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Arsenic Recovery from Irradiated Germanium Targets

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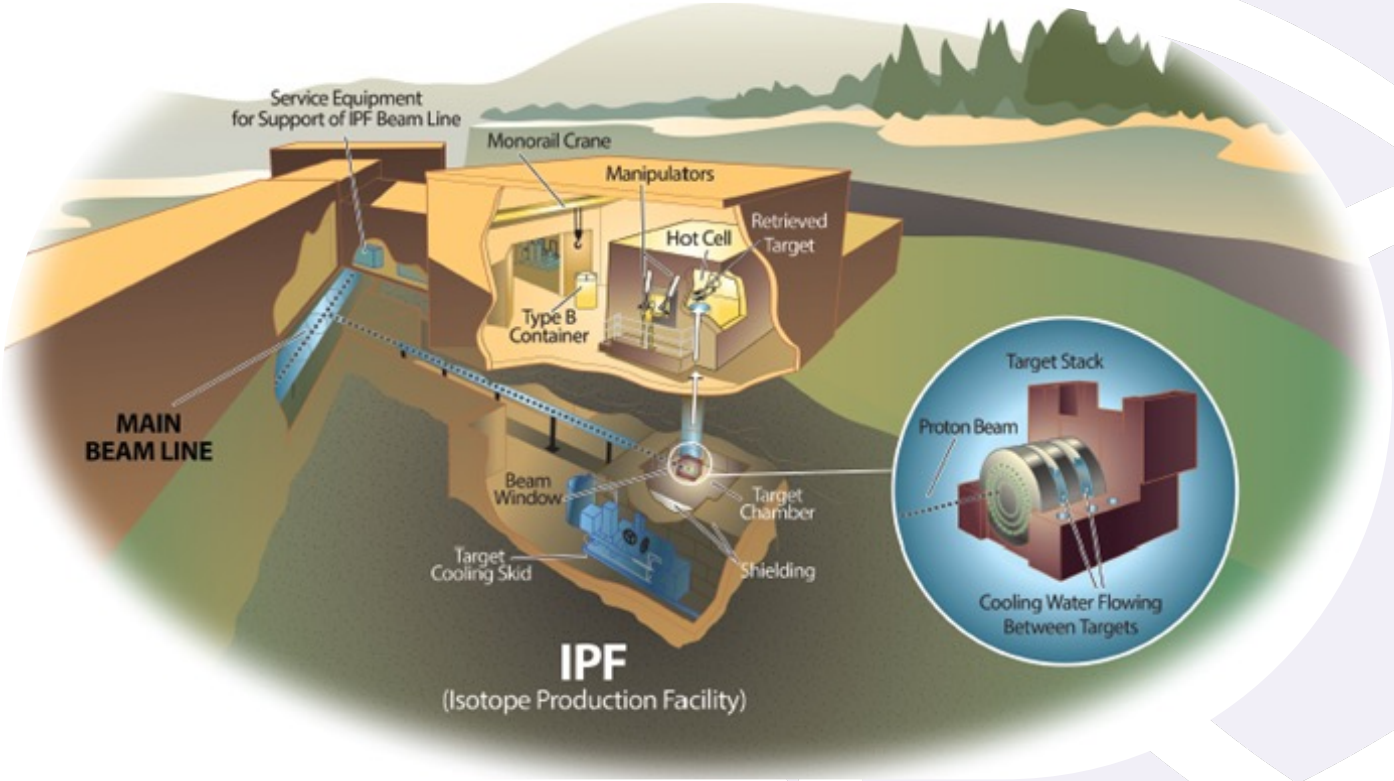
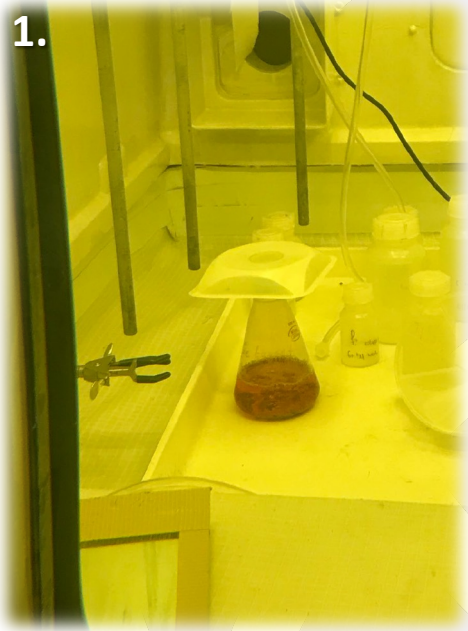
Current Process – Isotope Production Facility



Current Process – Chemistry

- Target dissolution
- $GeCl_4$ distillations
- $AsCl_3$ distillations
- Conversion to As(V)



1. Dissolution of the Ge target in 2:1 HCl:HNO_3 con. Mixture
2. A white precipitate forms (bulk Ge as GeO_2)

Current Process – Possible Improvements

- Target dissolution
- **$GeCl_4$ distillations**
- **$AsCl_3$ distillations**
- Conversion to As(V)

Series of long and delicate distillations required
Critical As volatilization could lead to loss of final product

Selected Literature: Anion-Exchange Separation in Hydrochloric Acid-Acetic Acid Medium

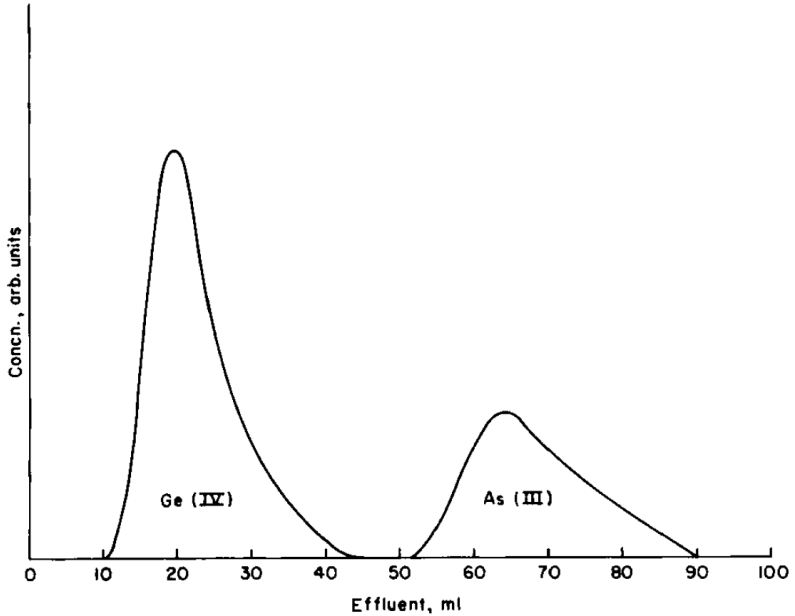


FIG. 1. Separation of 14 mg of germanium from 3.2 mg of arsenic(III) on a 10-g column of Dowex 1-X8 using 90 vol. % acetic acid–10 vol. % 9 *N* hydrochloric acid mixture and a flow rate of 1 ml/4.5 min.

Highly selective method allows for separation of Ge (IV), As(III) and As(V)

Best conditions found to be
90% A. Acid – 10% 9N HCl

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Korkisch, J.; Feik, F. *Sep. Sci.* **1967**, *2* (1), 1–9.

Selected Literature: Anion-Exchange Separation in Hydrochloric Acid Medium

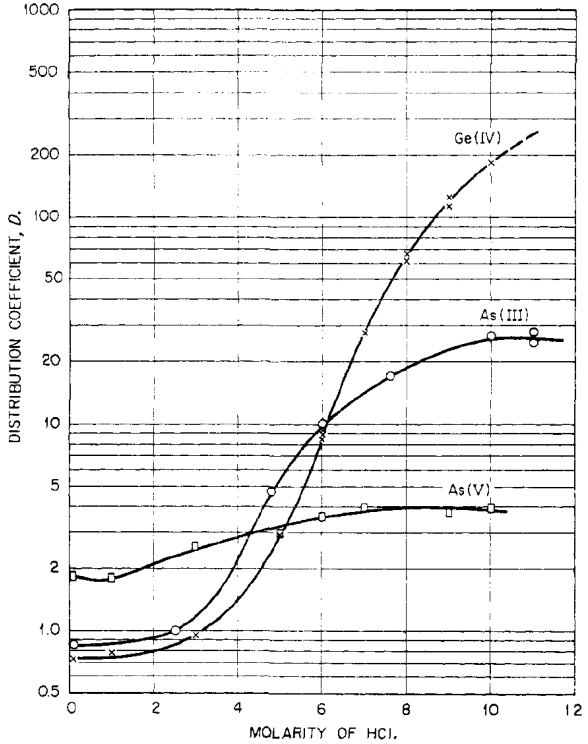


Fig. 1.—Adsorption of Ge(IV), As(III) and As(V).

Strong adsorption of both Ge(IV) and As(III) at high HCl concentration

As(III) adsorption levels off at 10M HCl while Ge(IV) still increases

Separation of As(V) from Ge(IV) readily achievable at high HCl concentration (As(V) and Ge(IV) would require larger volumes)

Nelson, F.; Kraus, K. A. *J. Am. Chem. Soc.* **1955**, *77* (17), 4508–4509.

Recovery of ^{73}As and ^{74}As

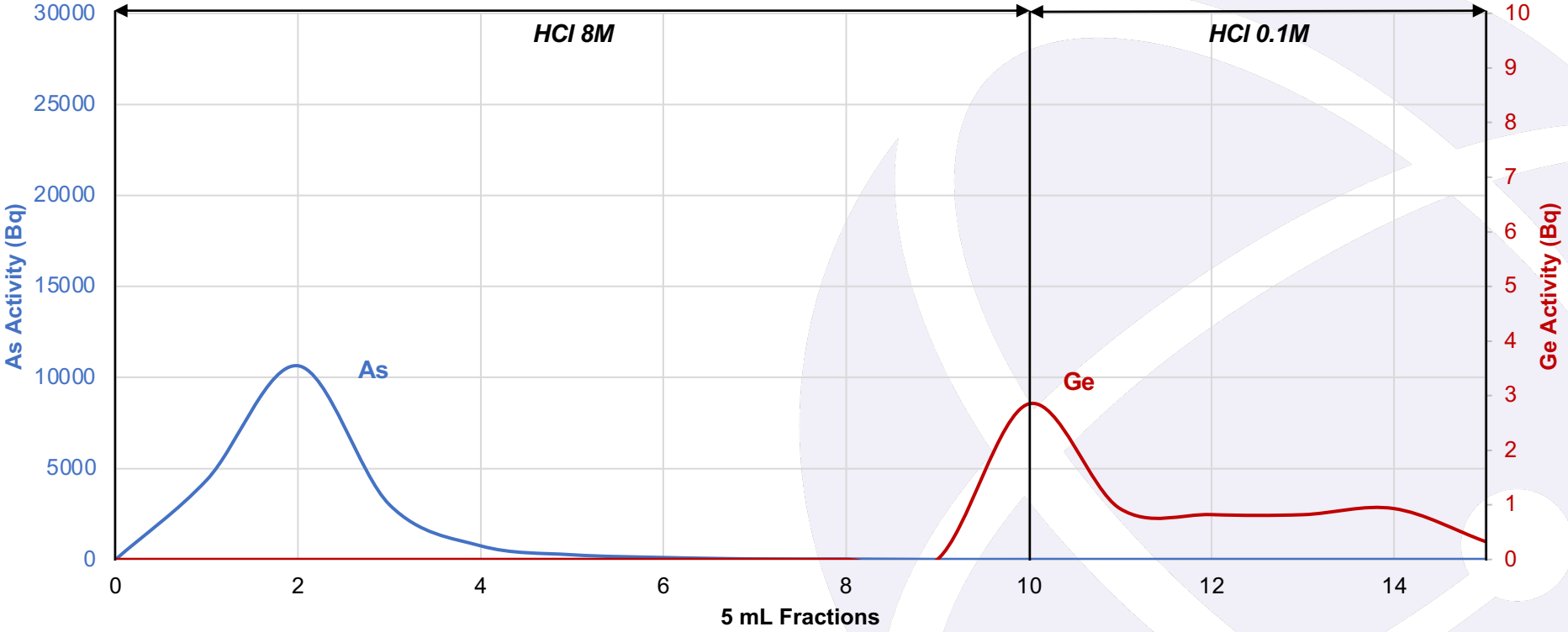
Normal process:
^{73}As (80.30 days, 53 KeV)
Co-produced ^{74}As (17.77 days, 595 KeV) is used as tracer during separation

^{68}Ge (270.93 days)
Its daughter ^{68}Ga (67.71 min, 1077 Kev) also used as tracer

R&D Improvement:
Older material (dissolved target solutions) available

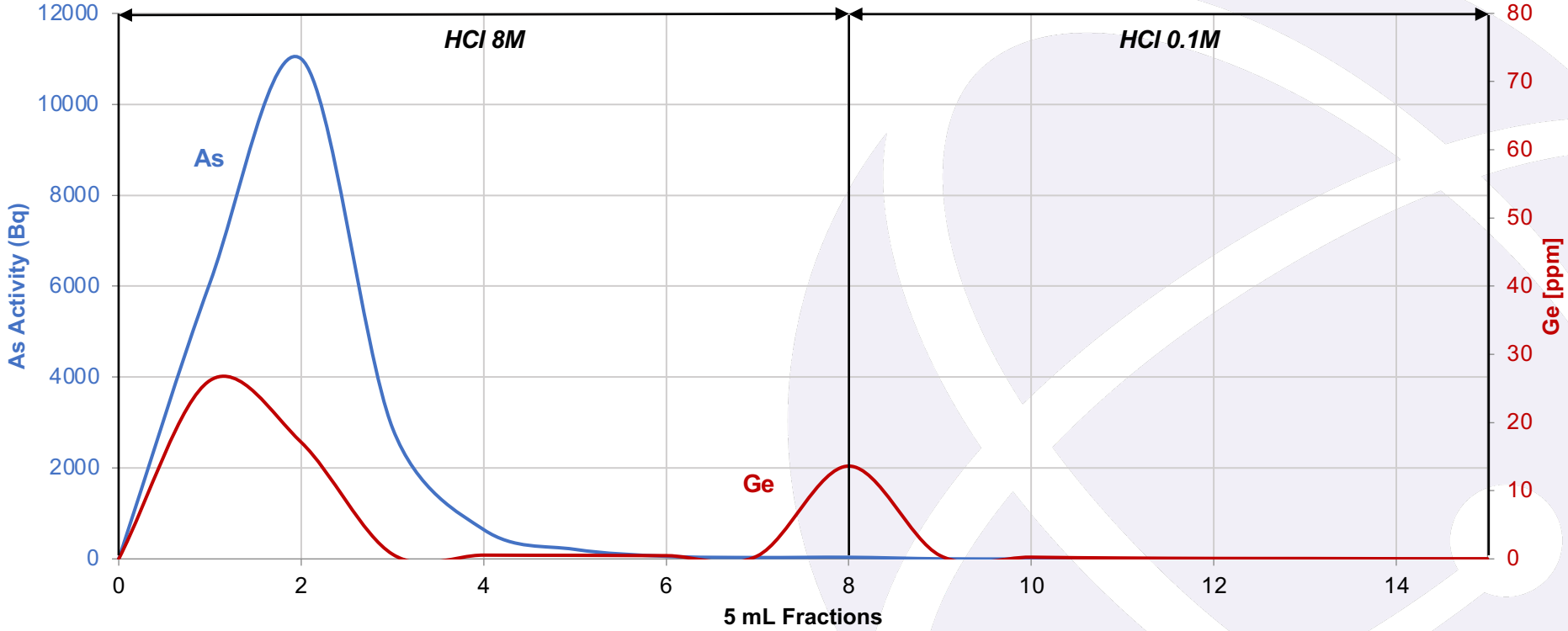
↳ Lower activity, and difficult tracking

Anion-Exchange Separation – 8M HCl



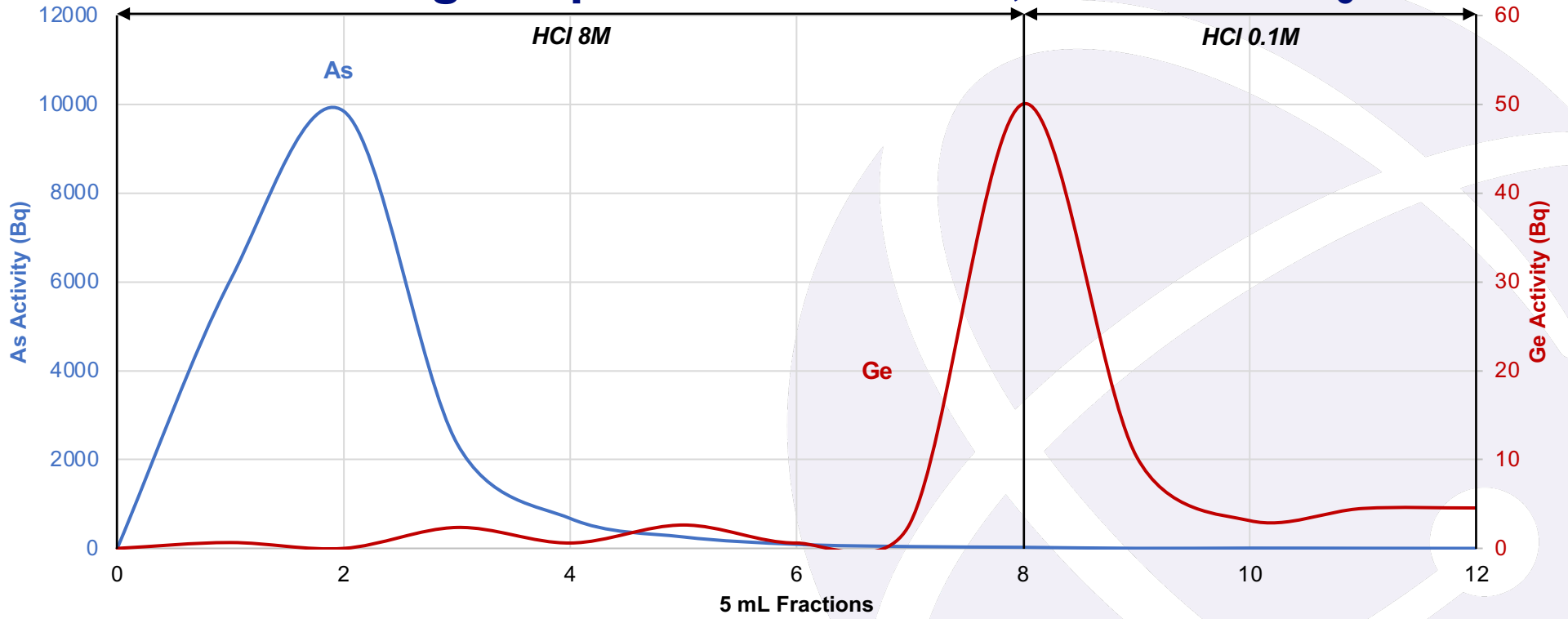
Elution profile of the separation of As and Ge in HCl media followed by γ–ray spectrometry.
5mL HCl spiked with 100 µL radiotracers solution, and loaded onto 1.2822 g of AG1-X8 resin.
(-) ^{68}Ge activity at 511 Kev, (-) ^{73}As activity at 53 Kev.

Anion-Exchange Separation – 8M HCl, Spiked Ge ICP Std



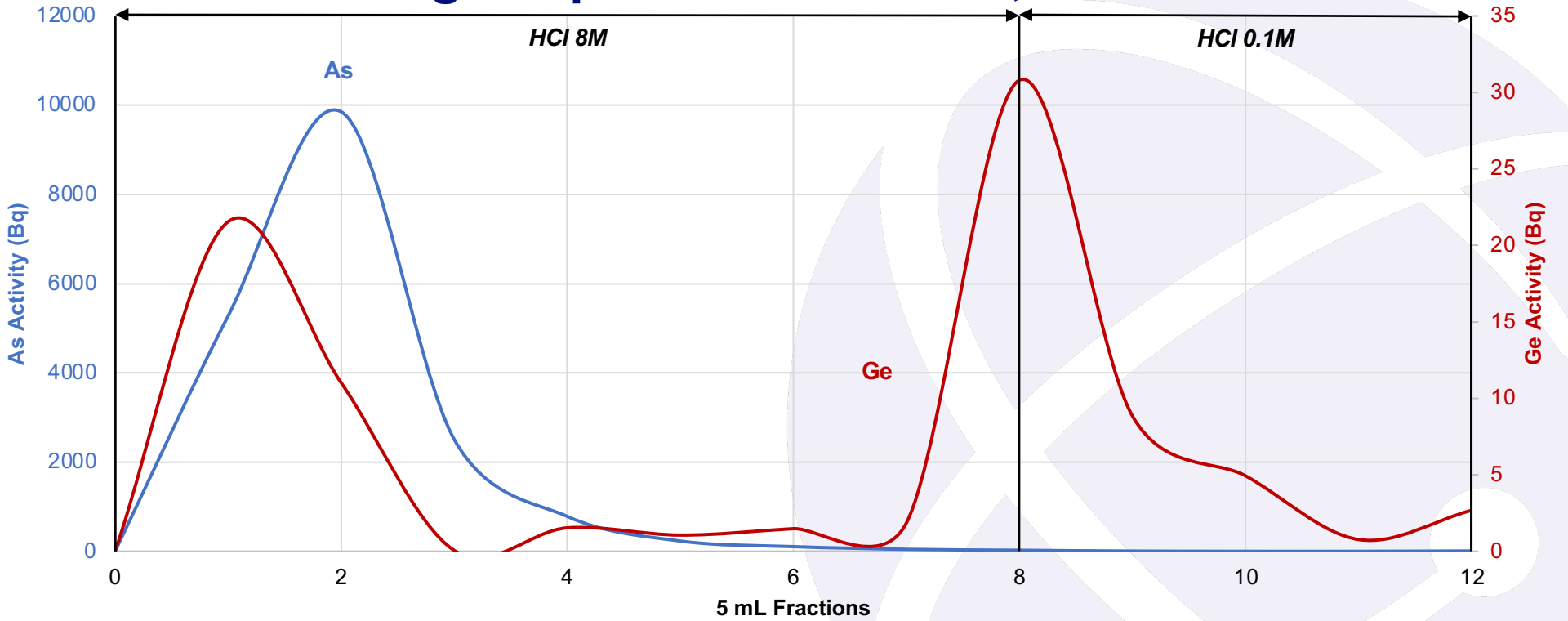
Elution profile of the separation of As and Ge in HCl media followed by γ–ray spectrometry and ICP-OES.
4mL HCl spiked with 750 µL radiotracers solution, 500 µL Ge ICP std, and loaded onto 1.3547 g of AG1-X8 resin.
(-) Ge at 209.426 nm, (-) ^{73}As activity at 53 Kev.

Anion-Exchange Separation – 8M HCl, Traces HNO_3



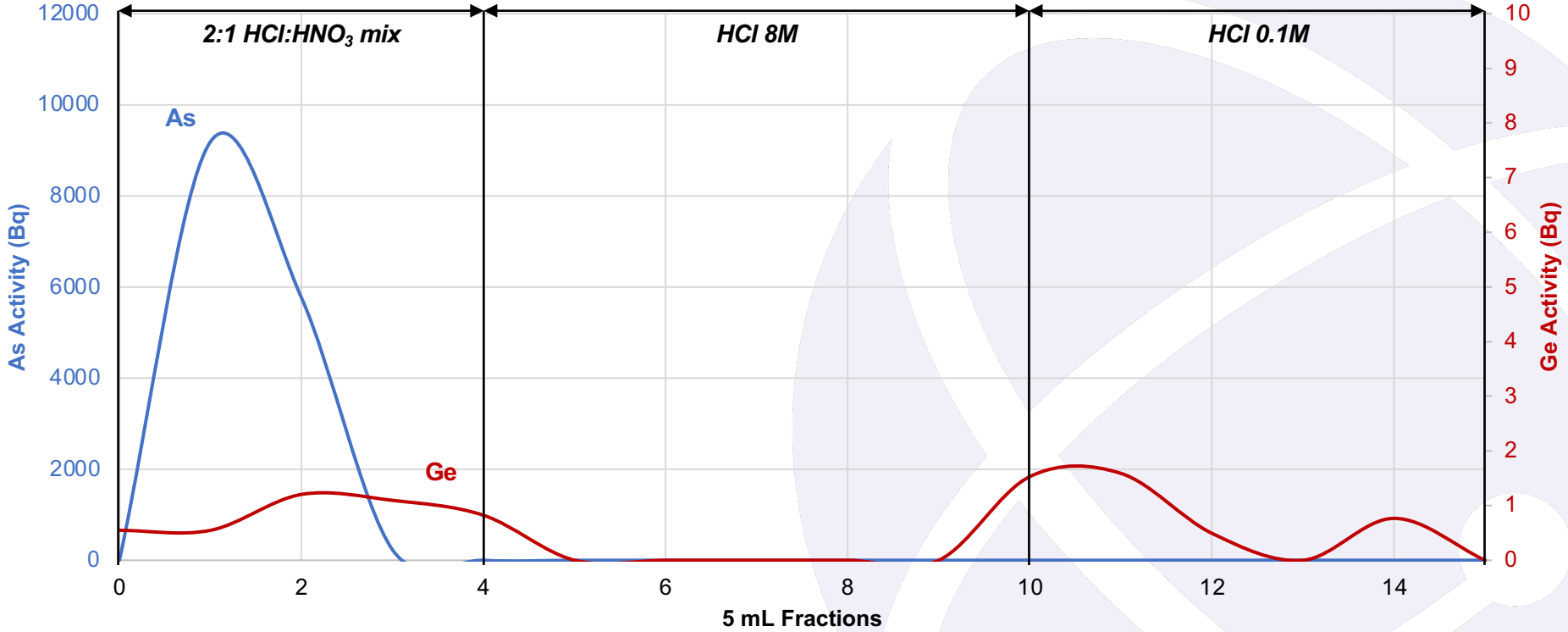
Elution profile of the separation of As and Ge in HCl media followed by γ–ray spectrometry.
4mL HCl spiked with 750 µL radiotracers solution, 33 µL HNO_3 con, and loaded onto 1.3383 g of AG1-X8 resin.
(-) ^{68}Ge activity at 511 Kev, (-) ^{73}As activity at 53 Kev.

Anion-Exchange Separation – 8M HCl, Traces HF



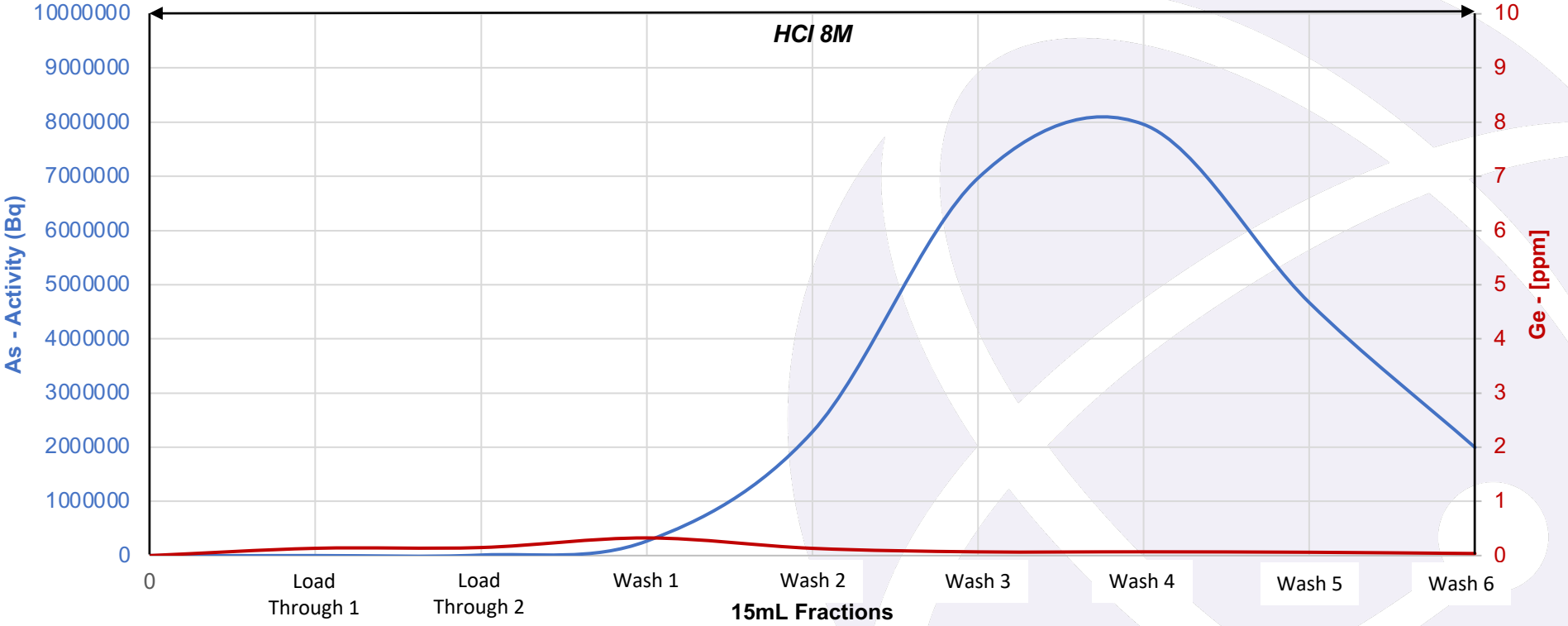
Elution profile of the separation of As and Ge in HCl media followed by γ–ray spectrometry.
4mL HCl spiked with 750 µL radiotracers solution, 20 µL HF con, and loaded onto 1.3526 g of AG1-X8 resin.
(-) ^{68}Ge activity at 511 Kev, (-) ^{73}As activity at 53 Kev.

Anion-Exchange Separation – 2:1 HCl:HNO_3 con



Elution profile of the separation of As and Ge in HCl media followed by γ–ray spectrometry.
5 mL of 2:1 HCl:HNO_3 mixture spiked with 100 µL radiotracers solution, and loaded onto 1.3211 g of AG1-X8 resin.
(-) ^{68}Ge activity at 511 Kev, (-) ^{73}As activity at 53 Kev.

Anion-Exchange Separation – Hot Cell test



Elution profile of the separation of As and Ge in HCl 8M followed by γ–ray spectrometry and ICP-OES. 31.3910 g of dissolved target load solution (30 mL) loaded onto 21.2188 g (30 mL) of AG1-X8 resin. (-) Ge at 209.426 nm, (-) ^{73}As activity at 53 Kev.

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Conclusions and Future Work

- Confirmed feasibility of As/Ge separation via anion exchange resin
- Fast, quantitative separation can be easily implemented in process
- Streamlined process

- Implementation on even larger scale for next batch (Oct 2022)

Acknowledgments

LANL Isotope Team

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Veronika Mocko

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