NO CARRIER ADDED SEPARATION OF TELLURIUM FROM ANTIMONY

Vladimir Tolmachev and Hans Lundqvist. Unit of Biomedical Radiation Sciences, Uppsala University, Box 535, S-751 21, Uppsala, Sweden

INTRODUCTION

Tellurium (Te) has industrial applications, which grow rapidly in importance and scale. It is therefore of interest to investigate tellurium physiology and toxicity (Taylor, 1996). Radioactive isotopes of tellurium might here be of great help. We have for this purpose considered the use of two long-lived and gamma emitting isotopes, $^{121}$Te ($T_{1/2} = 16.8$ d) and $^{121m}$Te ($T_{1/2} = 154$ d). These isotopes can be produced with high specific radioactivity by proton irradiation of antimony targets.

Methods for separation of tellurium from antimony, which were developed earlier, were based on co-precipitation with isotopic carrier (Novey, 1951; Bukshpan et al., 1975) or non-isotopic selenium carrier (Glendenin, 1975). This led to long and complicated separation procedures and gave low separation yields. The disadvantages of the existing techniques inspired us to develop a new no carrier added separation method based upon ion-exchange.

EXPERIMENTAL

Radioactivity measurements were carried out with an ultra-pure germanium detector (ORTEC, Oak Ridge, TN, USA) connected to a 8192 channel PC-based multichannel analyzer (The Nucleus inc., Oak Ridge, TN, USA). The detector was calibrated for energy and efficiency with a standard $^{152}$Eu source. Dead-time losses were always below 10% during measurements. Gamma lines from $^{121}$Te (573 keV) and $^{120}$Sb (197 keV) were used to monitor the separation process.

Elemental antimony target of natural isotopic composition was irradiated at Gustaf Werner cyclotron (The Svedberg laboratory, Uppsala, Sweden) with 28 MeV protons and a beam current of 10 $\mu$A. Integrated beam current was about 70 $\mu$Ah.

During the development of the separation technique, small samples of the target material were used. They were dissolved in aqua regia during heating, evaporated close to dryness, re-dissolved in concentrated hydrochloric acid and then evaporated again. This procedure was repeated twice after which the residue was re-dissolved in 4 M HCl. The solution was loaded in a column containing an ion-exchange resin (2 mL AG 1 x 8, 100-200 mesh) pre-equilibrated with 4 M HCl. Hydrochloric acid of various concentration (1-3.5 M) was tested to elute antimony. Gamma spectra of the eluted fractions were measured to determine the relative amount of Sb and Te.

Bulk target was dissolved and treated as above. The target solution was loaded in a column containing 15 mL of the ion-exchange resin. The column was then washed with 2 M HCl and the radioactivity of $^{120}$Sb in collected fraction was measured. When no antimony associated radioactivity could be detected, the column was washed additionally with three column volumes of 2 M HCl. Tellurium was eluted with 8 column volumes of 1 M HCl.

For concentration (and additional purification), the tellurium containing eluate was mixed with equal volume of 8 M hydrochloric acid and passed through a column containing 2 mL of the ion-exchange resin. After washing with 20 mL 2 M HCl, tellurium was removed from the column with 11 mL of 1 M HCl.
RESULTS AND DISCUSSION

According to the literature data (Samuelson, 1963), Dowex 1 ion-exchange resin absorbs tellurium strongly for all concentrations of HCl but Sb(V) is not absorbed when the molarity of HCl is below 2 M. Initially, we intended to trap Te on an ion-exchange resin and then to elute it by varying the composition of the eluent. To our surprise, Te was eluted from the column at low HCl concentration. To optimize the separation conditions for Sb and Te, the hydrochloric acid concentration was varied between 1 to 4 M.

Both antimony and tellurium were strongly absorbed at 4 M HCl and no breakthrough was observed neither for Te, nor for Sb. Decreasing the HCl molarity to 3.5 M caused some elution of antimony and the elution rate increased with decreasing hydrochloric acid concentration. At concentration 1.5 M, HCl tellurium started to be eluted. At 1 M HCl the elution rate of tellurium was large and did not change by further decrease of acid molarity. In all separations about 20 % of antimony associated radioactivity remained on the column after tellurium elution, firmly bound. Since this is an expected behavior of Sb(III), we assumed that not all antimony was oxidized. However, additional treatment with hot aqua regia did not decrease this part of bound antimony. The column bound antimony did not disturb the separation and did not contaminate eluted tellurium.

In the ideal use of ion-exchange separation, the target material should pass through the ion-exchange resin but the wanted nuclide should be trapped. In principle, such condition can be realized for the antimony and tellurium separation at hydrochloric acid concentration below 3 M. However, in our hands, the antimony can be dissolved only in hot acid in these conditions. After cooling to a room temperature, target material started to precipitate with co-precipitation of tellurium.

Figure 1. Elution profiles of radioactive antimony and tellurium from Dowex1 ion-exchange resin.
For this reason, we used solution of the target material in 4 M HCl, which provided strong absorption of both antimony and tellurium to the ion-exchanger, with subsequent elution at lower acid concentrations. The disadvantage of such a procedure is that a large volume of ion-exchange resin is necessary in order to provide sufficient absorption capacity of the column. A typical elution profile is given in figure 1.

Sb(V) was completely eluted by washing with ten column volumes of 2 M HCl. After that, the column was additionally washed with three column volumes of the same acid in order to ensure an absence of target material in the eluent. Tellurium was eluted in 5-8 column volumes of 1M hydrochloric acid, which gave a separation yield of 95 %.

Repeating the separation procedure with a smaller column gave an additional purification step as well as a concentration of the final solution reducing the volume to about 10 mL. An overall separation efficiency was 90 % and no trace of $^{120}$Sb could be measured in the elute, which indicates a successful separation between tellurium and antimony.

CONCLUSION

A new method for the separation of tellurium isotopes from irradiated antimony targets is presented. The method does not require the addition of a carrier and is easier to perform than co-precipitation techniques that were applied earlier. After double purification, tellurium isotopes can be extracted with an yield of more that 90 %.

REFERENCES

• Samuelson O. Ion exchange separations in analytical chemistry, p. 311, Wiley & Sons, New York (1963)