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Cross Sections and Excitation Functions for the Production of Long-lived Radionuclides in Nuclear Reactions of Lead and Bismuth with Protons

D. Schumann[1,*] and J.-Ch. David[2]

[1] *Paul Scherrer Institute, 5232 Villigen PSI, Switzerland*
[2] *CEA, Centre de Saclay, IRFU/Service de Physique Nucléaire, F-91191 Gif-sur-Yvette, France*

Radiochemical separation procedures were developed allowing isolating the fractions of several long-lived radionuclides from lead and bismuth targets irradiated with protons in an energy interval from 0.1 GeV to 2.6 GeV. The cross sections of the isotopes ^{10}Be, ^{26}Al, ^{129}I, ^{108m}Ag, ^{53}Mn, ^{60}Fe and ^{36}Cl were determined and their excitation functions were calculated. The results are compared with theoretical predictions obtained by combination of INCL4 for the intra-nuclear cascade (first stage of the spallation reaction) and ABLA for the deexcitation phase (second stage).

I. INTRODUCTION

Lead and lead-based alloys are among others - considered as target materials for high power spallation sources, either as a part of the driver device in accelerator driven systems (ADS) or as a scientific facility for neutron applications. At the Paul Scherrer Institute (PSI), solid lead targets have been used very successfully already for more than 10 years in the Swiss Neutron Source SINQ on a Megawatt level. In 2006, also at PSI, a demonstration experiment had been performed showing the qualification of the liquid metal alloy Lead-Bismuth Eutectic (LBE) as suitable target material (MEGAwatt PIlot Experiment - MEGAPIE) [1]. A very recently launched project at SCK-CEN Mol (Belgium), aimed to demonstrate the feasibility of ADS in industrial scale (MYRRHA), uses also LBE as target material [2].

Besides the knowledge on target and structure material behavior under extreme conditions, corrosion due to the liquid aggregate state of the metal, embrittlement, radiation damage and many others - also the radionuclide inventory, induced during the proton irradiation by a broad variety of nuclear reactions, is of vital importance for both a safe operation of the facility and a final or intermediate disposal of the waste. Theoretical predictions based on several nuclear reaction models help to estimate the amount of produced residues, but need benchmarks for checking the reliability. Moreover, the quality of calculations depends on the accuracy of the nuclear data to be used. Especially the data on cross sections for the production of long-lived isotopes need further improvement.

We studied the proton-induced production of long-lived radionuclides like ^{10}Be, ^{26}Al, ^{36}Cl, ^{129}I and others from lead and bismuth using thin targets and proton energies up to 2.6 GeV and derived the excitation functions for these isotopes. Most of those radionuclides, having half-lives up to million of years, cannot be measured by conventional radiation measurement technique like γ-spectroscopy, but have to be determined by accelerator mass spectrometry (AMS) after a chemical isolation of the elemental fractions from the matrix elements. Highly-sophisticated separation procedures, applied step-by-step, had to be developed to obtain samples suitable for AMS. We will present in this paper a summary of these cross section measurements and discuss the results.

II. EXPERIMENTAL

A. Irradiations

Proton irradiations of Pb and Bi targets (99.999% purity) with a thickness of 0.25 mm and a typical weight of 200 mg to 600 mg were carried out at the SATURNE II synchrotron of the Laboratoire National Saturne at Saclay (LNS) and at the cyclotron of The Svedberg Laboratory at Uppsala (TSL) during the years 1993–1997. Proton fluences between 10^{14} and 10^{16} per cm^2 were applied. A special stacked-foil technique was used for the simultaneous irradiation of a multitude of targets at different energy points. Details of the irradiation technique are described in [3].

* Corresponding author: dorothea.schumann@psi.ch

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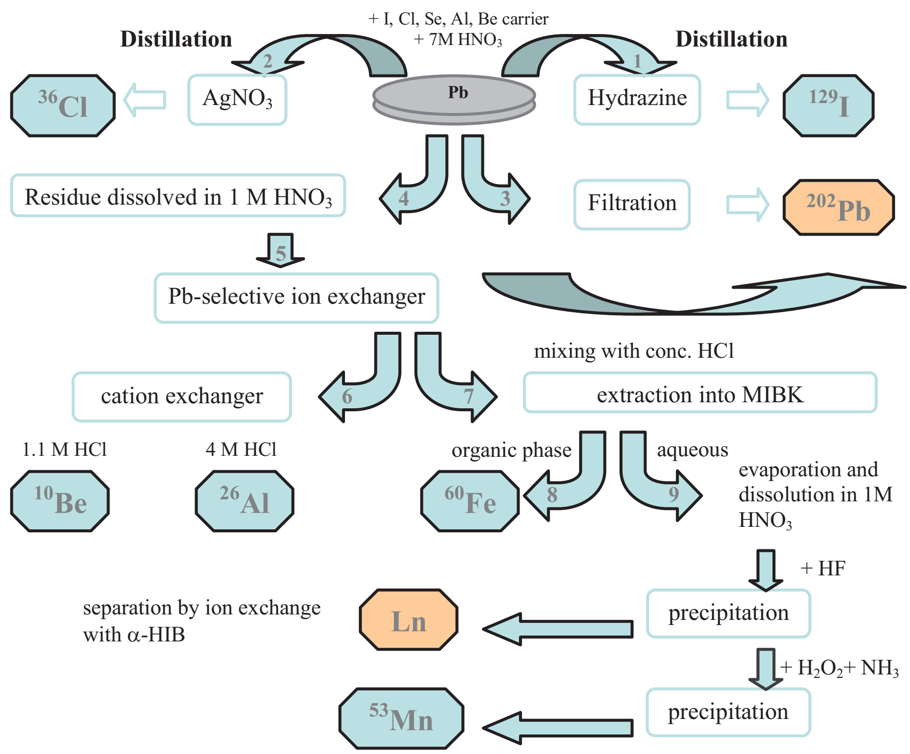

FIG. 1. Flow scheme for the chemical separation of long-lived radionuclides from irradiated lead samples.

B. Chemical Separations

Since only one sample per energy was available, subsequent separation procedures had to be developed, in order not to loose material for further investigations. In Fig. 1, a flow scheme for the separation from the lead targets is shown.

The Pb target material (about 200 mg) was dissolved in half concentrated HNO_3. Cl and I can be separated from the target material by distillation in a N_2 stream. I is distilled into hydrazine solution, Cl into $AgNO_3$ solution. The hydrazine solution was acidified with HNO_3, AgI was precipitated, filtrated, washed with bi-distilled water and dried at 80°C. The precipitate of AgCl was dissolved in NH_3, precipitated again and then treated as the I samples. The macro-amount of lead can be separated by a so-called "concentration precipitation" step, which means that a compound with a relatively high solubility - in our case $Pb(NO_3)_2$ - can be precipitated if the solution product is exceeded by partial evaporation of the solvent. 99% of the lead could be removed by filtration using this principle. The remaining solution in the flask was evaporated to dryness and the residue was dissolved in 10 ml 1 M HNO_3. Traces of Pb were removed by absorption onto a Pb-specific ion-exchange column (Eichrom) [4]. The eluate was divided into two equal parts. One part was fed onto a cation-exchange column (i.d. 0.5 cm, length 3 cm, DOWEX 50x8, 150 - 200 mesh, H^+-form). To remove boron, the column was washed with 20 ml 0.1 M HNO_3. Be was eluted with 15 ml of 1.1 M HCl, then Al with 4 ml 4 M HCl. The hydroxides were precipitated with NH_3, washed with bi-distilled water, and glowed at 800°C. The oxides were mixed with Cu powder and pressed into special target holders. The second part was used for the determination of ^{60}Fe and ^{53}Mn. These chemical procedures are described in detail in [5].

For the processing of the bismuth samples, a similar separation procedure as for lead was applied. Unfortunately, the concentration precipitation - in this case $BiONO_3$ - did not work efficiently enough for removing the macroamount of target material. Therefore, this step had been slightly modified. The in the flask remaining nitric solution was evaporated to dryness and the residue was dissolved in diluted HCl, which immediately precipitates the macroamount of bismuth in form of BiOCl, while dissolving all the other wanted elements. The precipitate was filtrated and traces of Bi were removed by loading the solution onto a cation exchange column (DOWEX50) and following elution of Bi with 0.4 M HCl. The description of the entire treatment can be found in [6].