



Separation of molybdenum and technetium

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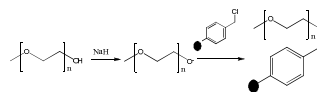


Introduction

- After the production of ^{99m}Tc via the $^{100}\text{Mo}(p,2n)^{99m}\text{Tc}$ reaction, there is a requirement to separate ^{99m}Tc from bulk ^{100}Mo . There are two challenges with this compared to commonly used generator systems; (i) the large amount of ^{100}Mo with respect to the ^{99m}Tc and (ii) the necessity of separating the expensive, highly enriched ^{100}Mo in a pure state for quantitative recycling.
- Although a number of separation methods (Liquid – liquid extraction, thermochromatographic separation etc.) have been demonstrated, the possibility of using a cartridge based system is particularly attractive for routine use in a GMP regulatory environment.
- Aqueous Biphasic Extraction Chromatography (ABEC™) was developed for selective ion (TcO_4^-) removal from nuclear waste [1]. Long polyethylene glycol chains bonded onto a water insoluble backbone yielded a Solid Phase Extraction (SPE) resin where pertechnetate was selectively adsorbed onto the resin from the strongly ionic solution while other more hydrophilic ions were not retained. Elution of the adsorbed pertechnetate was accomplished with deionized water according to Hydrophobic Interaction Chromatography (HIC) theory [2].
- This work uses ABEC™ as a starting and reference point to explore a variety of other synthetic and commercially available hydrophobic resins.

Methods

PS-DVB-PEG Resin synthesis



Grafting of PEG to polystyrene beads

PS-DVB-PEG resin synthesis [1]. The synthesis consists of grafting polyethylene glycol (PEG) to polystyrene beads.

1. Pre-swell the PS beads in diglyme (or THF) RT 15 min
2. Add PEG
3. 50°C 1 hour
4. Add sodium hydride at 35°C
5. Heat to 70°C for 17 hours.

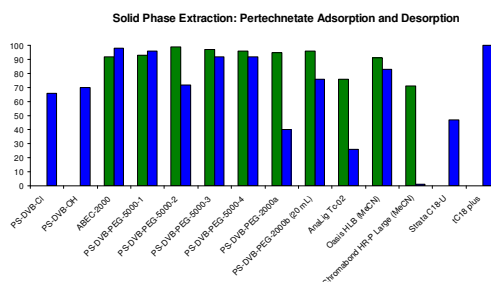
Total synthesis time was 20 hours. The yield of the synthesis was over 2.5 g PS-DVB-PEG resin per g polystyrene starting material.

SPE Separations

- Loading solution consisted of 1.5M $(\text{NH}_4)_2\text{CO}_3$ [^{99m}Tc]pertechnetate in NaCl eluted from a generator.
- Elution was performed with 10 mL D.I. H_2O unless otherwise stated.
- All cartridges were conditioned according to manufacturers instructions and subsequently with 3M $(\text{NH}_4)_2\text{CO}_3$.

Results

- "In-house" produced PEG resins showed similar adsorption characteristics as ABEC™ resin obtained from Eichrom Technologies LLC. however elution profiles of the pertechnetate were more variable. Differences could not be attributed to synthesis or resin analysis (elemental analysis).
- PS-DVB-Cl (Chloromethylated polystyrene divinylbenzene beads) and PS-DVB-OH were included in the study as references to determine the adsorption due to PEG grafted on the beads.
- Pertechnetate adsorbed strongly on the RP-SPE cartridge Chromabond® HR-P (Waters Inc.). H_2O , MeCN, DCM, DMSO were not successful to desorb the pertechnetate.



- RP-SPE Oasis® HLB (Waters Inc.). was quite successful in retaining pertechnetate but acetonitrile was needed for desorption.
- Waters tC18 plus and Phenomenex Strata® C18-U were not successful at retaining pertechnetate.

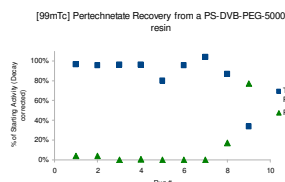
- Both adsorption and desorption of pertechnetate on AnaLig® Tc-02 (IBC Advanced Technologies Inc.) were found unsatisfactory under these non-optimized conditions. Hot H_2O was found to increase desorption, however not to completion.

Molybdenum Breakthrough Analysis

- As expected the PEG resins all exhibited very low ^{99}Mo retention and thus low breakthrough to the Tc fraction. The highest breakthrough observed was 0.08% for any PS-DVB-PEG resin.
- AnaLig® Tc-02 had a breakthrough of 0.2%, while the HR-P cartridge adsorbed 0.4% of the ^{99}Mo .
- Oasis® HLB did not adsorb [^{99}Mo]molybdate.

Degradation Analysis: PS-DVB-PEG

Polyethylene glycol is reported to be susceptible to free radical oxidation which leads to low molecular weight degradation products [3].



Method:

The same PS-DVB-PEG-5000 cartridge was used in a series of experiments. The loading solution consisted of 1.5M $(\text{NH}_4)_2\text{CO}_3$, 6% H_2O_2 , and [^{99m}Tc]pertechnetate in NaCl eluted from a generator. H_2O_2 content was experimentally measured to mimic the residual after the target plate dissolution step. The cartridge was washed with 1M Na_2CO_3 before the elution step.

Results:

- No effect was observed on adsorption or desorption of pertechnetate.
- After seven loadings the back-pressure from the cartridge increased, leading to longer loading and elution times.
 - Ammonium salts might be deposited in the resin.
- For the last data point (Run #9) the elution was never finished due to the resin resistance.

Conclusions

- HIC-SPE with PEG as stationary phase is an efficient and easily automated method to separate molybdate and pertechnetate.
- PEG degradation does not affect the performance of the resin.
- RP-SPE using Waters Oasis HLB shows promise for successful separation of molybdenum and technetium and work continues to extend this technology to readily available commercial SPE cartridges.

References

- [1] Gula M., Harvey, J., Separation, Concentration, and Immobilization of Technetium and Iodine from Alkaline Supernate Waste, Final Report March 11, 1998 U.S. Department of Energy, DE-AC21-97MC33137.
- [2] Roettger, B. F., Ladisch, M. R., Hydrophobic interaction chromatography, Biotechnology Advances, (7) 1989, 15-29.
- [3] Han, S., Kim, C., Kwon, D., Thermal/oxidative degradation and stabilization of polyethylene glycol, Polymer, (38) 1997, 317-323.

Acknowledgements

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