A SIMPLE METHOD FOR CARRIER-FREE SEPARATION OF $^{77}$Br FROM METALLIC SELENIUM

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A simple method was developed for separation of bromine from metallic selenium. This method enables easy preparation of $^{77}$Br by proton bombardment of metallic selenium (enriched $^{77}$Se or $^{78}$Se). The method consists of heating the metallic selenium with H$_2$O to 300 °C in a small autoclave for about three hours. The $^{77}$Br is distilled from the obtained solution after the addition of H$_2$SO$_4$ + K$_2$Cr$_2$O$_7$. The step also removes the arsenic produced together with the selenium.

Introduction

The usefulness of $^{77}$Br for nuclear medicine is now well-recognized$^{(1)}$ and $^{77}$Br is produced routinely by the reactions $^{75}$As ($\alpha$, 2n) $^{77}$Br$^{(2,3)}$ and Br + p or d → $^{77}$Kr → $^{75}$Br$^{4,5}$. The production through $^{77}$Kr is limited since it requires protons or deuterons of energy higher than those available with most of the compact cyclotrons. JANSSEN et al.$^{6}$ have measured the yield from the $^{77}$Se (p, n) $^{77}$Br and $^{78}$Se (p, 2n) $^{77}$Br reactions and reconfirmed that these reactions lead to higher yields than those of the $^{75}$As ($\alpha$, 2n) reaction. Since the use of enriched $^{77}$Se or $^{78}$Se is required, in order to decrease the contamination from $^{76}$Br and $^{82}$Br, it is very important to use a separation process which will cause a loss of selenium as low as possible. NORTON et al.$^{7}$ developed a method to separate about 90% of the $^{77}$Br and 97% of the enriched selenium. This procedure involves the dissolution of the whole target and subsequent precipitation of AgCl + AgBr. MADHUSUDHAN et al.$^{8}$ improved and separated about 98% of the $^{77}$Br with quantitative recovery of the isotopically enriched $^{78}$Se. In a study of the determination of trace elements in metallic selenium, VOGEL and ETTEN$^{9}$ found that metallic and non-metallic contaminants of Se, including Br, can be separated quantitatively by reaction of Se with water, aqueous ammonium hydroxide or acids at 300 °C in sealed quartz ampoules. The possible explanation for this separation is that at this temperature the metallic Se is melted and the SeBr$_2$ is decomposed by reduction with water$^{10}$ to give inorganic bromides which are found in the aqueous phase.

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This work was done to study if this method can be used to separate the $^{77}\text{Br}$ produced by proton bombardment of Se, and to determine the purity of the separated $^{77}\text{Br}$.

**Experimental**

$^{77}\text{Br}$ was prepared by bombardment with 20 MeV protons (1 mA for 30 minutes) of 250 mg pressed pills (12 mm diameter) of natural selenium. Due to the high activity it was impossible to seal it in quartz ampoules and the irradiated Se pill was introduced into a 30 ml stainless-steel autoclave. Twenty ml of distilled water was added to the autoclave which was later heated to 300 °C for three hours. After fast cooling in an ice-water bath the solution was filtered through a 0.22 μ millipore filter.

**Measurement of Se contamination and reprocession**

$^{75}\text{Se}$ was used to measure the contamination of the solution with the parent selenium and the yield of the reprocessed selenium. Metallic $^{75}\text{Se}$ was prepared by irradiation of the same metallic selenium as that used for $^{77}\text{Br}$ production, in a Triga reactor at a flux of $6 \times 10^{12}$ n.cm$^{-2}$s$^{-1}$ for 60 minutes. 50–100 mg of this metallic $^{75}\text{Se}$ was added to the autoclave together with the Se–$^{77}\text{Br}$ pill.

**Results**

The results of eight experiments show that in this treatment more than 85% of the $^{77}\text{Br}$ (in most cases more than 90%) was found in the aqueous phase while more than 98% of the $^{75}\text{Se}$ was found in the precipitate, in the form of a pellet of metallic Se. The temperature and the time of heating is important due to the fact too low temperatures and short heating time reduce the yield of $^{77}\text{Br}$ while too high temperatures increase the loss of Se.

The loss of Se occurs by two mechanisms. (1) Conversion to the red allotropic form which forms a colloidal solution. Filtering of the solution through the 0.22 μ millipore filter removes part of this red selenium, but the filtered solution still has a slightly reddish color, indicating the presence of colloidal selenium. (2) Reaction with water to form selenides and H$_2$Se as can be proven by the odor of the gas released when the autoclave is opened. This process increased considerably with the temperature.

Besides the contamination of the aqueous solution by selenium as a chemical contamination, it was found that the elution process also leads to the elution of radioactive arsenic produced from the selenium by the $(p, \alpha)$ and $(p, 2p)$ reactions. $^{76}\text{As}$ and $^{77}\text{As}$ were not determined due to the same γ lines as those of the Br isotopes.