

Pm/Sm Separation via Ln Resin

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Promethium

- from *Prometheus*, stole fire from heaven
- 4th member of the lanthanide group of elements
- Pm only REE which doesn't occur naturally
- 17 known isotopes of Pm, with atomic masses from 134 to 155
- Longest-lived isotope Pm-145 ($t_{1/2} = 17.7$ y); Pm-147 ($t_{1/2} = 2.6$ y) has been used with a phosphor to produce light or for nuclear-powered batteries.

58 59 60 61 62 63 64

| | | | | | | | |
|----|----|----|----|----|----|----|-----|
| Ce | Pr | Nd | Pm | Sm | Eu | Gd | ... |
|----|----|----|----|----|----|----|-----|

Samarium

- Found in several minerals including monazite and bastnasite
- 21 isotopes of Sm exist
- natural Sm a mixture of several isotopes, 2 of which are radioactive with long half-lives, ^{147}Sm , ^{148}Sm
- ^{151}Sm less abundant as a fission product than ^{147}Pm

58 59 60 61 62 63 64

| | | | | | | | |
|----|----|----|----|----|----|----|-----|
| Ce | Pr | Nd | Pm | Sm | Eu | Gd | ... |
|----|----|----|----|----|----|----|-----|

Important Fission Products

$T_{1/2} > 100$ days

| Radionuclide | $T_{1/2}$ | Decay-mode |
|---|---------------------|----------------------|
| ^{89}Sr | 50.5 d | β^- , γ |
| $^{90}\text{Sr} \rightarrow ^{90}\text{Y}$ | 28.5 y | β^- |
| ^{99}Tc | 2×10^5 y | β^- |
| ^{129}I | 1.6×10^7 y | β^- , γ |
| $^{137}\text{Cs} \rightarrow ^{137\text{m}}\text{Ba}$ | 30.2 y | β^- , γ |
| $^{144}\text{Ce} \rightarrow ^{144}\text{Pr}$ | 285 d | β^- , γ |
| ^{147}Pm | 2.62 y | β^- (224 keV) |
| ^{151}Sm | 90 y | β^- (76 keV) |

Pm-147 accounts for ~10% of the total activity in recent spent fuel while ^{151}Sm and Eu isotopes account for <1%

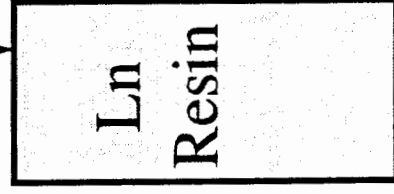
General Separation Approach

*Separate Pm from
common ions and most
other REE's on Ln Resin*

CaHPO₄
scavenge

+

Sm carrier



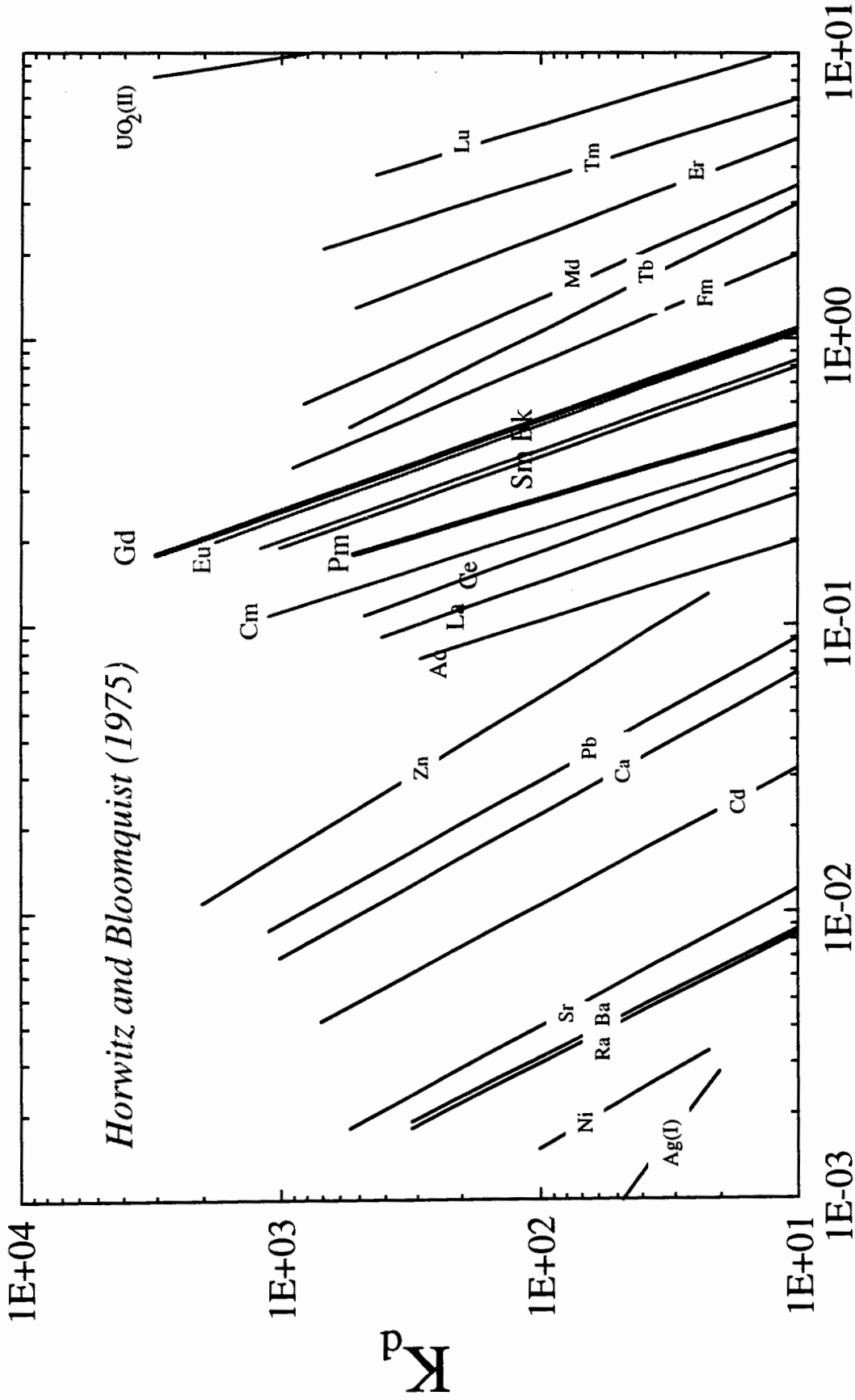
Source prep
OH- ppt

LSC
or
proportional counter

Characteristics of Ln Resin

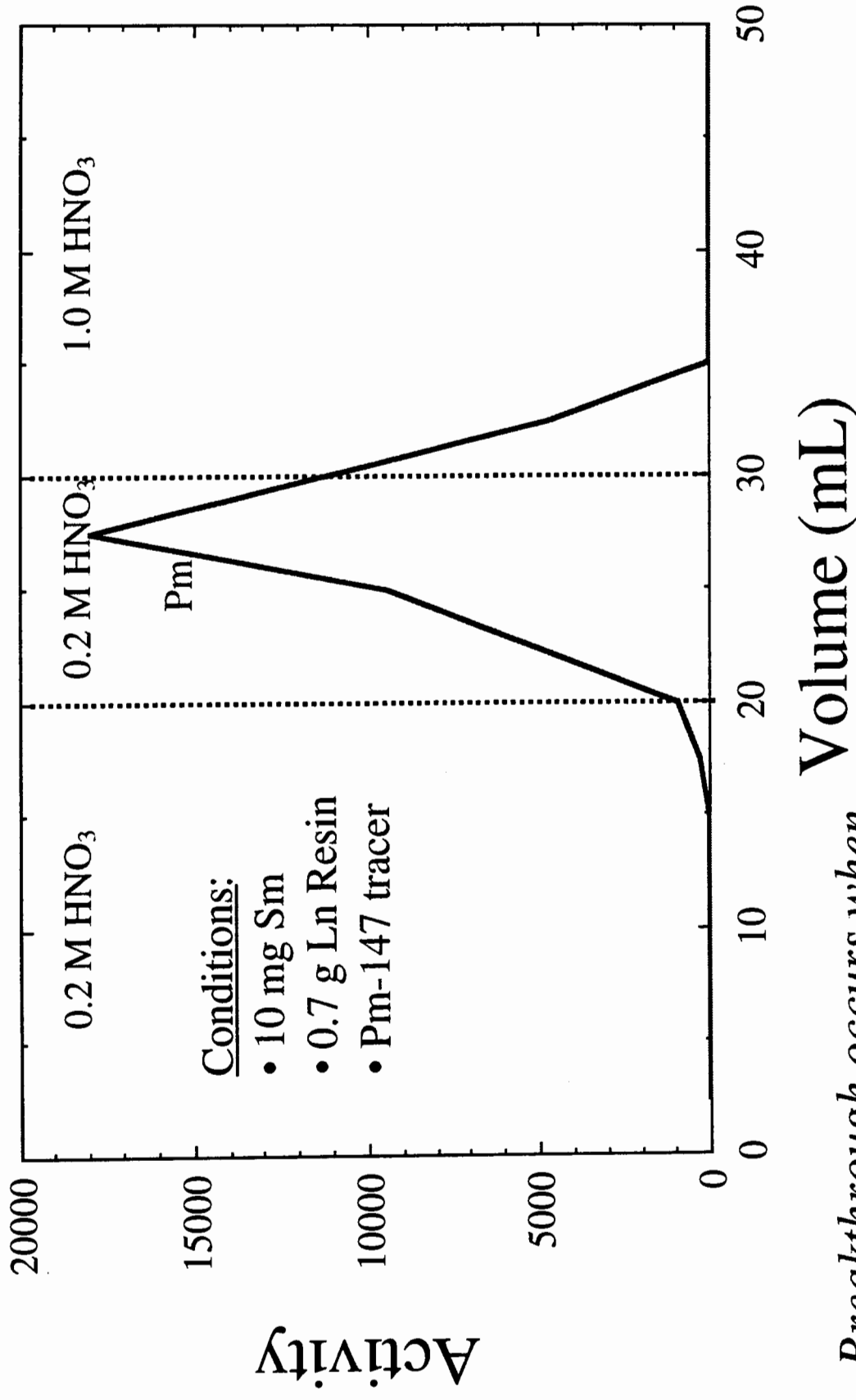
- stationary phase (extractant) →
HDEHP: di(2-ethylhexyl)
orthophosphoric acid
- support → Amberchrom™ CG-71
- particle diameter → 80-160 μm
- Extractant loading → 40 wt %
- 0.7 g bulk Ln Resin packed into Isolab
columns

Chemical Separations Using Ln Resin



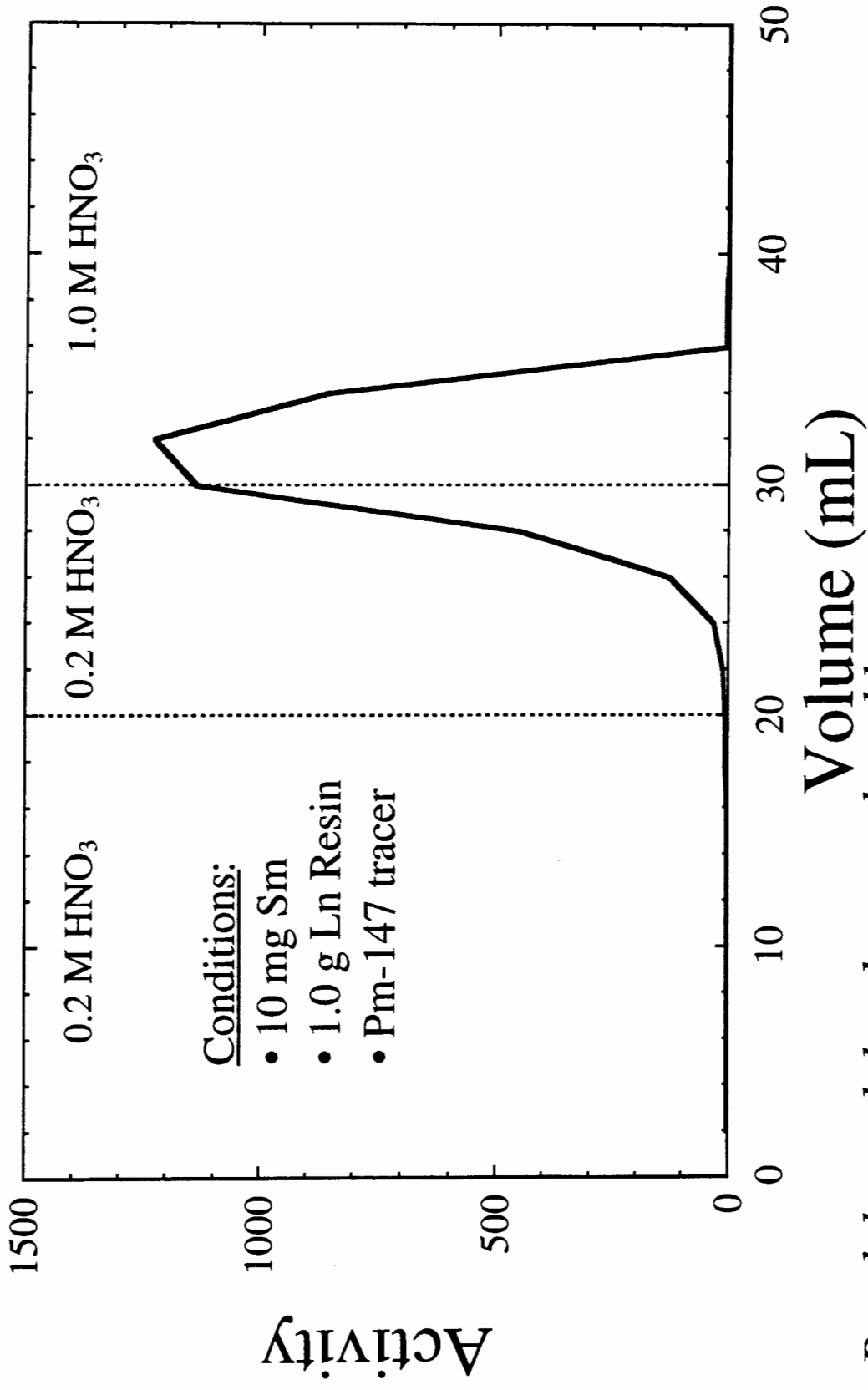
Molarity HNO_3

Sm as a Stable Carrier



*Breakthrough occurs when
using 10 mg Sm*

Increase Amount of Resin

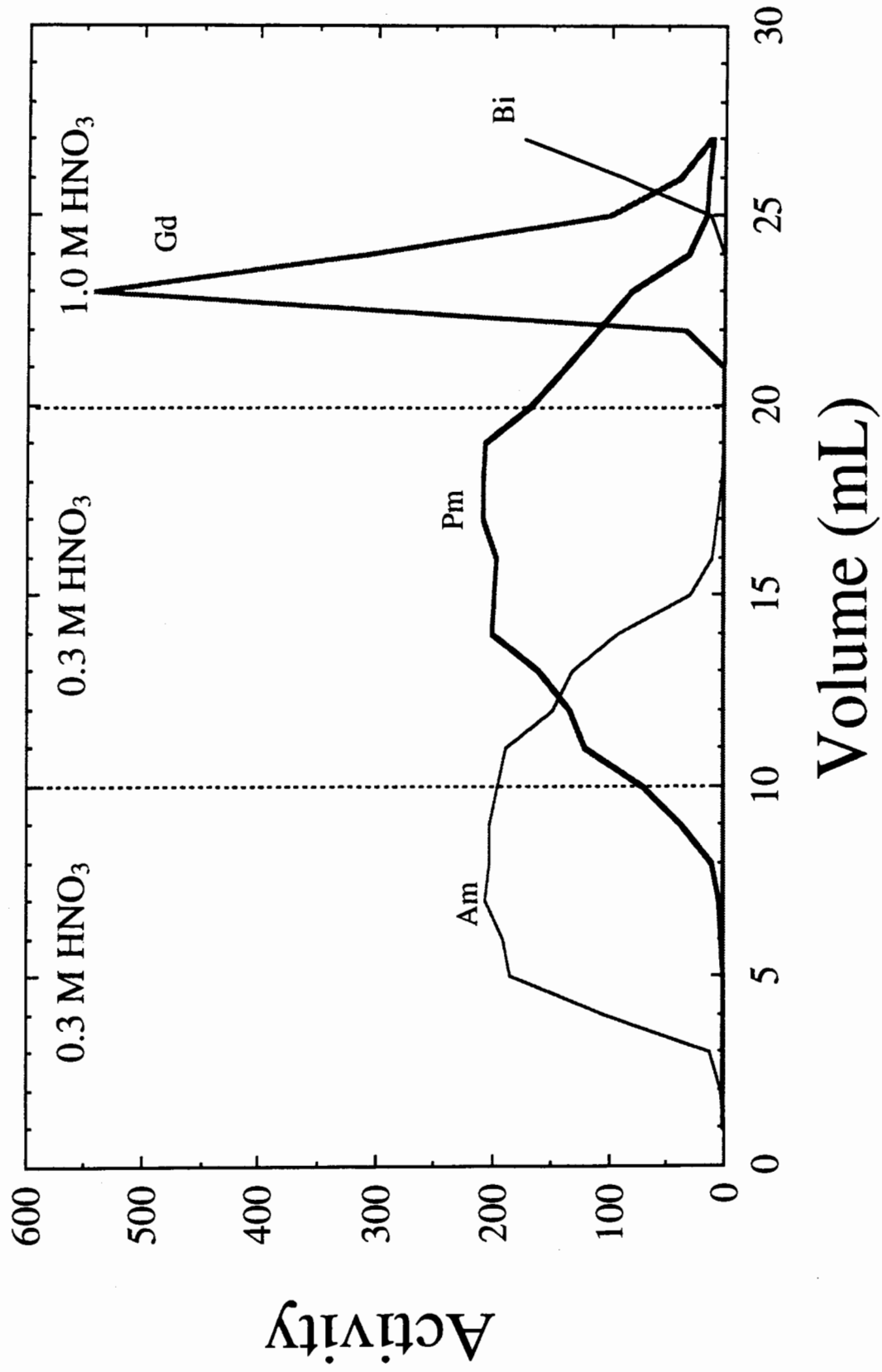


Breakthrough has been reduced but still remains a problem

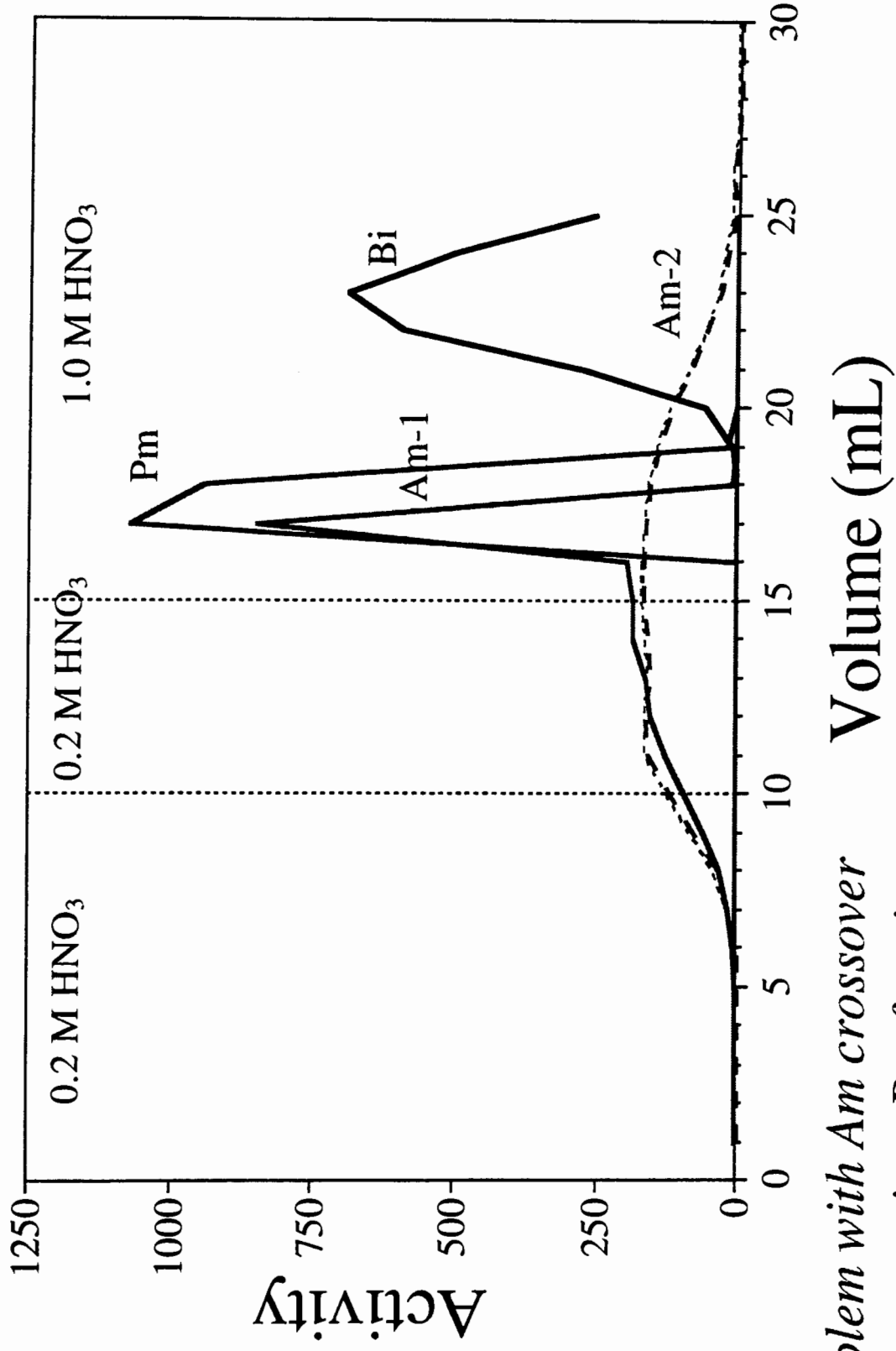
Gd-148 as a Tracer

- Atomic #64 (Pm atomic #61)
- Valance = 3+; very similar in chemistry to Pm
- Pure α -emitter, energy = 3.18 MeV
- $t_{1/2} = 75$ y
- Stable Gd used to make phosphors for color TV's and the metal has unusual superconductivity properties

Load in 0.3M HNO₃



Load in 0.2M HNO₃

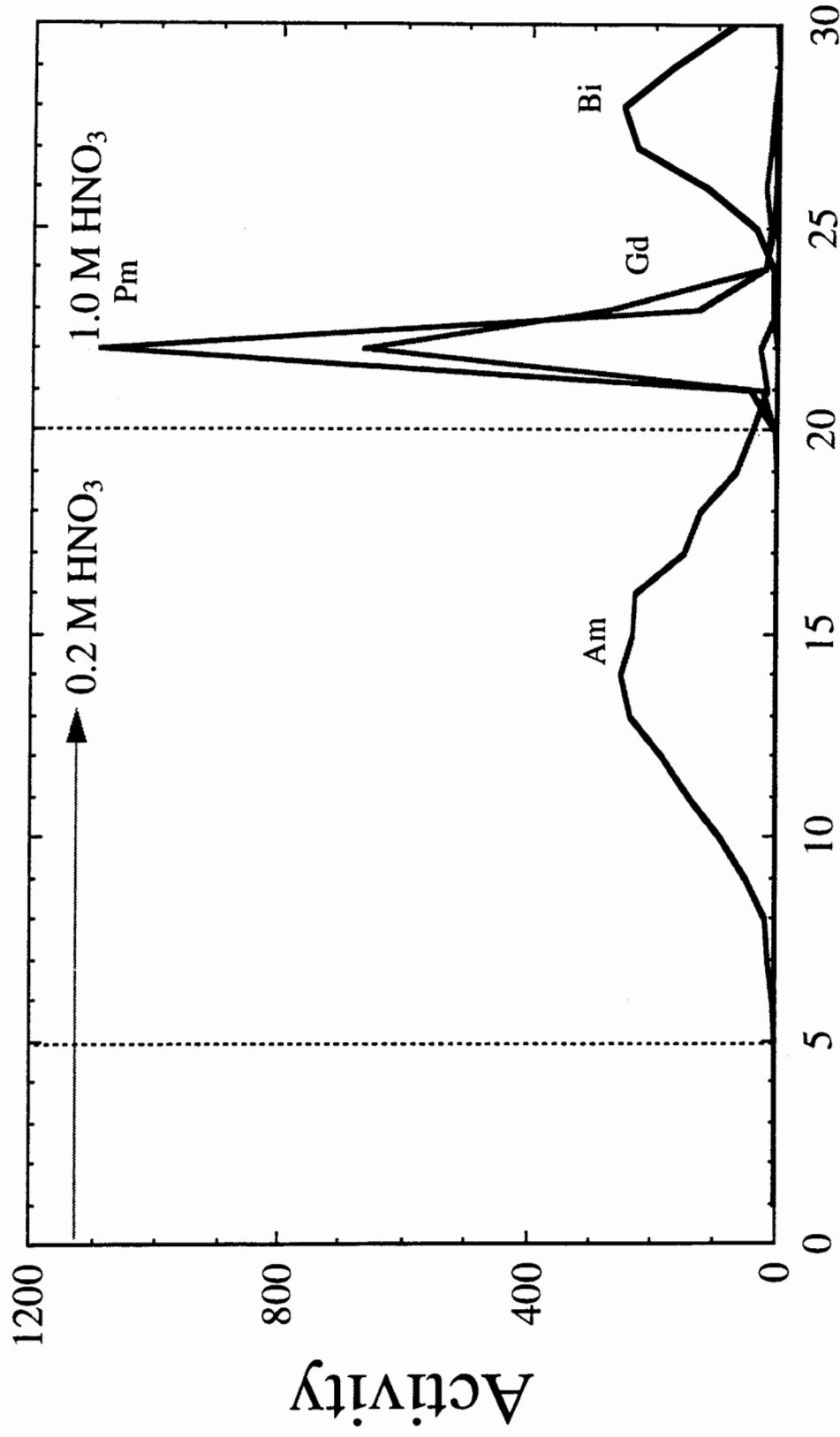


*Problem with Am crossover
into Pm fraction*

Am-2 – no elution step

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Adjust Rinse Volume

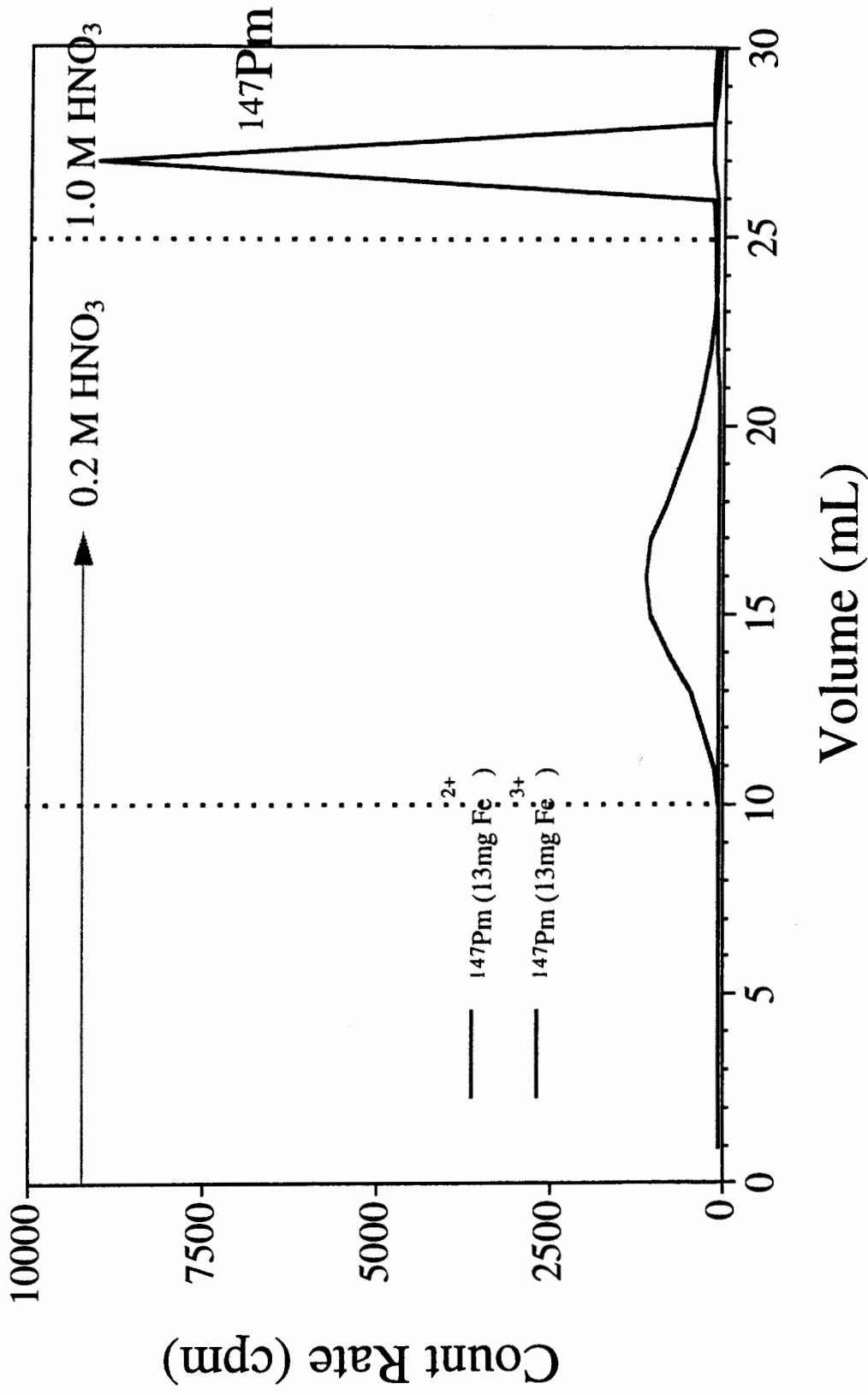


Increase volume of 0.2 M HNO₃ rinse to wash out Am

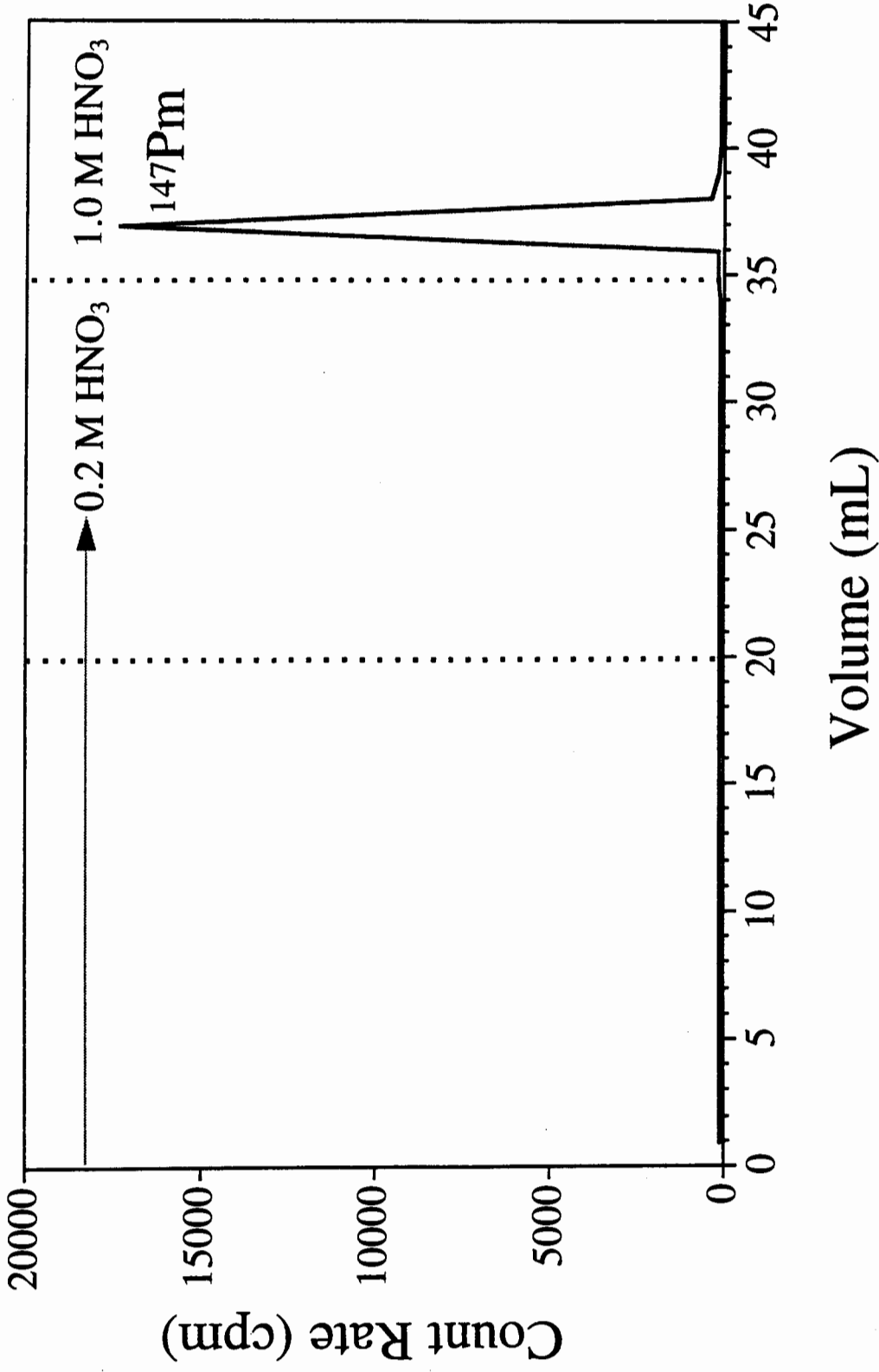
HNO₃ rinse to wash out Am

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Fe Matrix Experiments



Fe Matrix II

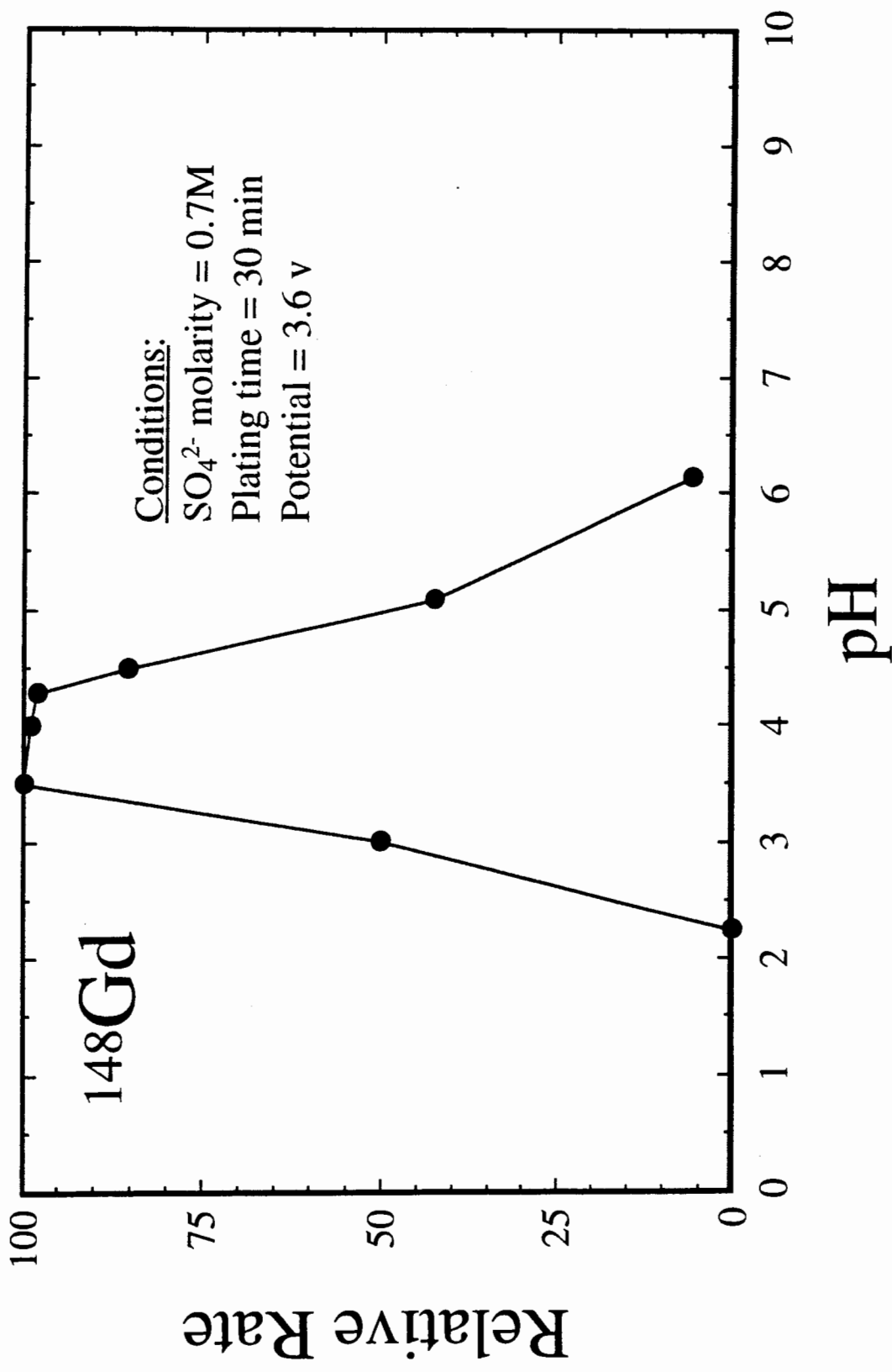


No breakthrough noted with 55 mg Fe(II)

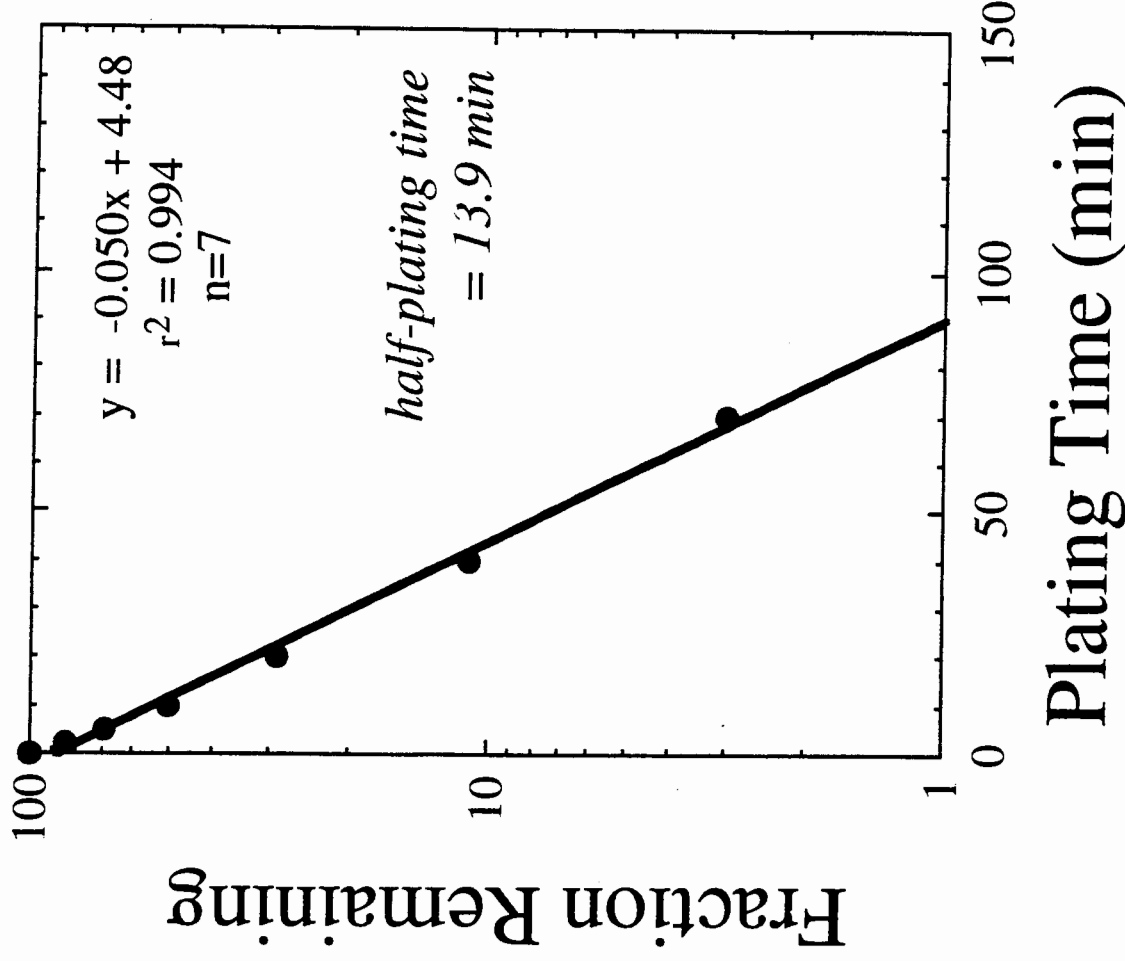
Source Preparation

- Prepare by CeF_3 co-precipitation using a constant amount of carrier (eg. 50 μg Ce), filtration; or
- Electrodeposition—must optimize parameters for Pm/Sm/Gd

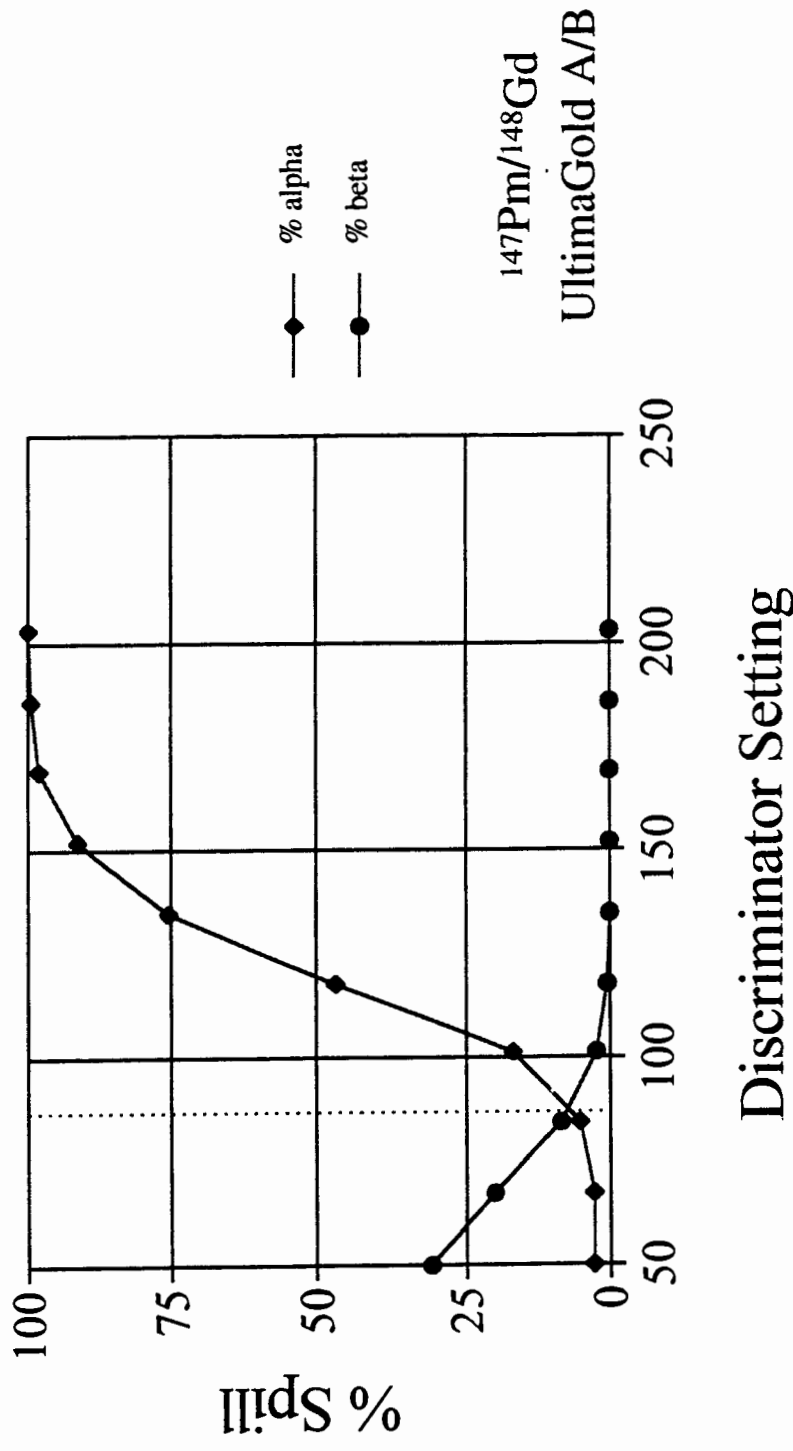
Electrodeposition: pH Variations



Plating Rate Determination

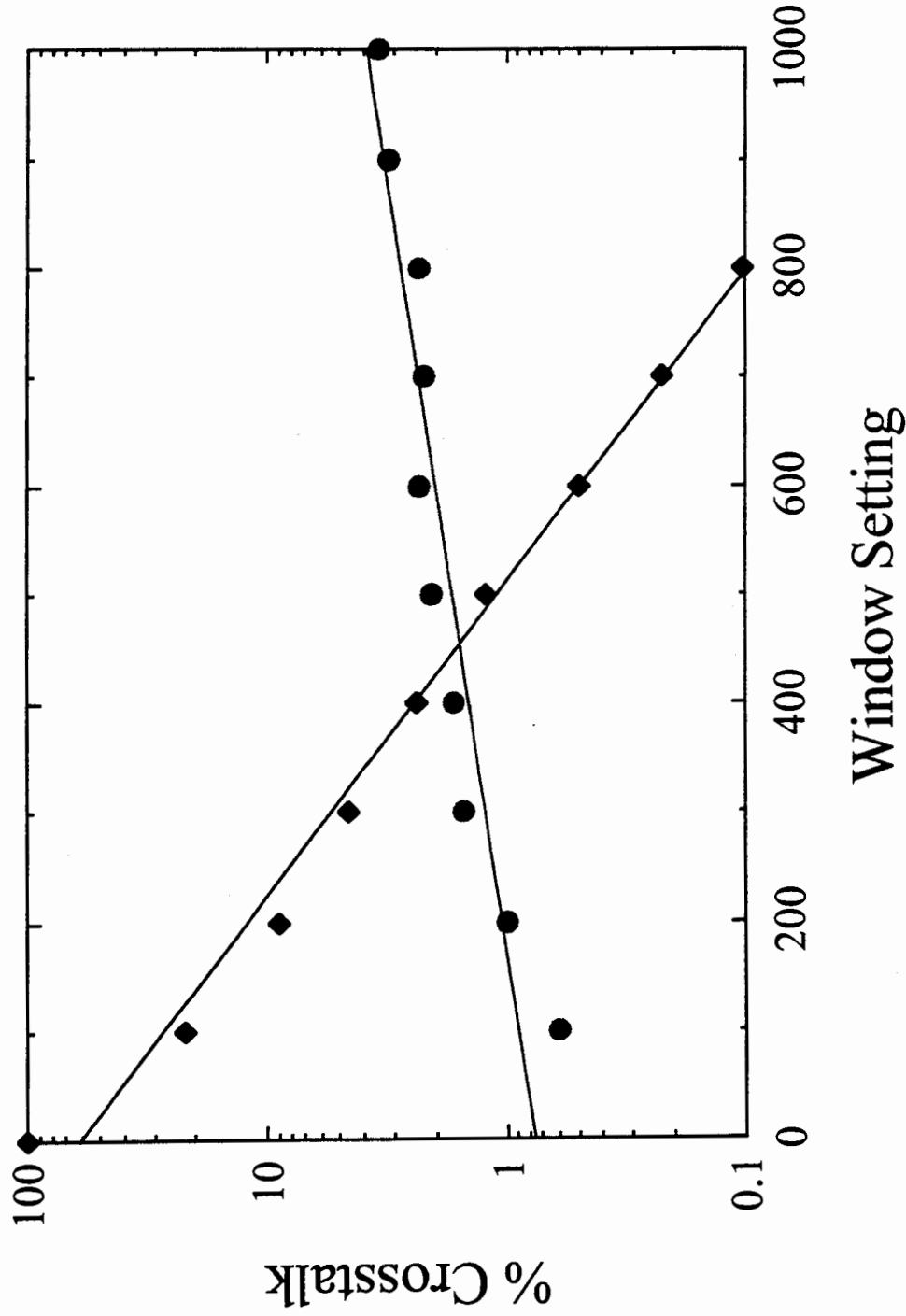


Counting - LSC



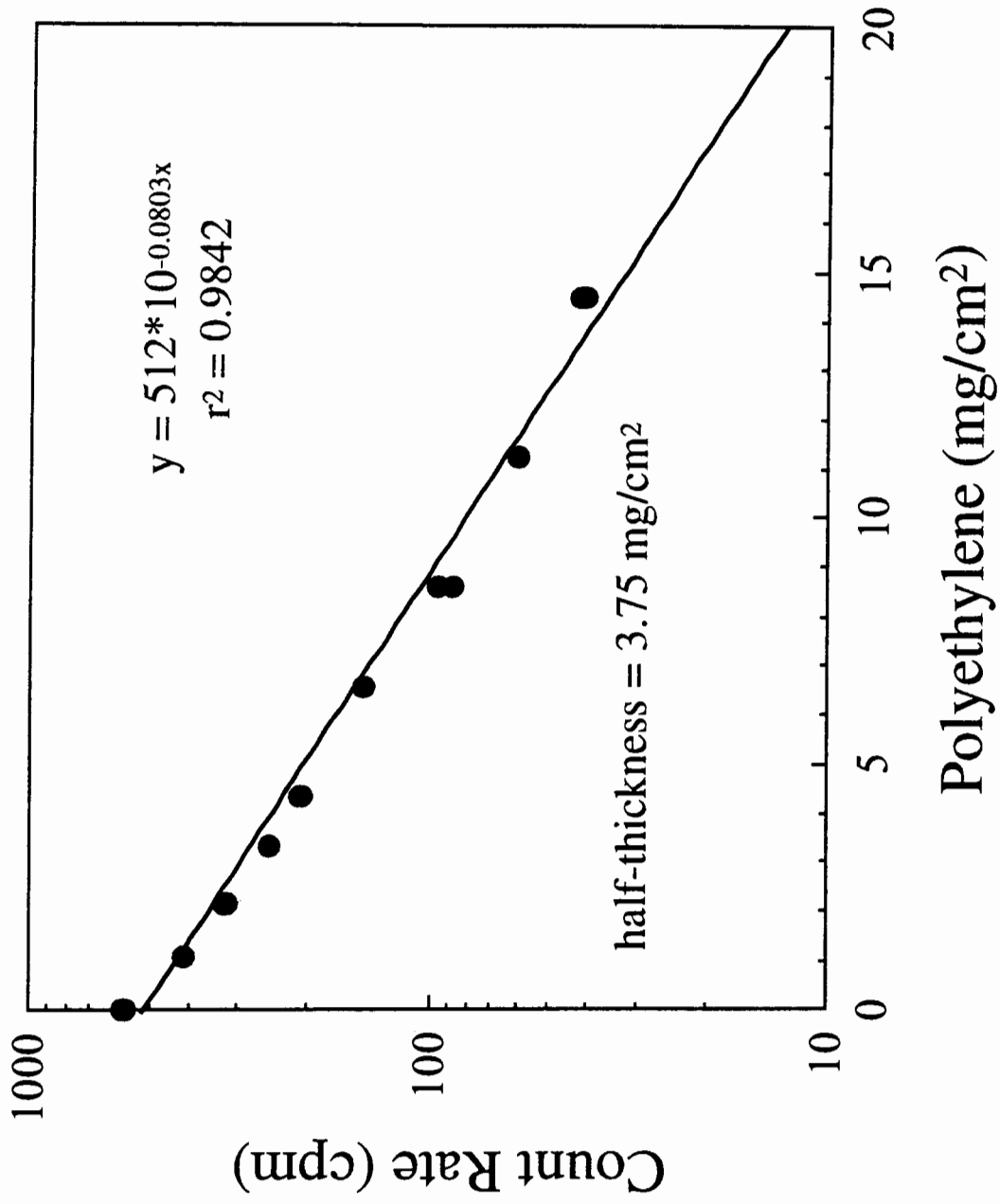
Percent misclassification curves can be used for simultaneous α/β counting by LSC

Proportional Counting



Curves generated using pure α (^{148}Gd) and pure β (^{147}Pm) — set window for minimum a/b crosstalk

Absorbers to Differentiate Pm/Sm



Some Preliminary Tests

- 2 deionized water tests (S1 & S2) with known amount ^{147}Pm (88.8 dpm)
- 1 decontamination test with ~250 dpm total betas (^{60}Co , $^{134,137}\text{Cs}$, $^{89,90}\text{Sr}$) and ^{147}Pm
- Add ^{148}Gd (111.6 dpm)
- 10 mg Ca^{2+} carrier, ppt as CaHPO_4
- Run column, prepare Ce_3F source
- Count on gas flow proportional counter set for simultaneous α - β counting

Results

| Sample | ¹⁴⁸ Gd Yield | ¹⁴⁷ Pm Activity dpm | ¹⁴⁷ Pm Recovery % |
|--------|----------------------------|--------------------------------------|------------------------------------|
| S1 | 98.6 | 77.8 | 88.3 |
| S2 | 97.3 | 77.9 | 88.4 |
| S3* | 93.5 | 84.3 | 95.7 |

*Rinse volume in decontamination experiment lowered to 13 mL

Summary/Future Work

- Ln Resin separates Pm from potentially interfering nuclides;
- ^{148}Gd appears to serve as a useful tracer for ^{147}Pm under the conditions studied;
- Need to improve Pm recovery which is slightly lower than Gd;
- Use beta absorption curves to distinguish ^{147}Pm , ^{151}Sm , ^{144}Pr , *etc.*