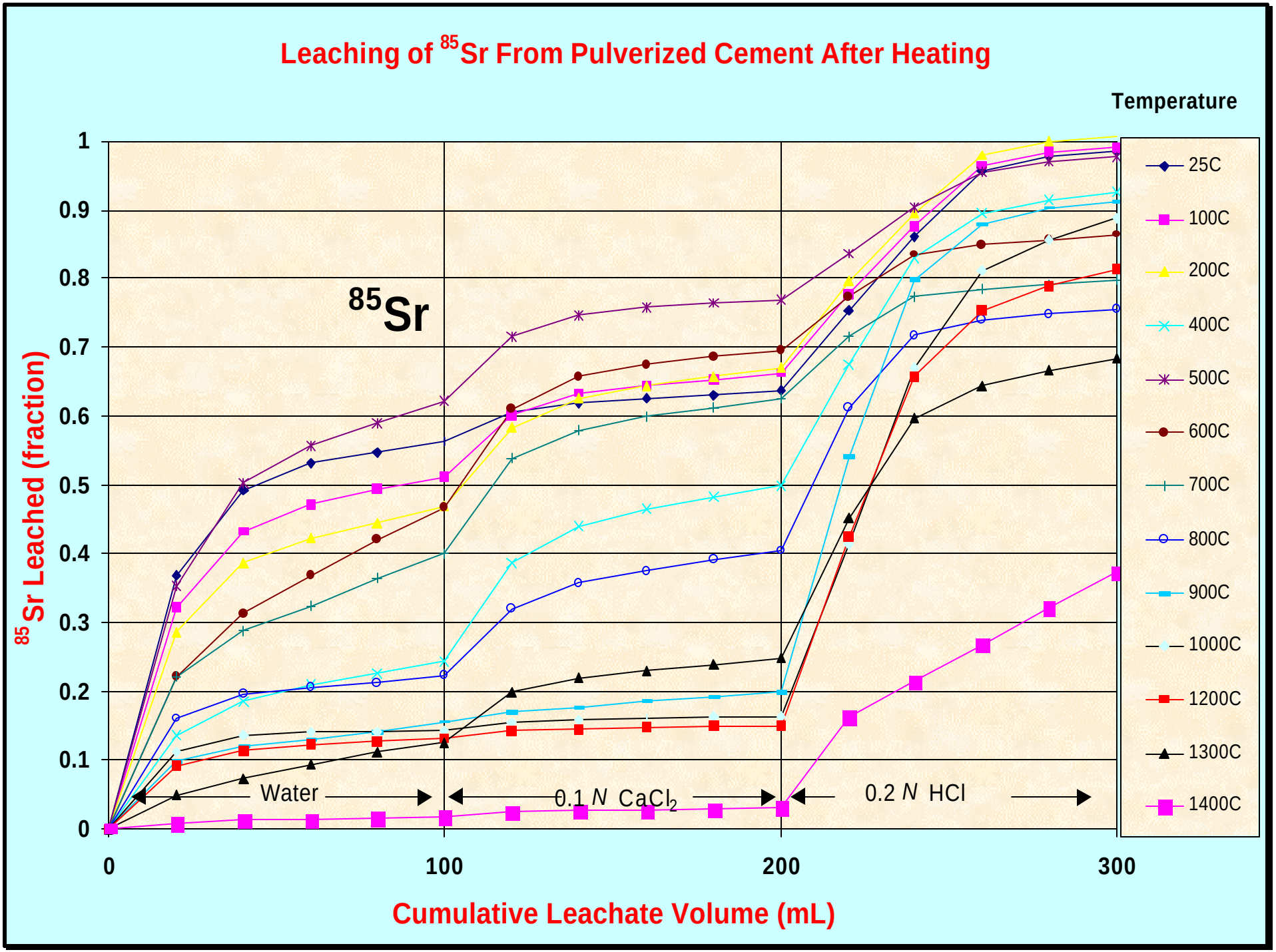

Radionuclide Volatilization From Pulverized Hydrated Cement
Isotope or Weight Retained (percent)
110
100
90
80
70
60
50
40
30
20
10
0
0
200
400
600
800
1000
1200
1400
Temperature (°C)
85Sr
134Cs
57Co
U
Weight

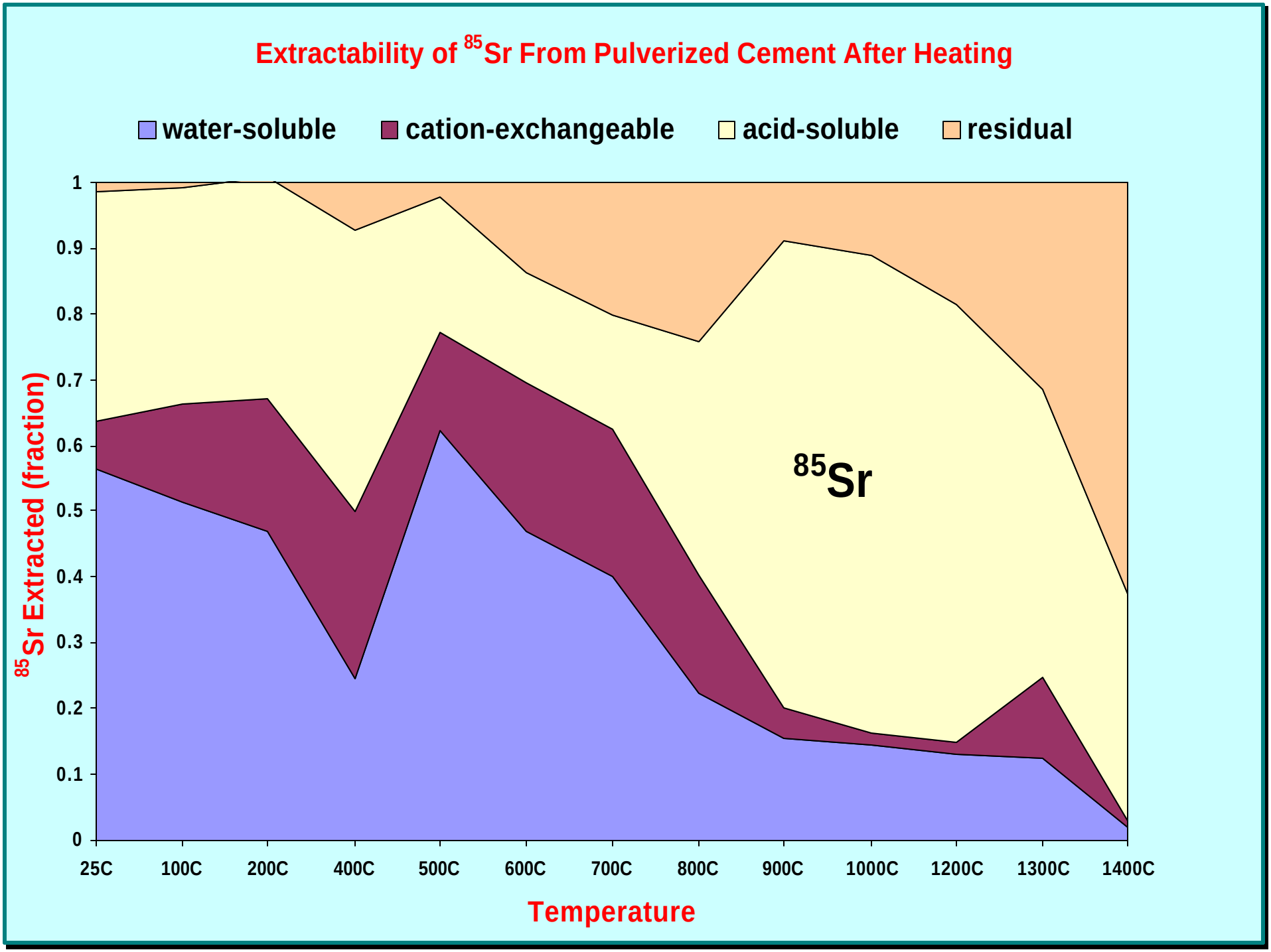

Radionuclide Volatilization From Surface-Spiked Cement Cylinders
Isotope Retained (fraction)
1.1
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
0
200
400
600
800
1000
1200
1400
Treatment Temperature (°C)
57Co
134Cs

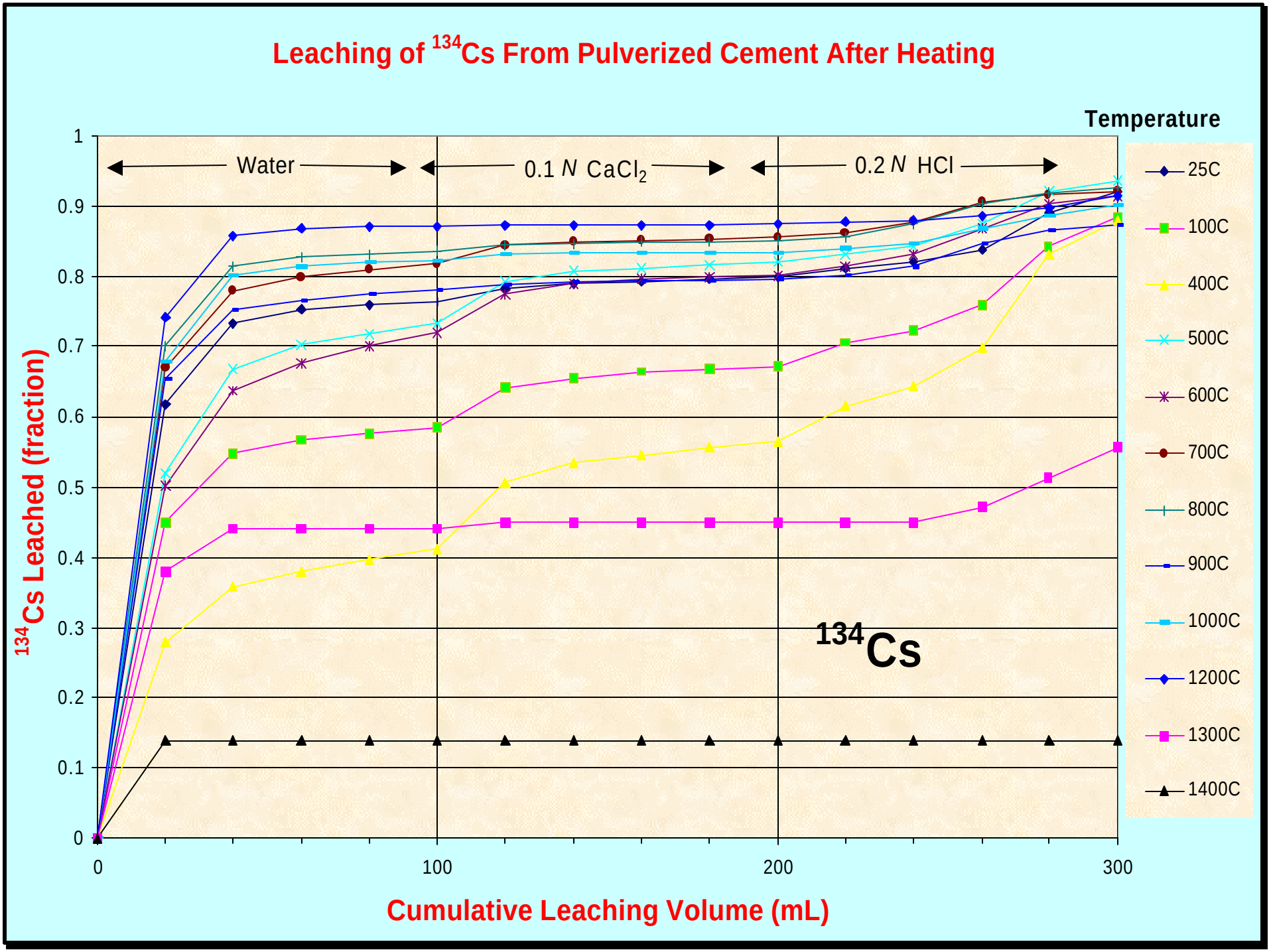

Extractability of Uranium From Pulverized Cement After Heating
Temperature:
Water
0.1 N CaCl2
0.2 N HCl
U
25C
100C
200C
400C
500C
600C
700C
800C
900C
1000C
1100C
1200C
1300C
1350C
1400C
Fraction of Uranium Leached
Cumulative Leaching Volume (mL)
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300

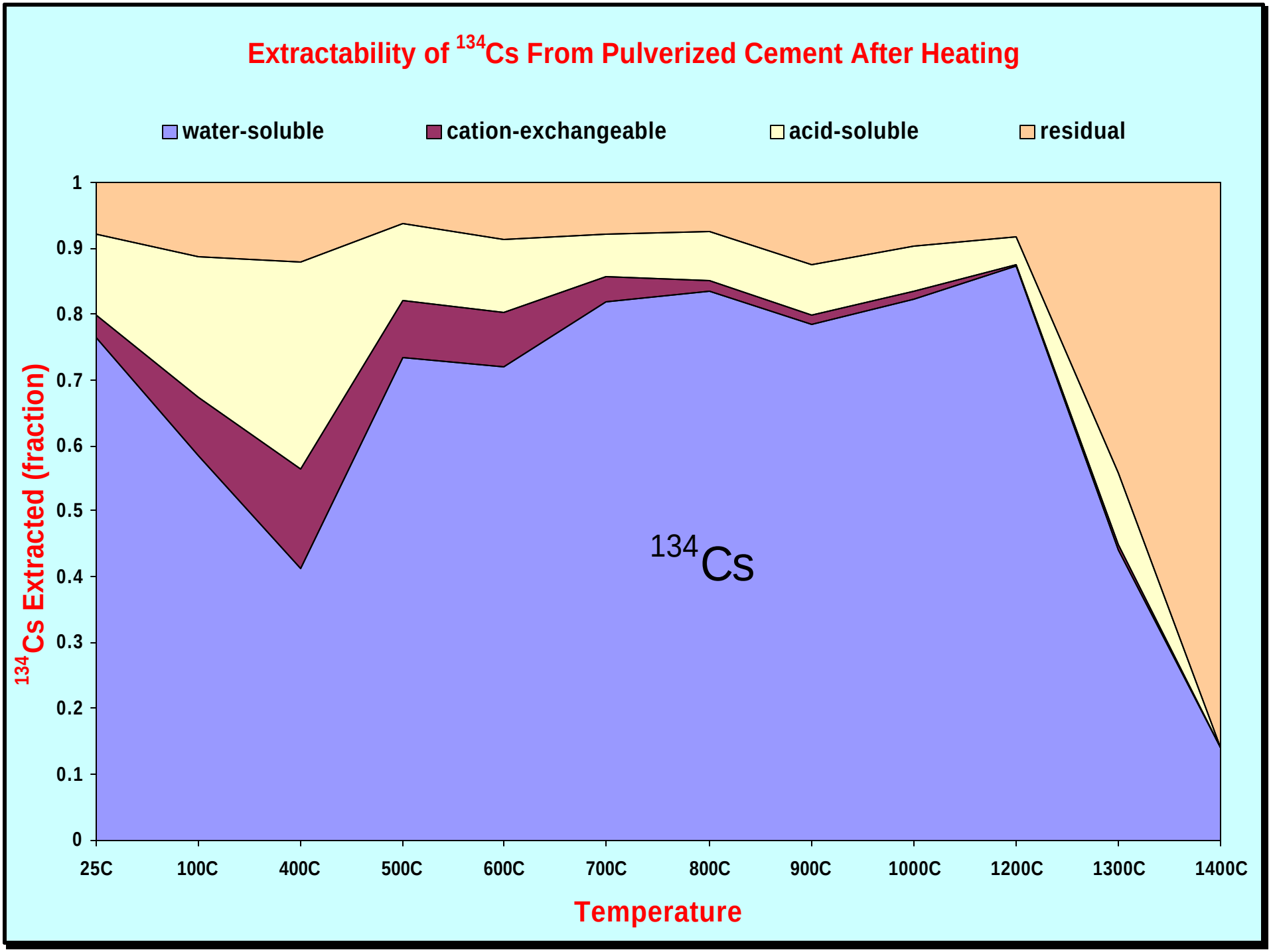

Extractability of Uranium From Cement After Heating
water-soluble
cation-exchangeable
acid-soluble
residual
Uranium Leached (fraction)
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
25C
100C
200C
400C
500C
600C
700C
800C
900C
1000C
1100C
1200C
1300C
1350C
1400C
Treatment Temperature (°C)

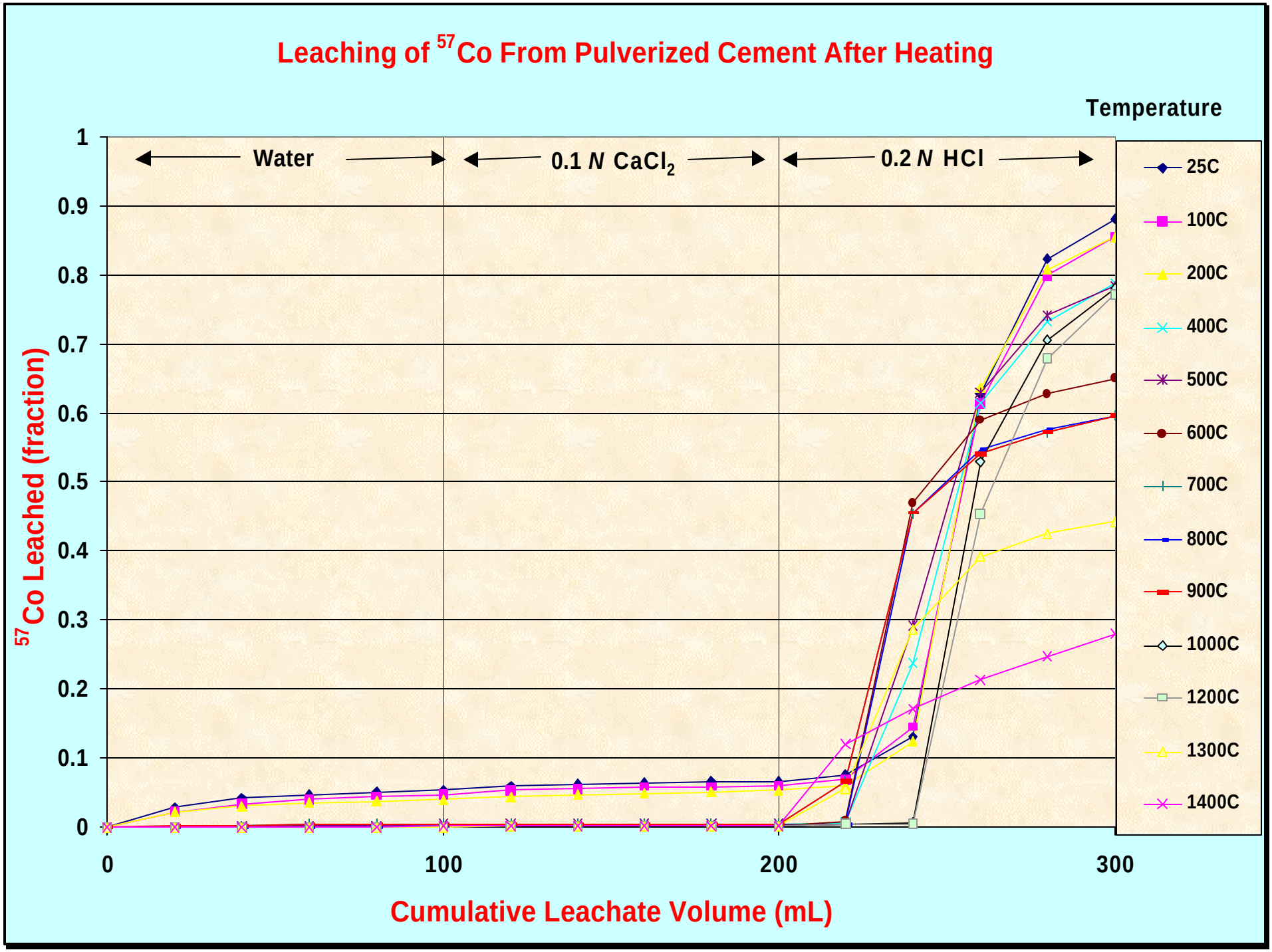

pH of Sequential Extracts of Pulverized Cement After Heating
Temperature
Water
0.1 N CaCl2
0.2 N HCl
25C
100C
200C
400C
600C
800C
1000C
1200C
1300C
1400C
Leachate pH
Cumulative Leachate Volume (mL)
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
0
100
200
300

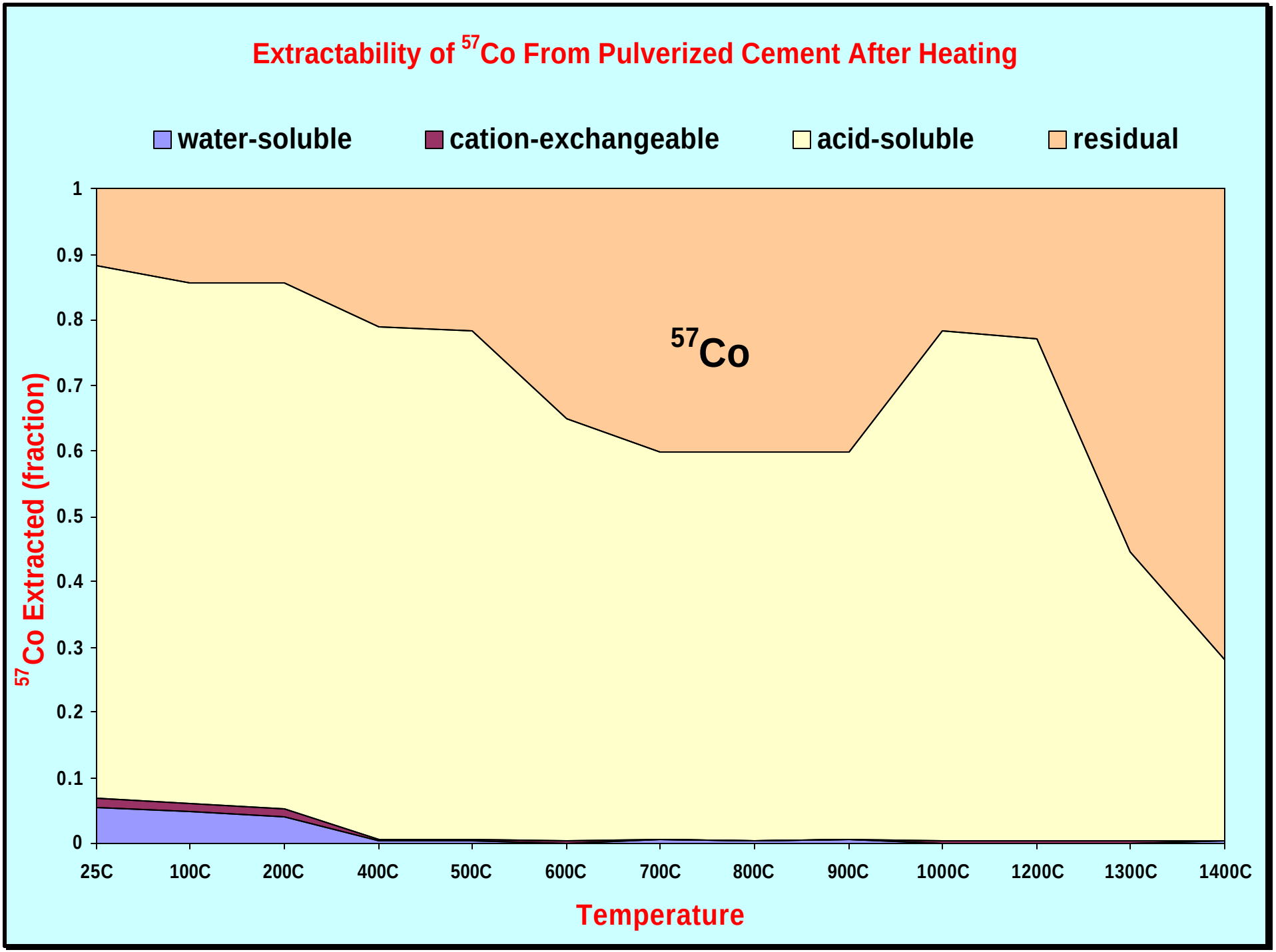

Leaching of 85Sr From Pulverized Cement After Heating
Temperature
85Sr
85Sr Leached (fraction)
Cumulative Leachate Volume (mL)
Water
0.1 N CaCl2
0.2 N HCl
25C
100C
200C
400C
500C
600C
700C
800C
900C
1000C
1200C
1300C
1400C
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300


Extractability of 85Sr From Pulverized Cement After Heating
water-soluble
cation-exchangeable
acid-soluble
residual
85Sr
85Sr Extracted (fraction)
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
25C
100C
200C
400C
500C
600C
700C
800C
900C
1000C
1200C
1300C
1400C
Temperature


Leaching of 134Cs From Pulverized Cement After Heating
Temperature
Water
0.1 N CaCl2
0.2 N HCl
134Cs
25C
100C
400C
500C
600C
700C
800C
900C
1000C
1200C
1300C
1400C
134Cs Leached (fraction)
Cumulative Leaching Volume (mL)
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300


Extractability of 134Cs From Pulverized Cement After Heating
water-soluble
cation-exchangeable
acid-soluble
residual
134Cs Extracted (fraction)
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
134Cs
25C
100C
400C
500C
600C
700C
800C
900C
1000C
1200C
1300C
1400C
Temperature


Leaching of 57Co From Pulverized Cement After Heating
Temperature
Water
0.1 N CaCl2
0.2 N HCl
25C
100C
200C
400C
500C
600C
700C
800C
900C
1000C
1200C
1300C
1400C
57Co Leached (fraction)
Cumulative Leachate Volume (mL)
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
0
100
200
300


Extractability of 57Co From Pulverized Cement After Heating
water-soluble
cation-exchangeable
acid-soluble
residual
57Co
57Co Extracted (fraction)
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
25C
100C
200C
400C
500C
600C
700C
800C
900C
1000C
1200C
1300C
1400C
Temperature

Current Conclusions and Planned Investigations

Interim Conclusions

- ^{137}Cs will volatilize if concrete is heated to temperature necessary for disintegration (1000°C).
- Uranium, ^{57}Co, and ^{85}Sr do not volatilize from concrete when heated even to its melting temperature (1350°C).
- Extractability of radionuclides mostly unaffected until onset of vitrification where it decreases sharply.
- Strength of cement is destroyed by thermal treatments above 1000°C.

Planned Tasks

- Radionuclide extractability by chelators/complexors relative to thermal treatment.
- Cement compressive strength versus thermal treatment measurements.

Decontamination of Concrete By Thermal Treatment
The Anticipated R&D Product

A comprehensive database on both the physical/engineering properties and the behavior of four major radionuclide contaminants in concrete as a function of temperature, heating rate, and the nature of its aggregates.

The end-user will be able to specify thermal conditions for the treatment and decontamination of particular concretes and predict the fate and behavior of its radionuclide contaminants.