CHEMICAL RECOVERY OF THALLIUM-203 FOLLOWING PRODUCTION AND SEPARATION OF LEAD-201


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Because of the cost and limited availability of isotopically enriched thallium (>92% $^{203}$Tl), its use in the $^{203}$Tl(p, 3n)$^{201}$Pb nuclear reaction necessitates chemical recovery. An adaptive method has been developed and evaluated. After the separation of $^{201}$Pb, the $^{203}$Tl(I) is oxidized to $^{203}$Tl(III) by Cl$_2$, Br$_2$ or [Fe(CN)$_6$]$_{3^-}$, precipitated as Tl(OH)$_3$ with NaOH and subsequently converted to Tl$_2$O$_3$ by heating. Due to potential loss during recovery, the solubilities of Tl(OH)$_3$ and Tl$_2$O$_3$ in aqueous solution as a function of pH have been studied using the internal tracer $^{203}$Tl ($T = 12.2$ d), produced during cyclotron irradiation. Effective solubility product constants have been determined to be $5.4 \times 10^{-48}$ and $2.5 \times 10^{-47}$ for Tl(OH)$_3$ and Tl$_2$O$_3$, respectively.

Introduction

Thallium-201 in the form of “no carrier added” thallous chloride has been shown to be an effective radiopharmaceutical for various medical applications. As a generator product, it is obtained from its parent, $^{201}$Pb, produced by the $^{203}$Tl(p, 3n)$^{201}$Pb nuclear reaction. Excitation functions, production yields, and processing procedures have been established for the various lead and thallium radionuclides produced by the proton irradiation of natural thallium and thallium enriched in $^{205}$Tl.

The $^{203}$Tl(p, 3n)$^{201}$Pb reaction can best be facilitated using a target enriched in $^{203}$Tl. Cost consideration and limited availability of enriched materials necessitates their chemical recovery, and in this investigation a literature method has been utilized requiring the oxidation of Tl(I) to Tl(III), precipitation of Tl(OH)$_3$ with base, and conversion to Tl$_2$O$_3$. As such, the successful recovery of the $^{203}$Tl depends on the solubility of Tl(OH)$_3$ in aqueous solution and its separation from

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**The precise chemical structure of this species is debated in the literature. In this paper it will be referred to as Tl(OH)$_3$. 

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undesirable chemical impurities. The inherent presence of the radionuclide $^{202}\text{Tl}$ in the recovered material afforded an excellent opportunity to investigate the solubilities of these thallium(III) compounds using isotopic dilution. Furthermore, uncertainties exist in the literature regarding the solubilities of these compounds, especially $\text{Tl(OH)}_3$, and therefore, comparison has been made with the results obtained in this investigation.

**Experimental**

Lead-201 is obtained from the $^{203}\text{Tl(p, 3n)}^{201}\text{Pb}$ nuclear reaction by the irradiation of enriched thallium (>92% $^{203}\text{Tl}$, obtained from Oak Ridge National Laboratories) electroplated on copper. The irradiations are routinely performed in the Cyclotron Facility of the Mount Sinai Medical Center, Miami Beach, Florida. A Cyclotron Corporation CS-30 having a maximum proton energy of 26.5 MeV is utilized. Separation of the various lead isotopes from the enriched $^{203}\text{Tl}$ and the thallium radionuclides produced during the irradiation is accomplished by coprecipitation with Fe(OH)$_3$ from 4N HNO$_3$ using NH$_4$OH in a manner similar to that reported. The activities of all radioactive products were measured by $\gamma$-ray spectroscopy using a Nuclear Data 600 series Multichannel Analyzer in conjunction with a Ge(Li) detector.

**Chemical recovery of enriched thallium-203**

The enriched target material, after precipitation and separation of the lead isotopes during chemical processing, is obtained in an ammoniacal solution. An initial assay is made to determine the amount of $^{202}\text{Tl}$ present. The $^{203}\text{Tl(I)}$ is oxidized to $^{203}\text{Tl(III)}$ by stirring the solution vigorously with a slight excess of Br$_2$ in CCl$_4$ (25 ml) for 15 minutes, then concentrated HBr (3 ml) is added and the stirring continued for an additional 15 minutes. Slow addition of 10N NaOH followed by an equal volume of concentrated NH$_4$OH precipitates $\text{Tl(OH)}_3$. Ammonium hydroxide facilitates the removal of copper impurities. The solution is filtered and washed thoroughly with concentrated NH$_4$OH and H$_2$O. Approximately 60 mg of the $\text{Tl(OH)}_3$ was removed for solubility determinations. The remaining material is dissolved in 6N HCl, precipitated again with 10N NaOH and concentrated NH$_4$OH, and separated by centrifugation. If the blue copper-ammonia complex is still present, the solid is redissolved in 6N HCl and reprecipitated with 10N NaOH and concentrated NH$_4$OH, then centrifuged and washed thoroughly with concentrated NH$_4$OH. After separation, the solid material is washed sequentially with hot water, ethanol, and ethyl ether. The $\text{Tl(OH)}_3$ is dried and heated at 200 °C for 10 minutes to effect conversion to $\text{Tl}_2\text{O}_3$. 

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