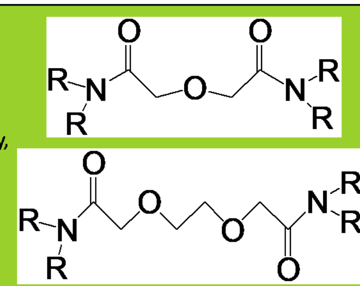
The Purification of Ac-225 using Extraction Chromatographic Resins

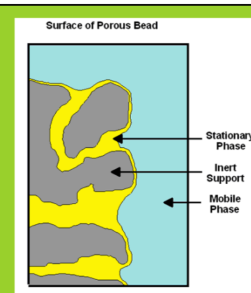
Madeleine A. Eddy[1], Ed Rush[1], Daniel R. McAlister[1]

[1]Eichrom Technologies, LLC Lisle, IL, USA

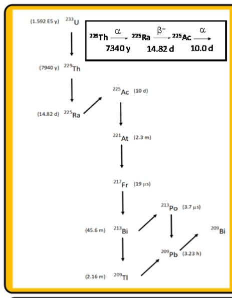
Abstract: Extraction chromatographic (EXC) resins consist of hydrophobic extractants physically adsorbed onto porous inert supports. Combining the selectivity of solvent extraction reagents with the ease of use of chromatography, EXC resins have been used extensively in the separation of radionuclides in a number of applications, including bioassay, environmental analysis, geochronometry, and radionuclide production for industry, radiochemistry, and nuclear medicine. EXC resins based on the diglycolamide (DGA) and dioxaoctanediamide (DOODA) extractants show particular promise for the purification of Ac-225 from Th-229, Th-232 spallation targets, and (p,n) reactions on Ra-226 targets. The DGA and DOODA resins purify Ac-225 from Th, Ra, Ra-226 daughters, Th-229 daughters, Th-232 spallation byproducts, and common reagent impurities. Utilizing both DGA and DOODA resins in combination facilitates the production of highly pure Ac-225 in a matrix compatible with chelator chemistry.

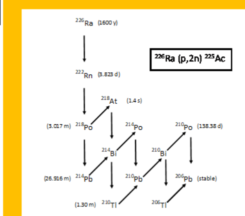


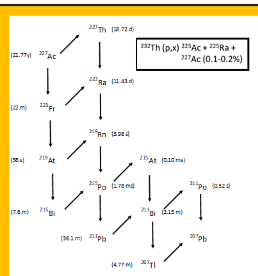
DGA and DOODA Extractants. R = n-octyl



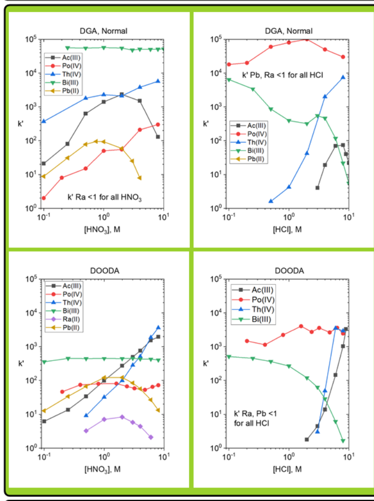
Extractant on EXC Resin





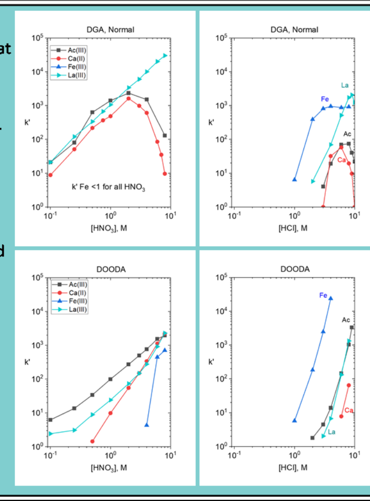


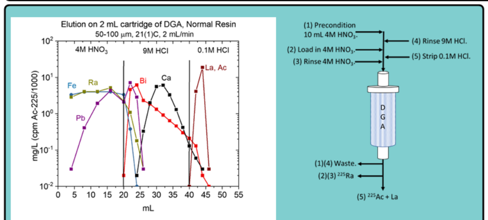
^{225}Ac production: There are several possible routes to large-scale production of ^{225}Ac, including the decay of long-lived ^{229}Th source material, proton irradiation of ^{226}Ra targets, and proton spallation on ^{232}Th targets. Each of these production routes poses unique separation challenges. Separation of ^{225}Ac and ^{225}Ra from **^{229}Th** source material starts with removal of the ^{229}Th using solvent extraction or ion exchange from HNO_3. DGA or DOODA resin can then be used to separate ^{225}Ac from ^{225}Ra and other ^{229}Th progeny and reagent impurities. Separation of ^{225}Ac from **^{226}Ra target** material and ^{226}Ra progeny can also be achieved using DGA or DOODA resin. Separation of ^{225}Ac from **^{232}Th proton spallation targets** also begins with removal of the bulk Th target, followed by purification of ^{225}Ac from ^{225}Ra and other spallation byproducts. Separation of ^{225}Ac from ^{140}La and other radio-lanthanide byproducts may also be necessary, and recovery of the ^{225}Ra is also important as a source of ^{225}Ac free of the long-lived ^{227}Ac spallation byproduct.

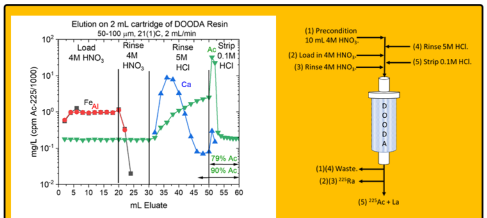


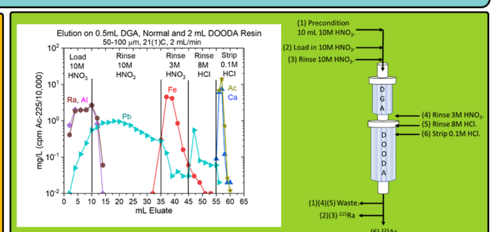
Separation of Ac from Th/Ra/decay series elements: DGA and DOODA resin both retain Ac strongly from HNO_3 and HCl. The Ac retention on DGA resin peaks at 2-3M HNO_3 and then decreases at higher HNO_3 concentrations. The retention of Ac on DOODA resin continues to increase even from 6-10M HNO_3. Ra has essentially no retention on DGA resin from HNO_3 or HCl, but is slightly retained (k' 4-5) in 1-3M HNO_3 on DOODA resin. Po(IV) and Bi(III) will remain on the DGA and DOODA resin during standard elution conditions, while Pb will be rinsed away using an HCl rinse prior to recovery of Ac using dilute HCl. Additional Th decontamination can be achieved on DGA resin by recovering Ac in 2M HCl (Th remains on DGA). From DOODA resin, Ac can also be recovered using dilute HNO_3.

Separtaion of Ac from La/reagent impurities: On DGA resin, La uptake continues to increase at high HNO_3 and HCl concentrations, while Ac retention decreases from 6-10M HNO_3. This behavior can be utilized to separate Ac from La. On DOODA resin, La and Ac uptake both continue to increase at high HNO_3 and HCl concentrations. The differences in La and Ac behavior between DGA and DOODA enable unique separation opportunities when DGA and DOODA resin are used in tandem. Fe(III) impurities can impact the labeling of the final Ac-225 product. Fe(III) can easily be removed from Ac on DGA resin by rinsing with any HNO_3 concentration and on DOODA resin by rinsing with 3-4M HNO_3. Many other common matrix impurities (Al, Na, K, Mg) are not retained by DGA or DOODA Resin. Ca can be removed with the appropriate HNO_3 or HCl rinse.

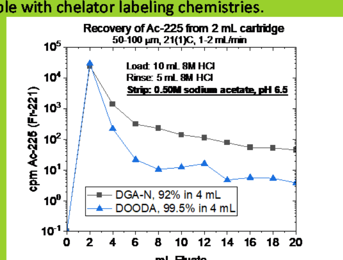


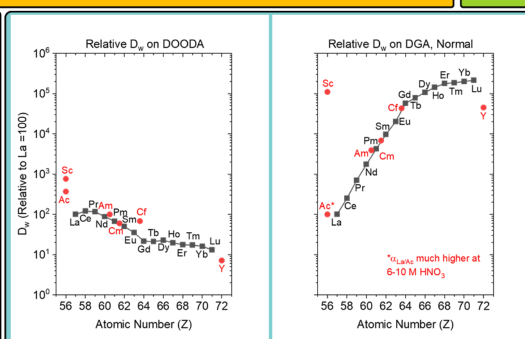






Recovery of Ac-225: Once purified, Ac-225 can be recovered from the DGA and DOODA resins in a small volume of buffer solution that is compatible with chelator labeling chemistries.





Future Work: There are many differences between the DGA and DOODA resins. In particular, the selectivity for adjacent rare earth elements (REE) and trivalent actinides is very different for the DGA and DOODA resins. DGA resin extracts heavy REE more strongly, while DOODA extractants the light REE more strongly. This selectivity difference has been utilized for the separation of Ac-225 from rare earths and other impurities using a combination of the DGA and DOODA resins. Future work will contrast the selectivity of DGA and DOODA for other separations, including the preparation of Sc-44, In-111, Y-90, Zr-89 and Ga-68.

eichrom TECHNOLOGIES
A QCI COMPANY
npo eichrom
1955 University Lane • Lisle, Illinois 60532 USA

References:

1) E. P. Horwitz, D. R. McAlister, A. H. Bond, R. E Barrans, Jr., "Novel Extraction Chromatographic Resins Based on Tetraalkyldiglycolamides: Characterization and Potential Applications," Solv. Extr. Ion Exch., 23, 319-344 (2005).
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3) M.A. Eddy, E. Rush, D. R. McAlister, "Novel Extraction Chromatographic Resins Based on the DOODA extractant: Characterization and Potential Applications," Solv. Extr. Ion Exch., submitted (2023).