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Polymer Filtration Process for Removal of Plutonium-238 from Aqueous Scrap Recovery Waste Streams

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Plutonium-238 is an excellent radioisotope for heat source applications in space because of its power density, useful lifetime, minimal shielding requirements and oxide stability. The U.S. is not presently producing new quantities of Pu-238. A glovebox facility is under construction at Los Alamos that will recover a significant quantity of the impure Pu-238 that exists in scrap and residues from past production operations. The general flowsheet consists of milling, acid dissolution, ion exchange, precipitation, calcination, oxygen isotope exchange, and waste treatment operations. Polymer Filtration will be used to remove Pu-238 from aqueous waste streams. Polymer Filtration uses water-soluble polymers to selectively bind Pu metal ions and ultrafiltration to concentrate the polymer-metal complex producing a permeate with low levels of Pu-238. This technology is being implemented on a process scale for the first time in the Pu-238 scrap recovery operations.

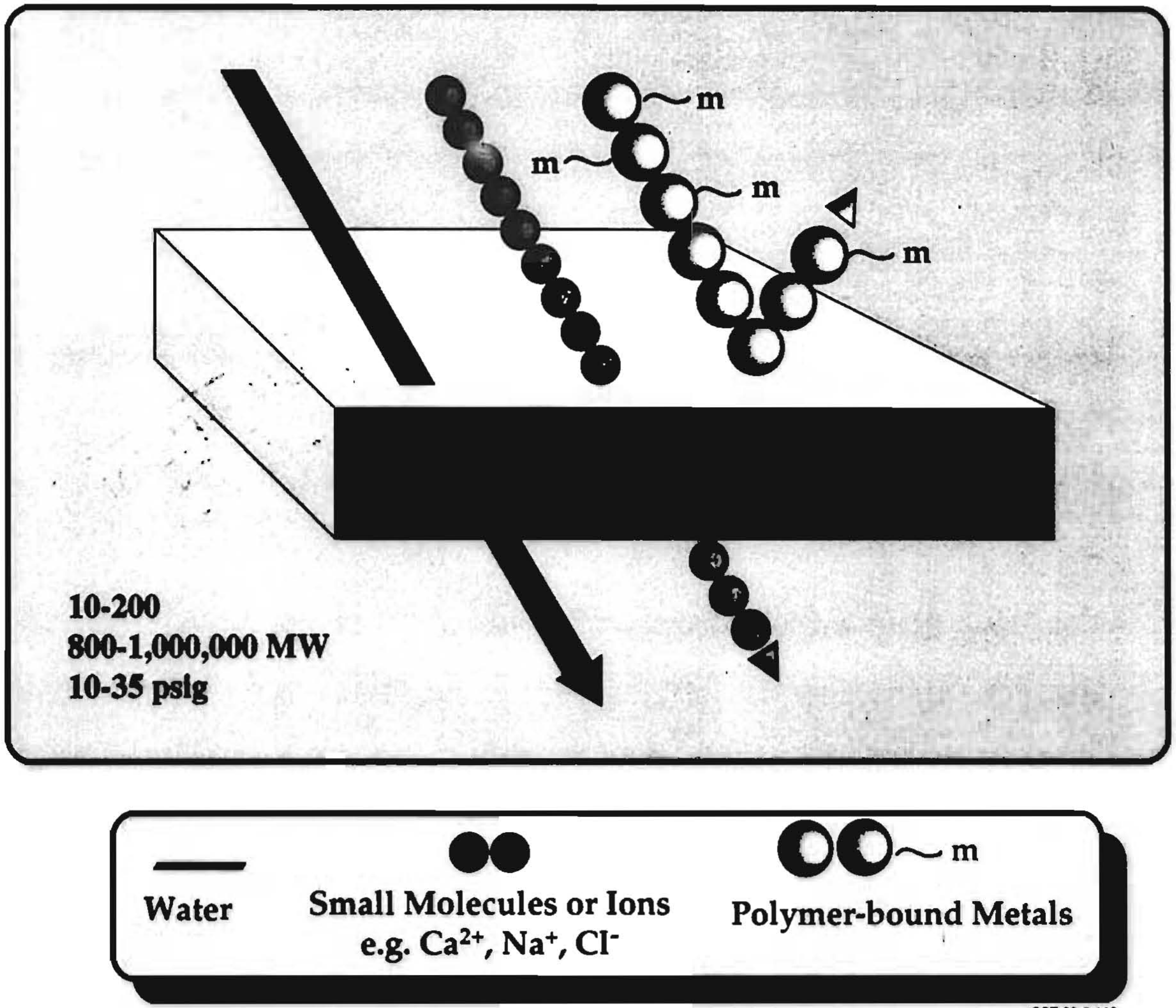
Poster Presentation: unclassfied, not UCNI

Introduction

Plutonium-238 is an excellent radioisotope for heat source applications in space because of power density, useful lifetime, minimal shielding requirements and oxide stability. Although the U.S. is not producing new quantities of Pu-238, Los Alamos will be recovering a significant quantity of the impure Pu-238 that exists in scrap and residues from past production operations.

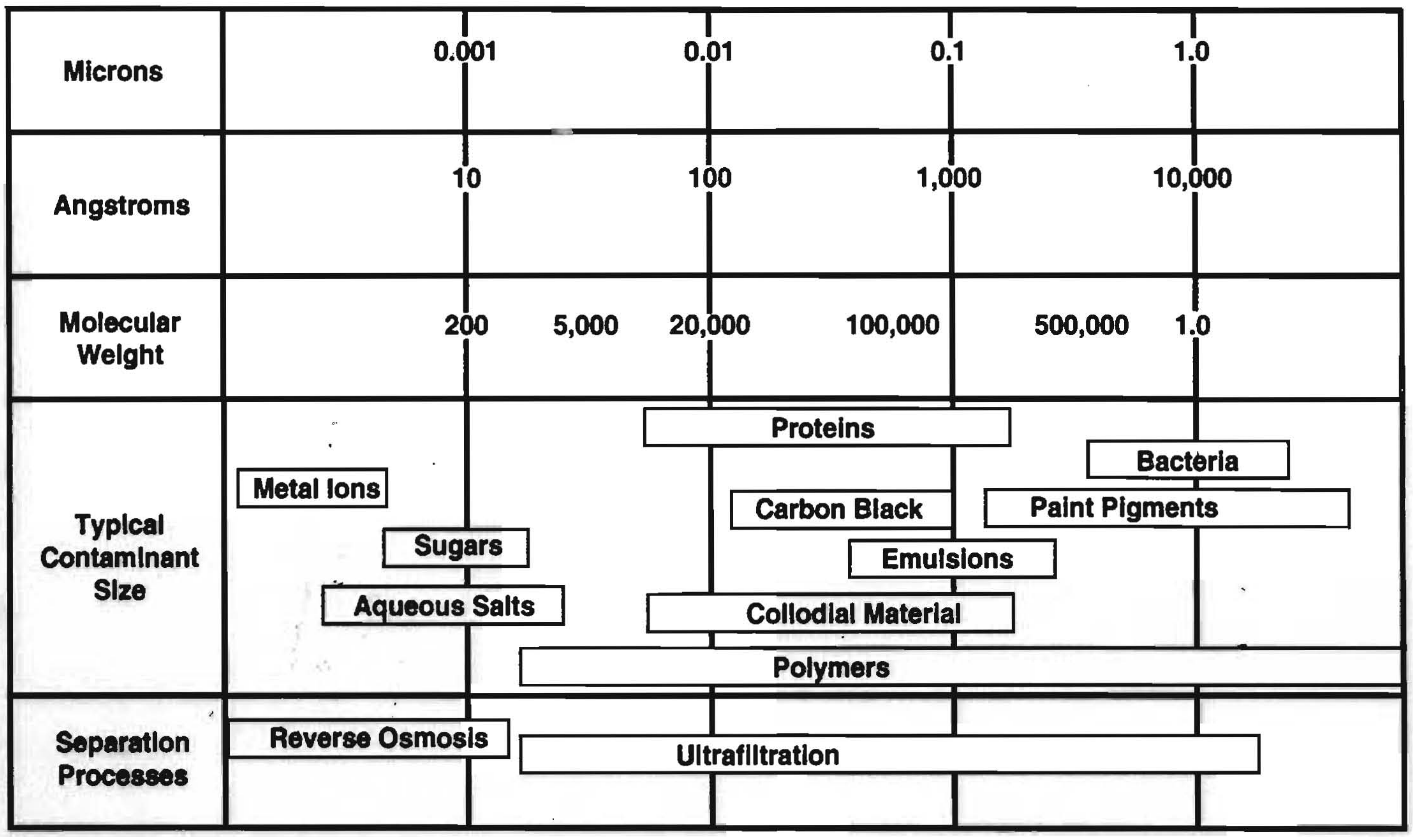
Polymer Filtration (PF) is being implemented on a process scale for the first time in the new Pu-238 aqueous scrap recovery operations. Use of PF in treating the effluents from these operations will ensure that regulatory limits are met, disposal costs are decreased, and waste is minimized.

Polymer filtration is based on the use of specially designed water-soluble polymers that selectively bind with metal ions. The polymer has a large molecular weight that allows it to be physically separated from the waste stream using ultrafiltration along with the metal ions bound to the polymer.

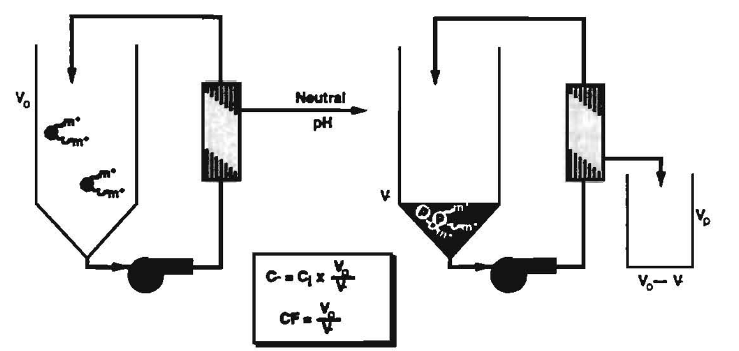

m
m
m
m
10-200
800-1,000,000 MW
10-35 psig
Water
Small Molecules or Ions
e.g. Ca^{2+}, Na^{+}, Cl^{-}
m
Polymer-bound Metals

CST 92-2,119

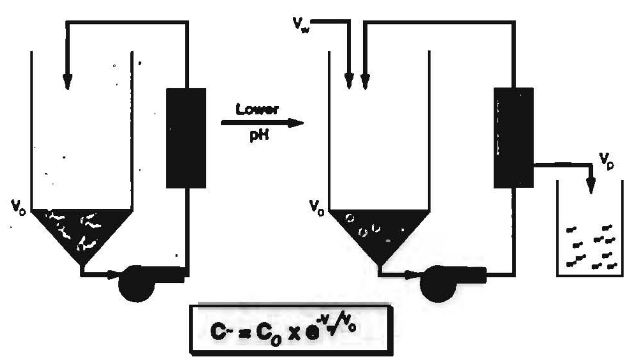
Filtration Spectrum



Polymer Filtration™ Process



Schematic of concentration process



Schematic of the regeneration
$V_w = V_p$

V_o = Starting volume
V_f = Final volume
V_p = Volume permeate
V_w = Volume water

Advantages of Polymer Filtration

- Reactions occur in a single phase
- All polymer structure can be used (higher capacity)
- Mixture of polymers can target a group of metal ions
- Ultrafiltration will remove colloidal material
- Scale-up of process straightforward
- Relatively low pressure operation – 10-50 psig
- No organic solvents used or "mixed waste" produced

Potential Drawbacks

- Fouling of ultrafiltration unit
- Single equilibrium stage for each ultrafiltration step

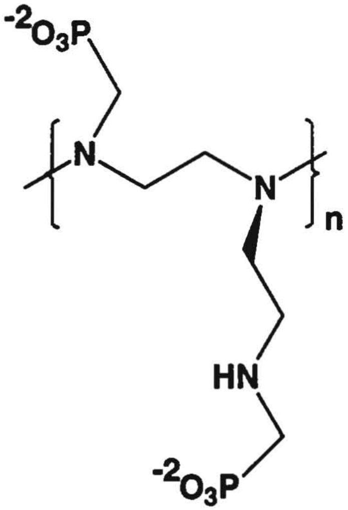
Preliminary Research

The use of polymer filtration to remove actinides and regulated metals has been studied over the past decade at Los Alamos. Polymer selectivity, loading capacity, and the influence of polymer binding parameters such as pH have also been investigated. The research for this project focused on confirming that polymer filtration of the Pu-238 scrap recovery waste stream was possible based on previous experimental data obtained in working with other actinides and metals.

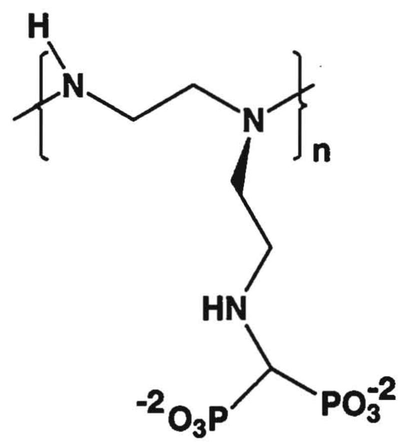
Considering the high radiation environment, it was important to study the effect of polymer degradation in the presence of Pu-238. The polymers used for this study have phosphonic acid functional groups attached to a polyamine backbone (see PEIP and PEIDP below). Equal amounts of process effluent with an activity of 1.0×10^8 CPMa / 0.01ml (1.22 Ci/L) were added to each of 2 centricon units (10K NMWC). 0.1wt % of PEIP was add to one and 0.1wt% of PEIDP was added to the other. A sample of the permeates were taken upon contact and periodically thereafter over 3 days.

Another goal was to demonstrate the efficiency of the ultrafilter without the use of polymers. This study involved the use of Pu-238 hydroxide filtrates with activity of ~ 5.0 X 10^4 CPMa /0.1ml (4 x 10^{-2} Ci/L) adjusted to various pH levels and ultrafiltered.

Structural Diagrams of Phosphonic Acid Polymers

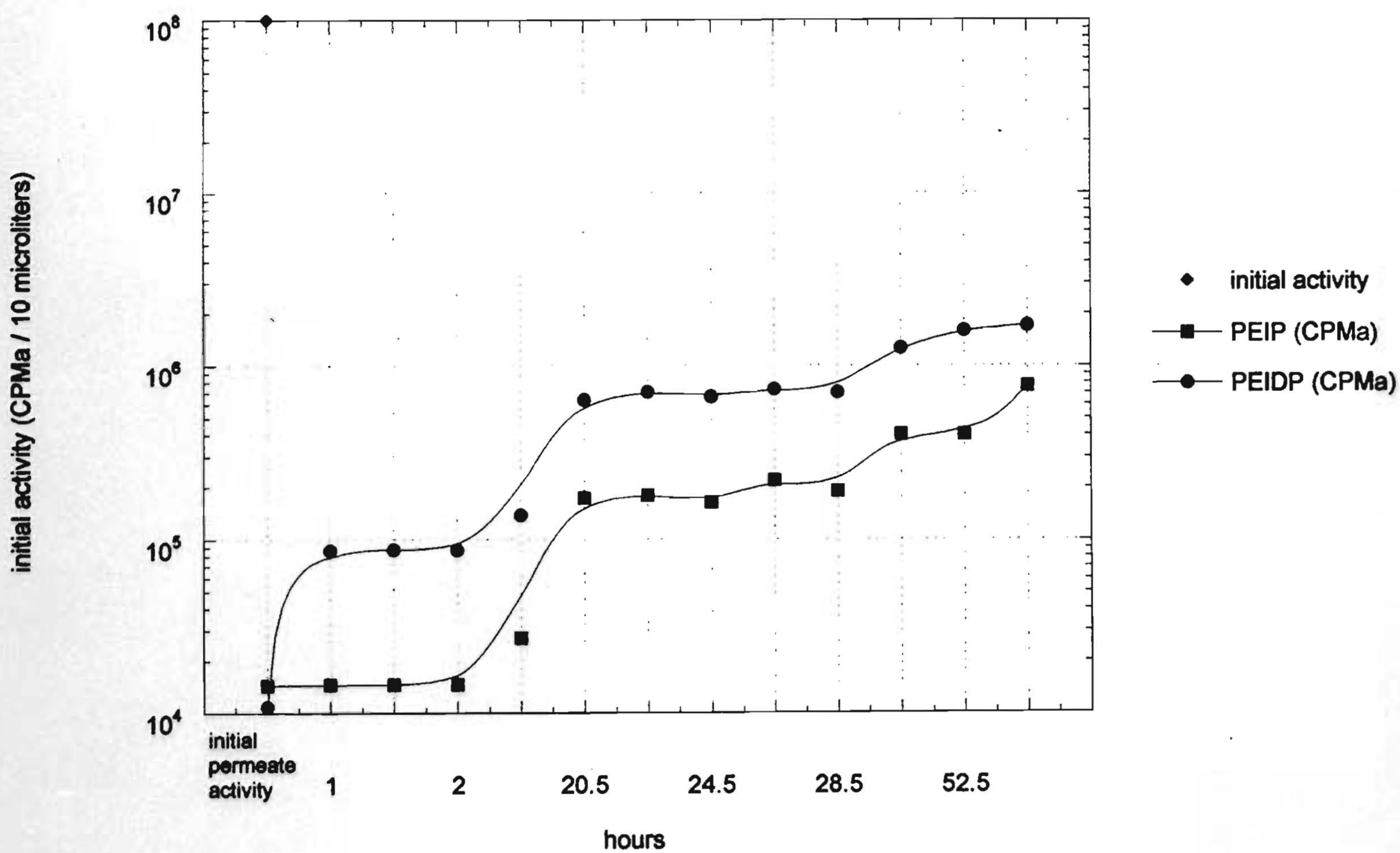


PEIP - high pH

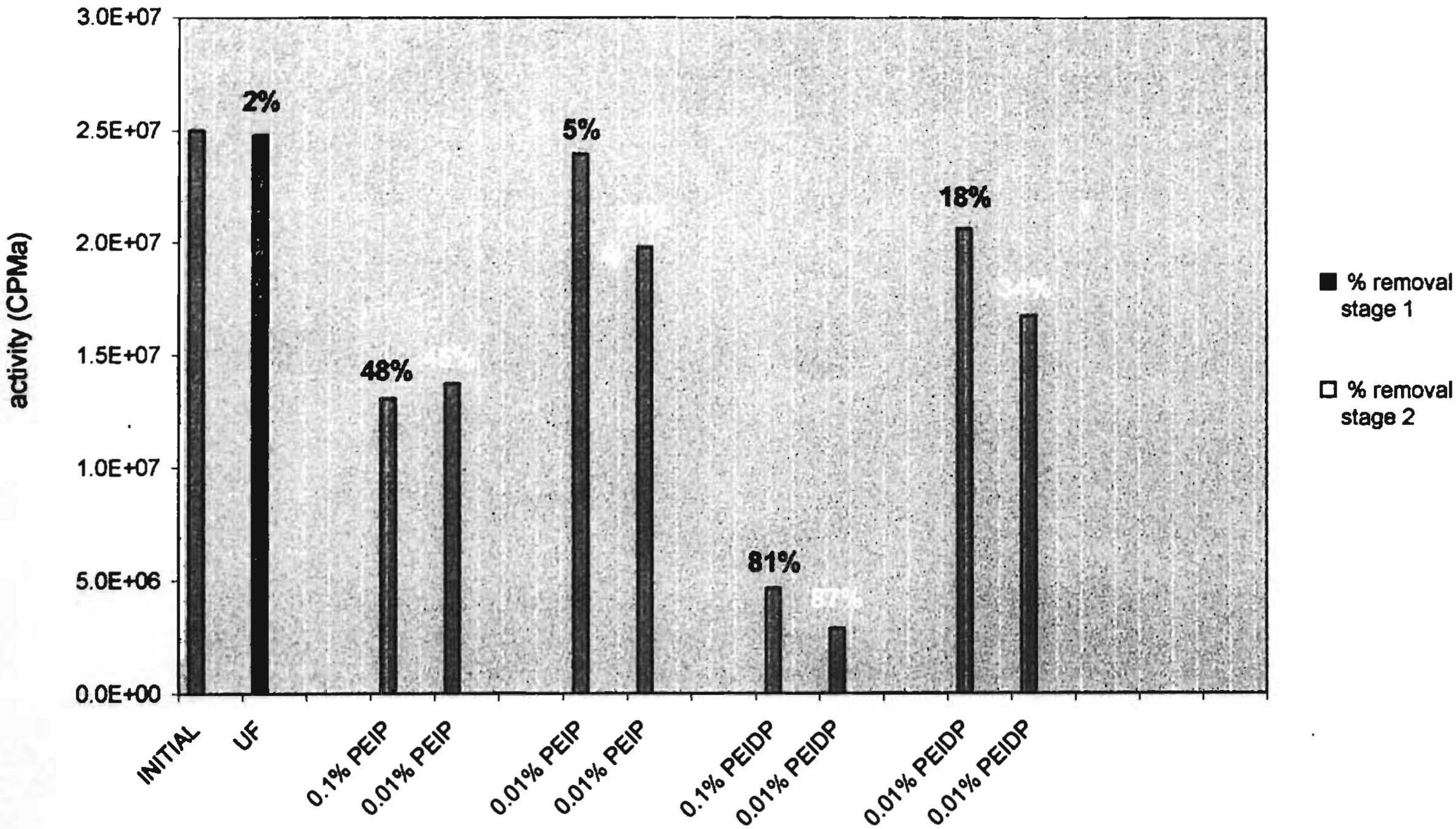


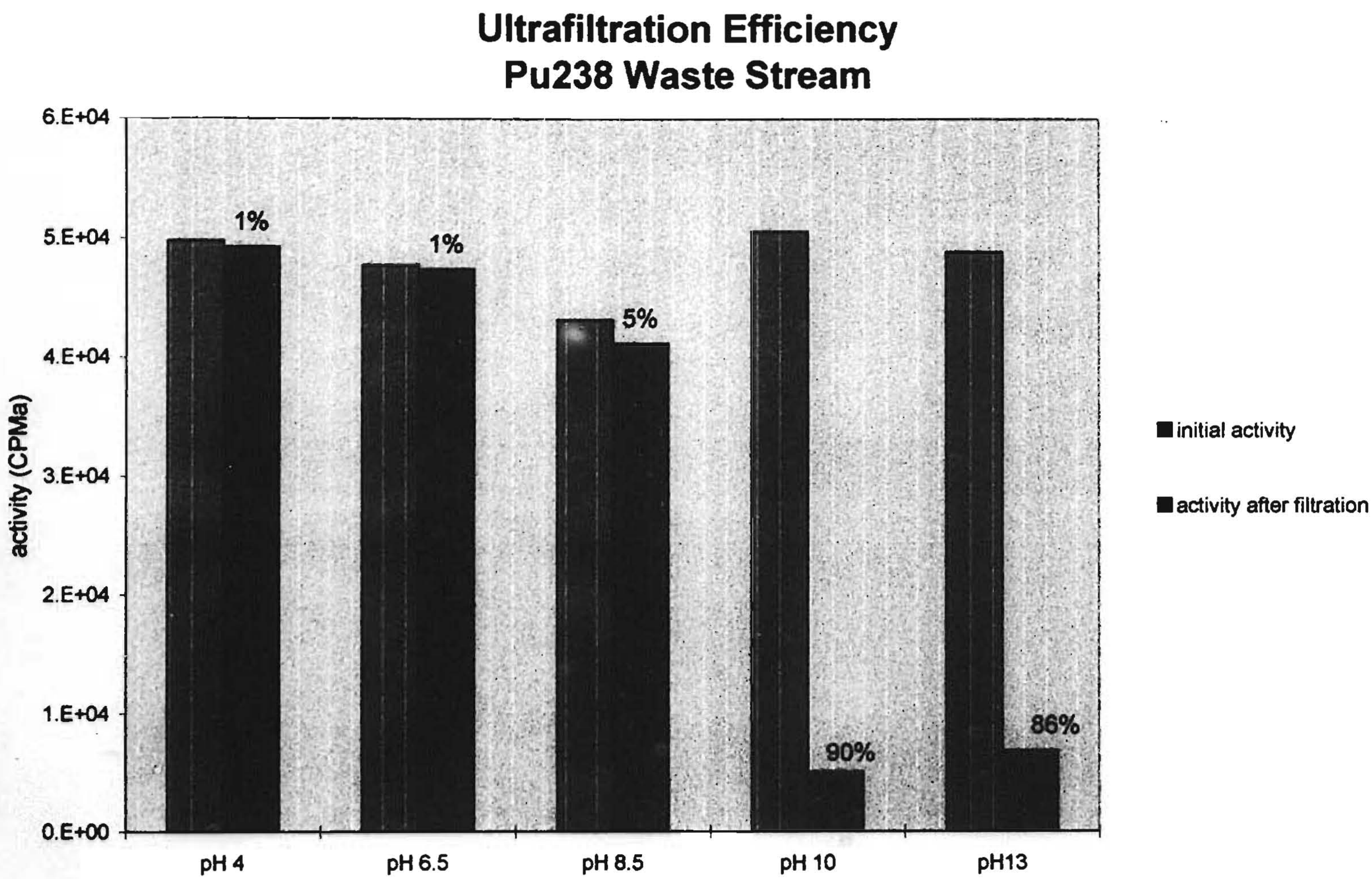
PEIDP - high pH

Polymer Degradation



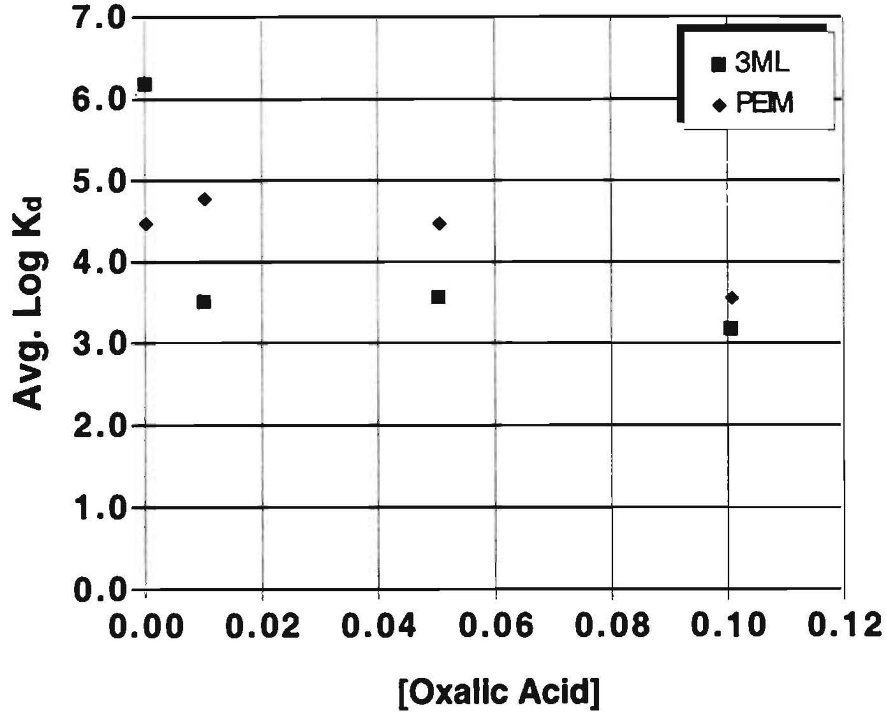
Pu-238 Ascorbic Acid Solution
pH 9





Ultrafiltration Efficiency
Pu238 Waste Stream
6.E+04
5.E+04
4.E+04
3.E+04
2.E+04
1.E+04
0.E+00
activity (CPMa)
1%
1%
5%
90%
86%
pH 4
pH 6.5
pH 8.5
pH 10
pH13
initial activity
activity after filtration

Distribution Coefficients for ^{238}Pu with 0.1% wt Polymer vs. Oxalic Acid Concentration at pH5



Ultrafiltration / Polymer Filtration Treatment of Pu-238 Hydroxide Filtrates

Sample	OHFLT-12A Ci/L (alpha)	OHFLT-12B Ci/L (alpha)
filtrate, pH 13-14	500	1200
UF, no polymer	250	460
pH 7, no polymer	280	680
pH 7, PF #1, 0.5%	48	18
pH 7, PF #2, 0.5%	0.96	0.42
pH 10, no polymer	290	620
pH 10, PF #1, 0.5%	9.9	18
pH 10, PF #2, 0.5%	0.57	0.65

Process Qualification

A series of qualification runs of the scrap recovery process was performed in FY00 on 50 – 100g batches of PuO_2. These runs allowed the throughput, yields and product quality of the process to be determined under realistic operating conditions. Two methods of purifying the Pu-238 were evaluated: direct oxalate precipitation and anion exchange followed by oxalate precipitation.

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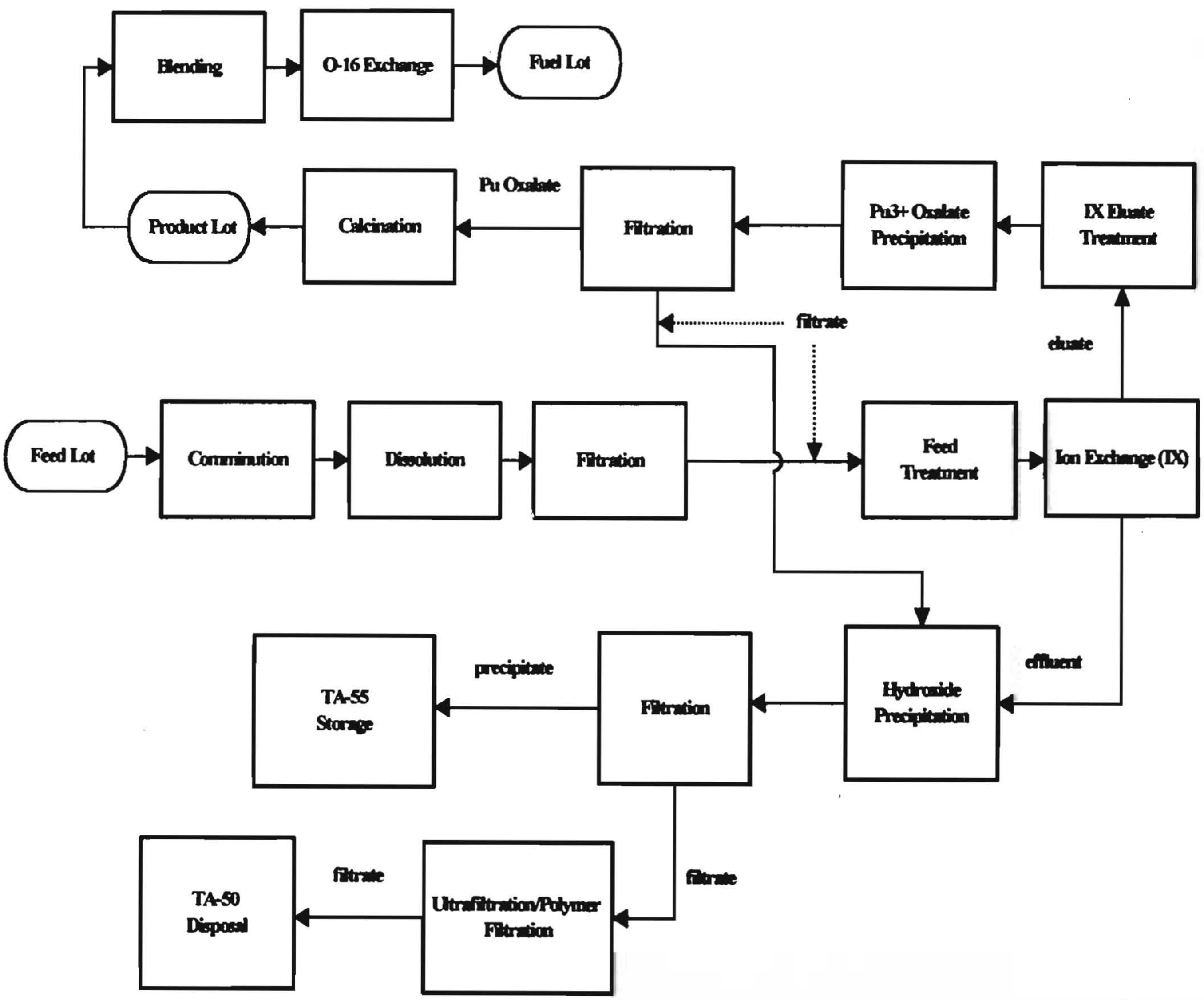
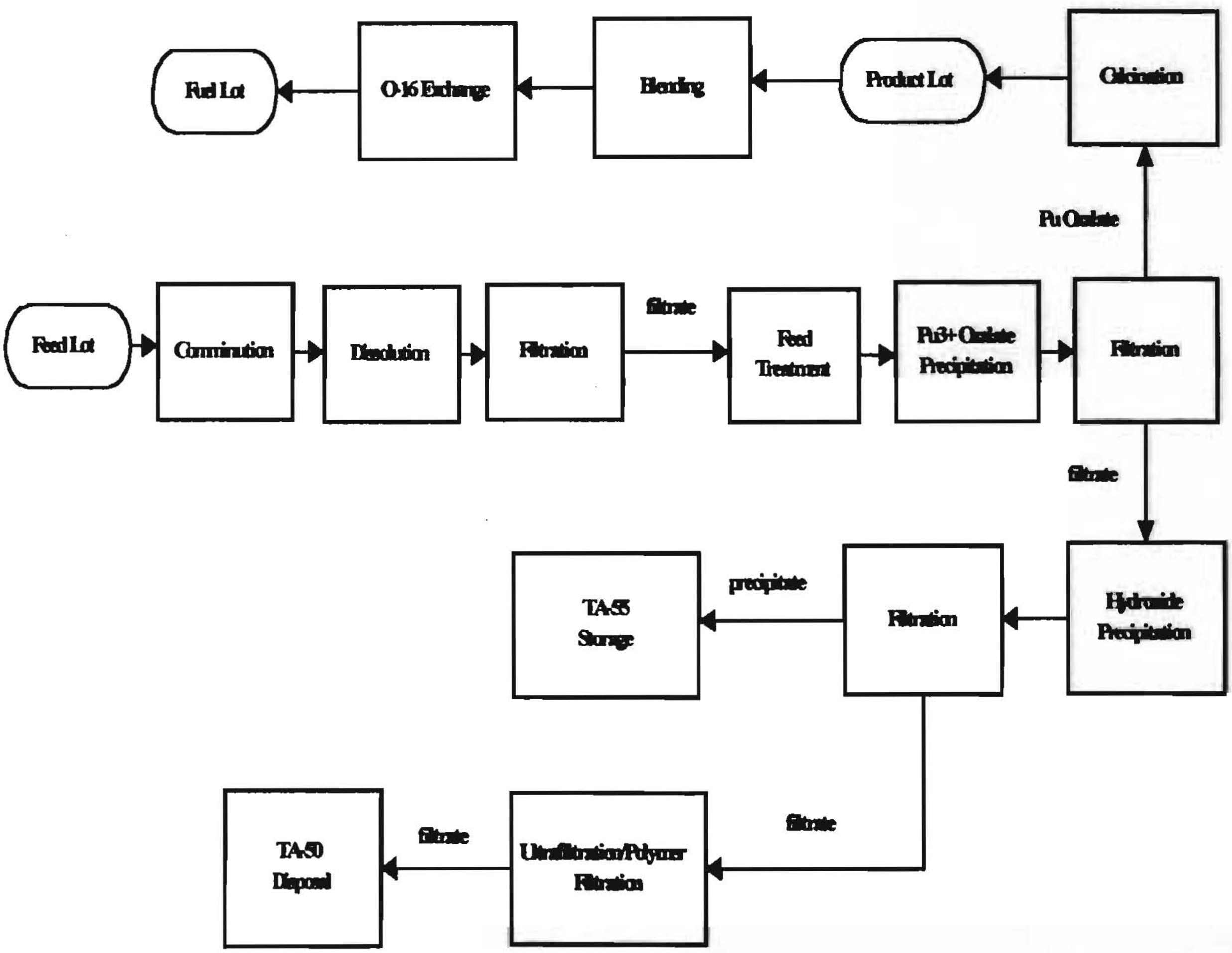
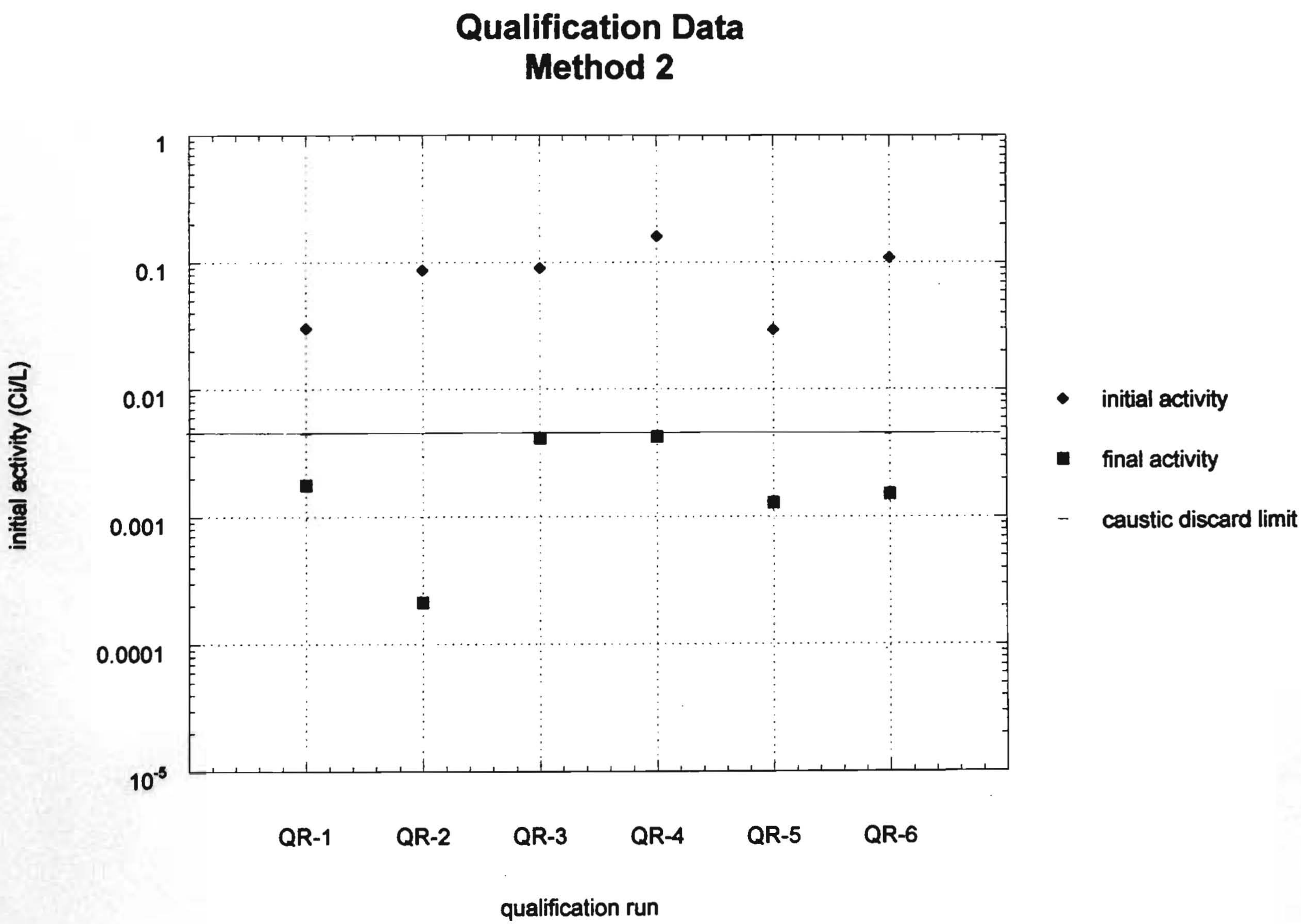
Method 1
Ion Exchange/Oxalate Precipitaion

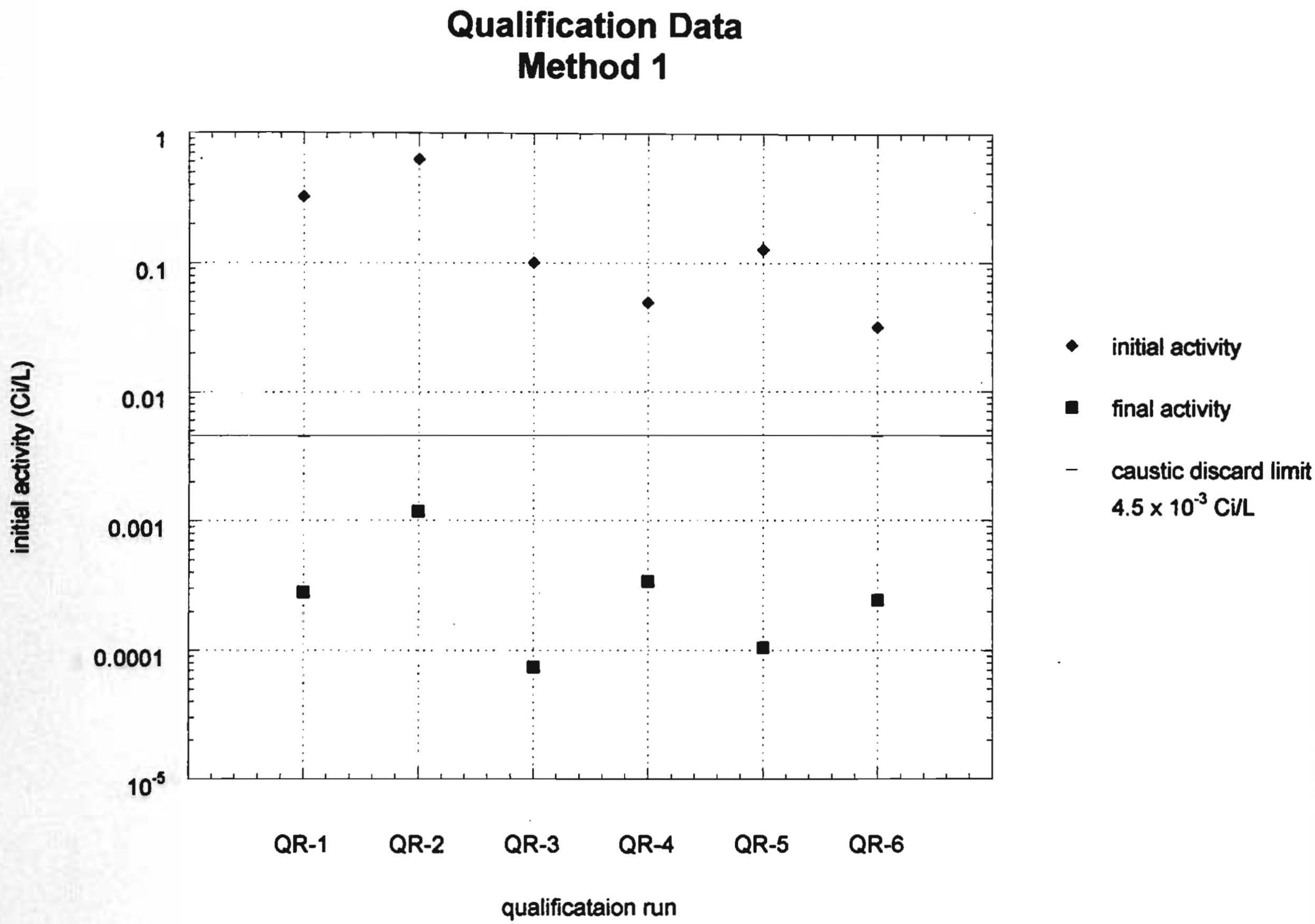


Method 2
Oxalate Filtrate

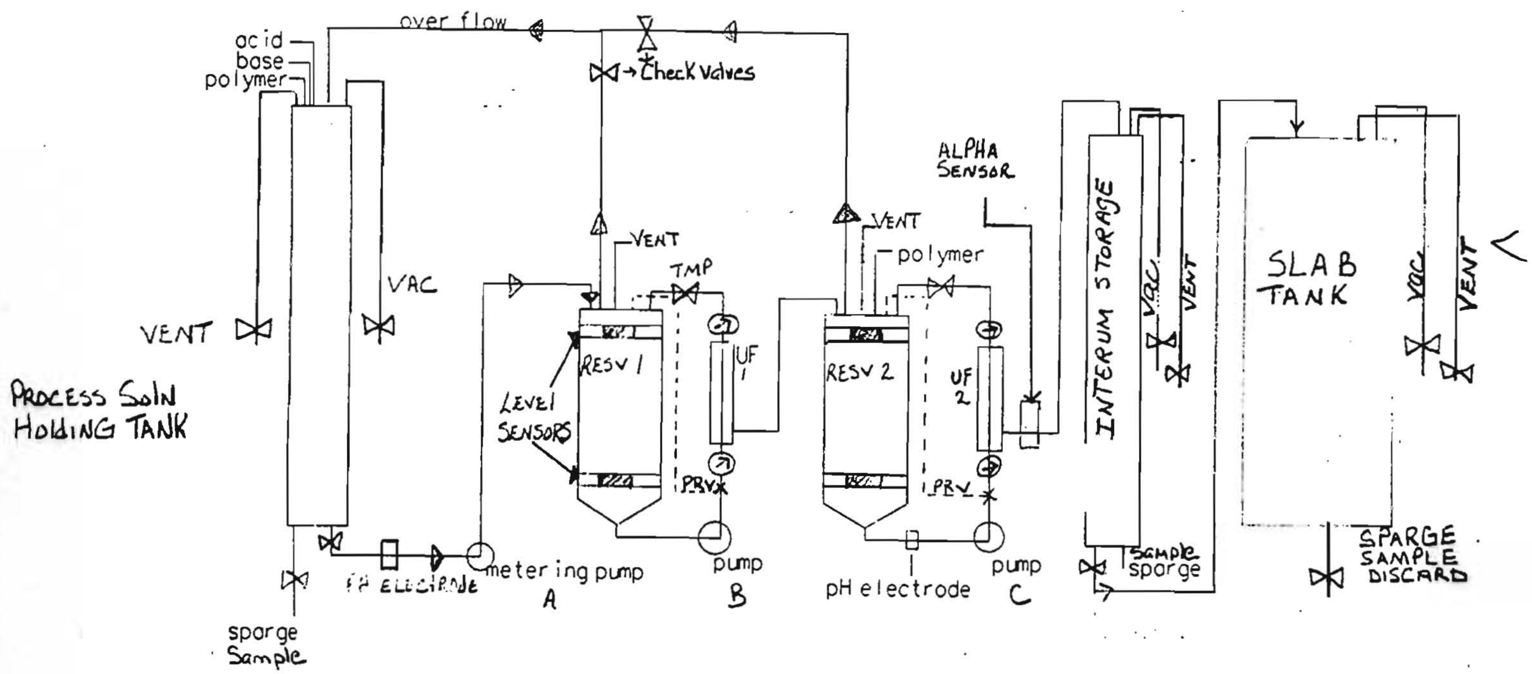




Qualification Data
Method 2
1
0.1
0.01
0.001
0.0001
10^{-5}
initial activity (Ci/L)
QR-1
QR-2
QR-3
QR-4
QR-5
QR-6
qualification run
initial activity
final activity
caustic discard limit


Qualification Data
Method 1
1
0.1
0.01
0.001
0.0001
10^{-5}
initial activity (Ci/L)
QR-1
QR-2
QR-3
QR-4
QR-5
QR-6
qualificataion run
initial activity
final activity
caustic discard limit
4.5 x 10^{-3} Ci/L

Pu 238 Polymer Filtration Process



Conclusions & Future Work

- The permeates from the polymer filtration process met the facility waste disposal requirements.
- Three options for disposition of the retentates include cementation, hydrothermal oxidation, and UV/H_2O_2 oxidation.
- Evaluate UV/H_2O_2 oxidation of oxalate, urea, and hydroxylamine.
- Full-scale scrap recovery operations to begin in FY01.

Acknowledgements

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Elizabeth Foltyn, NMT-9