Evaluation of Column Separation Methods for Simplification of the Wet Chemistry Approach to Isolation of ^{211}At

S. Watanabe[a,b], K. Gagnon[a,c], D.K. Hamlin[a], M.-K. Chyan[a], E. Balkin[a] and D.S. Wilbur[a,1]

[a]Department of Radiation Oncology, University of Washington, Seattle, WA 98105
[b]Medical Radioisotope Application Group, Japan Atomic Energy Agency, Takasaki, Japan
[c]Department of Radiology, University of Alberta, Edmonton, AB, Canada

Introduction

Difficulties with reproducibility of isolation yields when distilling ^{211}At from irradiated bismuth targets led us to use a "wet chemistry" approach for that process[1]. The wet chemistry approach has provided ^{211}At isolation yields of ~ 78 % after decay and Bi attenuation corrections[2]. However, the use of diisopropyl ether (DIPE) in the separation process has made it difficult to reach our goal of automating the ^{211}At isolation. Therefore, we have investigated the use of column materials to simplify the isolation of ^{211}At and remove DIPE from the process. In this investigation we evaluated the use of a strong anion exchange resin (AG1×8), a strong cation exchange resin (AG MP-50) and a polyethylene glycol (PEG)-coated resin for separation of ^{211}At from the bismuth target material.

Material and Methods

Anion and cation resins AG1×8 and AG MP-50 were obtained from commercial sources. A PEG-coated resin was prepared by reaction of the Merrifield resin with mPEG-OH 2000 in the presence of tBuOK at 80 °C for 3 days, followed by drying under vacuum. Prior to use of the PEG resin, it was soaked in H_2O. Resins (400–800 mg) were loaded into polypropylene columns (Applied Separations, Inc.). Column elution studies were conducted with and without reductants (0.75M $FeSO_4$/1M H_2SO_4 or $Na_2S_2O_5$) to determine their effect on capture of ^{211}At. After target dissolution in HNO_3 (and in most cases subsequent removal of HNO_3 by distillation and redissolution of solid in 8M HCl), ^{211}At solution was loaded onto the column, then the column was washed with 2M HCl or H_2O to separate the Bi, and finally was eluted with strong base to remove the ^{211}At.

Initial studies were conducted with stable iodine to determine if reductants were effective in the presence of large amounts of bismuth ions. Studies with AG1×8 used ^{125}I to determine if that radiohalogen could be captured and recovered from the column when eluting with boric acid buffers at pH 5.3, 8.0 or 10, or H_2O at pH 7. Capture and recovery of ^{211}At was evaluated under the same conditions. Further studies with AG1×8 involved eluting with 4M H_2SO_4. A limited study with AG MP-50 resin used 1M HCl as eluant. Studies with PEG-coated columns used 2M HCl, 4M HCl, 8M HCl, 16 M HNO_3 and 8M HNO_3 as initial (capture) eluants. Strong base (0.2, 1 or 12.5 M NaOH; 15M NH_4OH) and 3 or 500 mM tetrabutylammonium bromide (TBAB) were evaluated for removal of ^{211}At from the columns tested.

Results and Conclusion

The efficiency for capture of ^{211}At on the AG1×8 column was high (99%) when loading with strong acid, but decreased when using 0.1–0.2M boric acid (69–91 %) buffer. Low ^{211}At capture efficiencies were obtained with AG MP-50 columns (15–29%). High ^{211}At capture efficiencies (96–100%) were obtained with PEG-coated resins when loading with 8M HCl or 8M HNO_3, irrespective of whether reductant was in the acid solution.

Four column washings (2 mL of 2M HCl each) were required to remove all Bi prior to elution of ^{211}At. No bismuth was detected in solution from the 4th washing in any of the elutions studied.

Low (< 6%) recovery of ^{211}At from the AG1×8 columns was obtained using the conditions studied. Good (60–79%) recovery of ^{211}At was obtained from PEG-coated resin using 15M NH_4OH.

Isolation of the ^{211}At from NH_4OH solution was accomplished by distillation. In an initial study ^{211}At distilled before obtaining a dry residue. However, later studies demonstrated that addition of NaOH prior to distillation kept the ^{211}At in the distilling flask.

These studies demonstrated that PEG-coated columns could be used to isolate ^{211}At from HNO_3-dissolved bismuth targets with good non-optimized (~60%) overall recovery yields. The studies are continuing with optimization of elution conditions and automation of the process.

References

1. R. D. Neirinckx, J. A. Smit: *Analytica Chimica Acta* **63**, pp. 201–204, 1973.
2. E. R. Balkin, D. K. Hamlin, K. Gagnon, M.-K. Chyan, S. Pal, S. Watanabe, D. S. Wilbur: *Appl. Sci.* **3**, pp. 636–655, 2013.

Acknowledgements. We thank the US DOE (DE-SC0010502) for funding this research.

[1]Corresponding author, E-mail: dswilbur@uw.edu