SEPARATION AND ANALYSIS OF RADIOIODINE IN NUCLEAR REPROCESSING SOLUTIONS OF HIGH IONIC STRENGTH


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Ion chromatography (IC) can be used to separate radioiodine from fission products and other ionic species in complex reprocessing solution matrices. A preliminary concentration and separation of the radioiodine is performed using a column filled with a platinum-coated copper bed which converts all forms of iodine to iodide and selectively adsorbs the iodide. The separation is completed by employing an anion chromatographic system. The iodide peak aliquot is collected for subsequent radioanalysis by low energy photon spectroscopy. Results indicate quantitative separation and recovery of iodine. The entire system is automated under computer control and is able to handle small (μl) and large (500 ml) sample sizes.

Introduction

In recent years, increasing attention has been focused on radioiodine as an environmental hazard. In the environment, radioiodine quickly enters the food chain and is passed on to humans where it accumulates in the thyroid gland.1 Because of this hazard, the U.S. government requires that radioiodine levels be closely monitored by operators of nuclear facilities.

Several different methods for separating radioiodine from various solutions for analysis have been reported. Most of the reported methods require that the iodine be volatilized after which it is separated and detected in the gaseous form.2-6 Other methods involve extraction and ion exchange.7,8 Various techniques for the subsequent detection of radioiodine have also been reported.9,10 Many of these methods for separation and analysis suffer from several disadvantages, including: long analysis times, labor intensive procedures, and the use of highly toxic or corrosive reagents which are hazardous to human health and difficult to eliminate.

We have developed a technique for the ion chromatographic concentration and separation of iodine which minimizes many of the problems associated with the above techniques. This procedure is easily automated, reducing the time and labor required for analysis. The process begins with use of a column of platinum coated copper as proposed
by ARINO and KRAMER\textsuperscript{11} to convert all oxidation states of iodine to iodide and to selectively adsorb the iodide. A potential problem of this system, namely the incompatibility of the copper with acidic solutions containing high concentrations of nitrate ions, can be resolved by reducing the nitrate ions with metallic zinc prior to introduction into the separation system. The iodide in the samples is then separated from potentially interfering anions on a commercially available ion chromatographic system.

**Experimental**

*Materials*

Five different solutions simulating different types of reactor fuel reprocessing solutions were prepared. The compositions of these solutions were matched as closely as possible to the typical concentrations of cations and anions in real reprocessing solutions. No radionuclides were added to the simulated solutions. The compositions of the simulated solutions are identical to those used in our earlier work on radiostrontium.\textsuperscript{12} An $^{127}$I$^-$ spike was added to the solutions as a carrier for radioiodine, a standard procedure in current radioiodine separation procedures.

*Simulated solutions*

The following reagent grade chemicals were used in preparing the five simulated solutions: Al(NO$_3$)$_3$ · 9H$_2$O; NaCl; Ca(NO$_3$)$_2$ · 4H$_2$O; NaHCO$_3$; CdCl$_2$; NaNO$_3$; Cd(NO$_3$)$_2$ · 4H$_2$O; Na$_2$PO$_4$ · 12H$_2$O; Ce(NO$_3$)$_3$ · 6H$_2$O; Na$_2$SiO$_3$; Co(NO$_3$)$_2$ · 6H$_2$O; Na$_2$SO$_4$; Cr(NO$_3$)$_3$ · 9H$_2$O; Nb$_2$O$_5$; CsNO$_3$; Eu(NO$_3$)$_3$ · 6H$_2$O; Pd(NO$_3$)$_2$; Fe(NO$_3$)$_3$ · 9H$_2$O; Rh(NO$_3$)$_3$ · 2H$_2$O; Gd$_2$O$_3$; RuCl$_3$; H$_3$BO$_3$; HCl; Sb$_2$O$_3$; SnO; HF; Sr(NO$_3$)$_2$; HgO; UO$_2$(NO$_3$)$_2$ · 6H$_2$O; HNO$_3$; ZrO$_2$; Mg(NO$_3$)$_2$ · 6H$_2$O; ZrOCl$_2$; MnCl$_2$ · 4H$_2$O; and MoO$_3$. Water used in making all solutions was purified to a specific resistance of 18 M$\Omega$ cm using a Milli-Q water system from Millipore Corporation. Solutions were prepared as outlined in a previous paper.\textsuperscript{12}

*Platinum coated copper*

The following reagent grade chemicals were used in preparing the platinum coated copper: granular copper metal, 20–40 mesh; 1.0M H$_2$SO$_4$; N$_2$H$_6$SO$_4$; 1.0M NaOH and 50 mg H$_3$PtCl$_6$ in 2M HCl. 100 g of copper was allowed to react with a solution of 60 ml of 1.0M sulfuric acid and 8 ml of the chloroplatinic acid mixture for 5 minutes. The copper was rinsed carefully with water. A solution of 2.0 g of hydrazine sulfate dissolved in 60 ml of 1.0M NaOH was added to the copper and heated to 70 °C for 30 minutes. The platinum coated copper was cooled, washed with water several times, and stored under water until used.