**Volatile Elements Production Rates in a 1.4 GeV Proton-Irradiated
Molten Lead-Bismuth Target**

L. Zanini^a, M. Andersson^a, P. Everaerts^b, M. Fallot^c, H. Fränberg^{a,b}, F. Gröschel^a, C. Jost^d,
T. Kirchner^c, Y. Kojima^e, U. Köster^b, J. Lebenhaft^a, E. Manfrin^a, E. Pitcher^f, H. Ravn^b,
Y. Tall^c, W. Wagner^a, M. Wohlmuther^a

^a Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland

^b CERN, CH-1211 Geneva, Switzerland

^c SUBATECH, 44307 Nantes-cedex 3, France

^d Universität Mainz, Fritz-Straßmann-Weg 2, D-55128 Mainz, Germany

^e Hiroshima University, 1-4-1-Higashi-Hiroshima 7398527, Japan

^f Los Alamos National Laboratory, Los Alamos NM 87545, USA

Abstract

Production rates of volatile elements following spallation reaction of 1.4 GeV protons on a liquid Pb/Bi target have been measured. The experiment was performed at the ISOLDE facility at CERN. These data are of interest for the developments of targets for accelerator driven systems such as MEGAPIE. Additional data have been taken on a liquid Pb target. Calculations were performed using the FLUKA and MCNPX Monte Carlo codes coupled with the evolution codes ORIHET3 and FISPACT using different options for the intra-nuclear cascades and evaporation models. Preliminary results from the data analysis show good comparison with calculations for Hg and for noble gases. For other elements such as I it is apparent that only a fraction of the produced isotopes is released. The agreement with the experimental data varies depending on the model combination used. The best results are obtained using MCNPX with the INCL4/ABLA models and with FLUKA. Discrepancies are found for some isotopes produced by fission using the MCNPX with the Bertini intranuclear cascade model coupled with the Dresner evaporation model.

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INTRODUCTION

In the development of key experiments in the frame of the research on Accelerator Driven System (ADS) for the nuclear waste transmutation¹, many issues arise which require dedicated experiments. One example is the development of an ADS target, where the isotope production following the interaction of an intense proton beam with a liquid target is of fundamental importance for safety reasons. In the European roadmap for developing accelerator driven systems for nuclear waste incineration, the key experiment for the target development is MEGAPIE².

The aim of the MEGAPIE (MEGAWatt Pilot Experiment) project is to demonstrate the feasibility of a liquid lead bismuth eutectic (LBE) target for spallation facilities at a beam power level of about 1 MW. During the design phase of such an innovative system, many safety aspects must be considered. One of them concerns the production of volatile elements during operation. This is important for several reasons: *i*) some stable gases, and in particular ⁴He and H, are expected to be produced in relatively large quantity (in the case of MEGAPIE, about 1 liter NTP per month) and a system must be designed to handle safely the gases and avoid excessive pressure buildups. Moreover, it is important to know the production of these light elements to estimate possible damage to structural materials. *ii*) the production of radioactive elements is of concern for safety reason. The long-lived elements are of major concern, but short-lived elements are also of interest in case of an accident.

In the last years a great research effort was devoted in basic nuclear studies of interest for ADS applications. This has resulted in a renovated interest in the study of isotope production following spallation reactions in heavy materials^{3,4}. Experiments performed in inverse kinematics have allowed the investigation over large mass regions of production cross sections in thin targets⁵. These experiments, in combination with the further development of Monte Carlo transport codes, have led to a deeper understanding of the spallation process and to the development of new theoretical models⁶.

In the case of an ADS target, where production of isotopes originates in a large volume of LBE, it is important to consider not only the production of volatile elements, but also their release rate out of the LBE volume. In the case of MEGAPIE, a cover gas system has been designed to properly handle the gas produced⁷. A verification of the production rates estimated by the codes used during the design of the cover gas system is therefore important.

We chose to perform a dedicated experiment to study the production rates of stable and radioactive volatile elements in a LBE target irradiated by a proton beam of the energy of the order of the energy of the SINQ synchrotron (590 MeV).

Results presented in the present paper should be considered as preliminary.

EXPERIMENTAL SETUP

The experiment was performed at the ISOLDE facility⁸. The spallation target consisted of a cylindrical tantalum container filled with liquid LBE. Protons pulses of 1.4 GeV and variable intensity (up to 10^{13} protons/pulse with a rate of one pulse every 16.8 s) impinged on the target. Following spallation reactions, the produced volatile elements exiting the liquid metal were ionized by means of a plasma ion source, then accelerated to 60 keV and sent to the magnetic mass separators and to the beam lines where the measuring stations were placed. An additional measurement was performed with a liquid Pb target.

Yields were measured using three different techniques of common use at ISOLDE. Online yields of stable isotopes and of some radioactive ones were measured by a Faraday cup inserted in the beam line. A special data acquisition system was developed to trigger the

current measurement by a picoampèremeter with the arrival of the proton beam on target, thus allowing the measurement of the gas release curves, characteristic of each element. For short-lived β emitting isotopes, beams were directed to a dedicated tape station and yields were measured with a plastic scintillator detector.

A third measurement method was used for longer lived ($T_{1/2} \geq 5$ min) γ emitting radioisotopes; ion beams were implanted on thin Al foils, then after irradiation an offline γ detection was performed using a calibrated HPGe detector.

In order to obtain the absolute production rates from the measured yields, the efficiency of the ion source had to be measured. For this purpose, known amounts of different gas mixtures (consisting of Ar/Xe, He/Ne/Ar/Kr/Xe, and ^3He /Ar/Xe mixtures) were leaked into the ion source, thus having the possibility to measure the efficiencies at any time during the experiment.

For the LBE target, the measurements were performed with the target at temperatures of 400 °C and 600 °C. The Pb target was at a temperature of 520 °C. These temperatures are in the range of the LBE temperature in MEGAPIE during operation, which varies from 300 °C to 400 °C depending on the position inside the target. Temperature differences within these ranges are not expected to affect the releases of the noble gases and of the Hg isotopes. On the other hand, differences are expected for some isotopes such as I, Cd and Po. Having performed the experiments at higher temperatures than in MEGAPIE will allow to conclude, in case the release of a specific isotope is not observed at 600 °C, that no release should be observed in MEGAPIE for the same isotope at 300-400 °C, even for longer irradiation times.

A selection of the data is presented in this paper, with emphasis on the γ -spectroscopy data.

FIRST RESULTS

Online measurements

The time-dependent releases of volatile elements were measured with the online measuring techniques of the tape station and the Faraday cup. Release curves of volatile elements have specific shapes typical for each element; in most of the cases the decay part can be fitted with the sum of two exponentials⁹.

The online measurement with the tape station allows correction for partial decay of produced isotopes inside the target, before the release. In fact, since the release is dependent on the chemical properties of a given element, it is possible for instance to fit the release functions of ^6He (measured with the tape station) and ^4He (measured with the Faraday cup) and correct for the partial decay of the ^6He .

During the first measurement, with the LBE target, it was found that the short term component exhibited discontinuities probably related to splashing effects in the target which reduced for a few tens of ms the ionization efficiency of the ion source. While this affects only slightly the absolute release, which is dominated by the long component, it makes it more difficult to fit the release curve. No such effect was observed during the second measurement, with the Pb target, where the proton beam intensity was reduced to 1.5×10^{12} protons/pulse.

In Fig. 1 the ^4He current measured by a Faraday cup for 6 s after the arrival of the proton beam on the Pb target is shown.

The ionization and transmission efficiency from the ion source to the Faraday cup was measured to be 0.05 % for ^3He . Assuming the same transmission efficiency for ^4He , the production rate for ^4He is 0.77 atoms/p, with a systematic uncertainty of about 20 %. This value is in good agreement with calculations with MCNPX with the Bertini/Dresner models, giving 0.84 atoms/p.

Offline measurements

Collection measurements were performed for a number of isotopes. We investigated the release of Ne, Ar, Kr, Xe, Br, Cd, Te, I, Hg, Po, and At radioisotopes. During the first measurement run more attention was concentrated on those isotopes which are critical for the operation of an ADS target such as MEGAPIE.

For a given isotope, the measured yield has two components, one from direct production from the target and one from the decay of parents. Isotopes were collected in an order chosen so that the first ones to be measured were the first reaching equilibrium, having parents with shorter half-lives. In this way most of the measured isotopes were in equilibrium with their parents, with only a few exceptions.

In Figure 2 the measured cumulative production rates for radioactive Hg isotopes are presented. Longer-lived Hg isotopes are expected to be completely released at the temperature of 600 °C. The ionization efficiency was not measured for Hg, as it was only measured for noble gases. In this case only indicative results can be extracted: based on previous results from Ref. 10, we considered an efficiency of a factor 1.5 higher than the measured Xe efficiency of 3.7(11) %.

The measured values are in line with expected cumulative production rates calculated using the Monte Carlo transport codes FLUKA¹¹ and MCNPX¹². The two codes were coupled with the evolution codes ORIHET3¹³ and SP-FISPACT¹⁴, respectively. In the case of MCNPX, results are shown here with two different model combinations for the intranuclear cascade and evaporation models. The circles represent results from using the Bertini intranuclear cascade model with the Dresner evaporation code. The diamonds are obtained using the recently implemented INCL4/ABLA⁶ model combination. The trend observed in the data as a function of the atomic mass is well reproduced by the three calculations. One should note that for ^{193}Hg , ^{195}Hg and ^{197}Hg , there are isomeric states of 11.1 h, 40 h and 23.8 h half-lives, respectively. For these three isotopes, equilibrium was not achieved between formation and decay of the respective isomeric states, a process which is difficult to properly calculate with existing Monte Carlo codes. Overall these results confirm the expected production rates of Hg isotopes in a thick LBE target.

Results for Xe isotopes, also measured with the LBE target at $T=600$ °C, are shown in Fig. 3. In this case there is a clear disagreement between the values calculated with MCNPX with Bertini/Dresner, and the results from the other two calculations. The data, with an ionization efficiency of 3.7 % for Xe isotopes seem to favor the other two calculation results, thus confirming recent experimental findings⁵.

Similar results are obtained for the iodine isotopes. However, I is not completely released and observed production rates at 600 °C are a factor 10 lower than the calculated FLUKA and MCNPX (INCL4/ABLA) values.

While production of Hg isotopes from Pb/Bi target is due to direct spallation, the Xe and I isotopes are the results from a later stage of the spallation process, the fission of highly excited spallation fragments, or as a two-step process due to neutron induced fission from high energy spallation neutrons. Thus the evaporation models, the Dresner and ABLA, are probably most responsible for the differences observed in the calculations.

Among the other isotopes measured, it is of particular interest to discuss the Po and At. Production rates of $^{207,208,209,210}\text{At}$ of the order of 10^7 atoms/ μC (assuming the same ionization efficiency as for Hg) were detected, with values an order of magnitude lower for ^{206}At . Such production rates are not of concern for an ADS. On the other hand, it is the first observation of At beam from a Pb/Bi target and this constitutes an interesting result with possible applications. Production of At comes from several possible reactions of Bi, but the most likely, given the high proton energy, is $^{209}\text{Bi}(p, \pi^- xn)^{210-x}\text{At}$. The At decay is responsible for the observed small quantities of Po isotopes, which contrary to At is expected to be produced in large amounts. However, as found in Ref. 15, little or no Po should be released at 600 °C.

Of the other isotopes measured in the first measurement, no release of Br was observed, while very little amounts of Cd isotopes were detected. For the Kr isotopes, some problems during the measurement rendered the analysis questionable and such measurement was repeated with the Pb target.

CONCLUSIONS

The first results from the measurements of production rates of volatile elements from irradiated LBE and Pb targets indicate that the results are consistent with the expectations from Monte Carlo calculations. Overall, these preliminary results confirm the expected production rates in an ADS target such as MEGAPIE, and therefore help in positively assessing such calculations, and the system designed to handle the released volatile elements.

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FIGURE 1. Current of ^4He (in pA) measured by the Faraday cup.

FIGURE 2. Production rates for Hg isotopes. Measured points (*black squares*) are compared with calculations: *open circles*: MCNPX (Bertini/Dresner model combination); *diamonds*: MCNPX (INCL4/ABLA); *stars*: FLUKA.

FIGURE 3. Same as Fig. 2 but for Xe isotopes.