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Production of noble gas isotopes by proton-induced reactions on lead and bismuth

I. Leya[a,b,*], R. Wieler[b], J.-C. David[c], S. Leray[c], L. Donadille[c,1], J. Cugnon[d], R. Michel[e]

[a] *Physikalisches Institut, University of Bern, Bern, Switzerland*
[b] *Institute for Isotope Geology and Mineral Resources, ETH Zürich, Zürich, Switzerland*
[c] *DSM, DAPNIA/SPhN, CEA-Saclay, Gif-sur-Yvette, Cedex, France*
[d] *Université de Liège, Institut de Physique, Sart Tilman, Liège, Belgium*
[e] *Center for Radiation Protection and Radioecology, University Hannover, Hannover, Germany*

Abstract

We measured integral thin target cross-sections for the proton-induced production of He-, Ne-, Ar-, Kr-, and Xe-isotopes from lead and bismuth from the respective reaction threshold up to 2.6 GeV. The production of noble gas isotopes from lead and bismuth is of special importance for design studies of accelerator driven nuclear reactors and/or energy amplifiers. For all experiments with proton energies above 200 MeV a new mini-stack approach was used instead of the stacked-foil technique in order to minimise influences of secondary particles. The phenomenology of the determined excitation functions enables us to distinguish between the different reaction modes fragmentation, hot and cold symmetric fission, asymmetric fission, and deep spallation. For lead more than 420 cross-sections for 23 nuclear reactions have been measured. While the lead data have already been published, here we present first results for the production of noble gas isotopes from bismuth. The experimental data are compared to results from the theoretical nuclear model code INCL4/ABLA. This comparison clearly indicates that experimental data are still needed because the predictive power of nuclear model codes, though permanently improving, does still not allow to reliably predict the cross-sections needed for most applications and irradiation experiments remain indispensable.

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1. Introduction

In recent years spallation neutron sources were built in various countries and accelerator based nuclear transmutation (ADS), e.g. [1,2] and energy amplification (EA), e.g. [2,3], devices were proposed as a possibility of closing and/or improving and cleaning the nuclear fuel cycle. However, a key issue for the design and construction of such devices is the reliable modelling of the production of radioactive and stable residual nuclides by proton- and neutron-induced reactions. For proper modelling the differential particle spectra and the excitation functions of the relevant nuclear reactions have to be known. While calculating differential particle spectra using state-of-the-art Monte Carlo codes is now very reliable, the thus calculated cross-sections are accurate to within a factor of 2 at best, which is not sufficient for most applications. Therefore, experimental cross-sections are still essential for design studies of spallation neutron sources and ADS/EA systems.

Here we present thin-target cross-sections for the proton-induced production of He-, Ne-, Ar-, Kr-, and Xe-isotopes from lead and bismuth. For lead the data base is fairly complete by now and more than 420 cross-sections for 23 nuclear reactions have been measured. The data together with a detailed discussion of the various reaction

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[*] Corresponding author. Physikalisches Institut, University of Bern, Bern, Switzerland.

E-mail address: Ingo.Leya@phim.unibe.ch (I. Leya).

[1] IRSN/DPHD/SDOS/LRDE, Fontenay-aux Roses, France

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modes are given in Ref. [4]. Here we present for the first time cross-sections for the production of He-, Ne-, Ar-, Kr-, and Xe-isotopes from bismuth. However, the database so far is rather scarce but first interesting results have been obtained and will be discussed. Such preliminary data are given in figures only. Further measurements are currently underway and the final data will be published in a subsequent paper.

The data are compared to theoretical excitation functions calculated by the combination of two codes, INCL4 [5] for the intranuclear cascade and ABLA [6] for the nucleus de-excitation. We choose INCL4/ABLA because for this system good results for the production of reaction yields in thin and thick targets have been demonstrated, e.g. [5]. Note that most of the samples studied here have already been analysed for residual radionuclides [7].

2. Experimental

As targets high purity materials were used to avoid interfering reactions from impurities. The targets were of natural isotopic composition. The irradiation experiments were performed at the SATURNE synchrocyclotron of the Laboratoire National Saturne at Saclay, France ($E > 200$ MeV), The Svedberg Laboratory at Uppsala, Sweden ($70\,\mathrm{MeV} < E < 200\,\mathrm{MeV}$), and the Paul Scherrer Institute at Villigen, Switzerland ($E < 72$ MeV). For proton energies below 200 MeV the *stacked-foil technique* was used, since the influences of secondary particles on the production of the nuclides studied here can be neglected. For the irradiation experiments above 200 MeV we used the so-called *mini-stack approach* to reduce secondary particle effects [4,7]. The flux densities were determined via the reaction ^{27}Al(p, 3p3n)^{22}Na using cross-sections given by Refs. [8,9].

The noble gas isotopic concentrations were measured by static noble gas mass spectrometry. After being loaded into the all metal noble gas extraction system the samples were preheated in order to release atmospheric surface contamination. The lead and bismuth samples were degassed in a Mo crucible held at 1000 °C and 800 °C for 15 min, respectively. The gases were cleaned on Zr–Ti and Al–Ti getters. He–Ne, Ar and Kr–Xe fractions were separated using cryogenic traps and measured separately. For further information see Ref. [4]. The error bars shown in Figs. 1–3 include the uncertainties of the mass and the thickness of the target foil, the uncertainty of the noble gas concentrations (blank corrections, calibrations) and the uncertainty of the monitor cross-sections.

3. Results for lead

Some selected excitation functions for the proton-induced production of noble gas isotopes from lead are shown in Fig. 1. Results of INCL4/ABLA calculations are shown by lines. The excitation functions for the production of ^{3}He and ^{4}He are shown in panel (a). The data indicate that low energetic reactions do not distinguish between ^{3}He

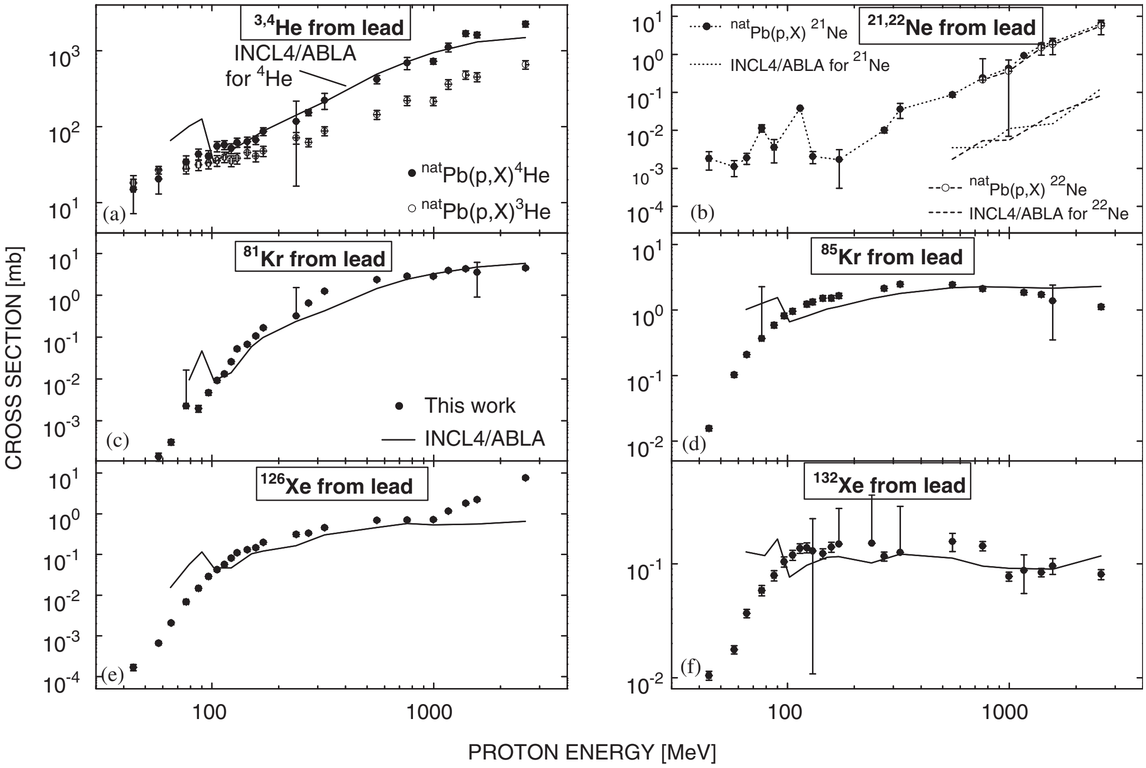

Fig. 1. Excitation functions for the proton-induced production of ^{3,4}He (panel (a)), ^{21,22}Ne (panel (b)), ^{81}Kr (panel (c)), ^{85}Kr (panel (d)), ^{126}Xe (panel (e)) and ^{132}Xe (panel (f)) from natural lead. The lines are results of INCL4/ABLA calculations. Error bars smaller than symbol sizes are suppressed.

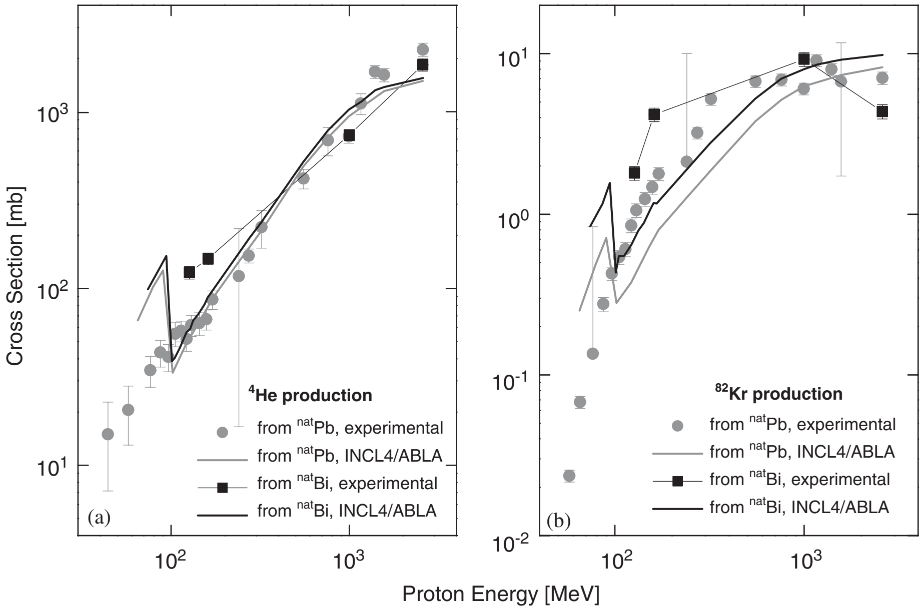

Fig. 2. Excitation functions for the proton-induced production of ^{4}He (panel (a)) and ^{82}Kr (panel (b)) from lead and bismuth. For further explanation see Fig. 1.

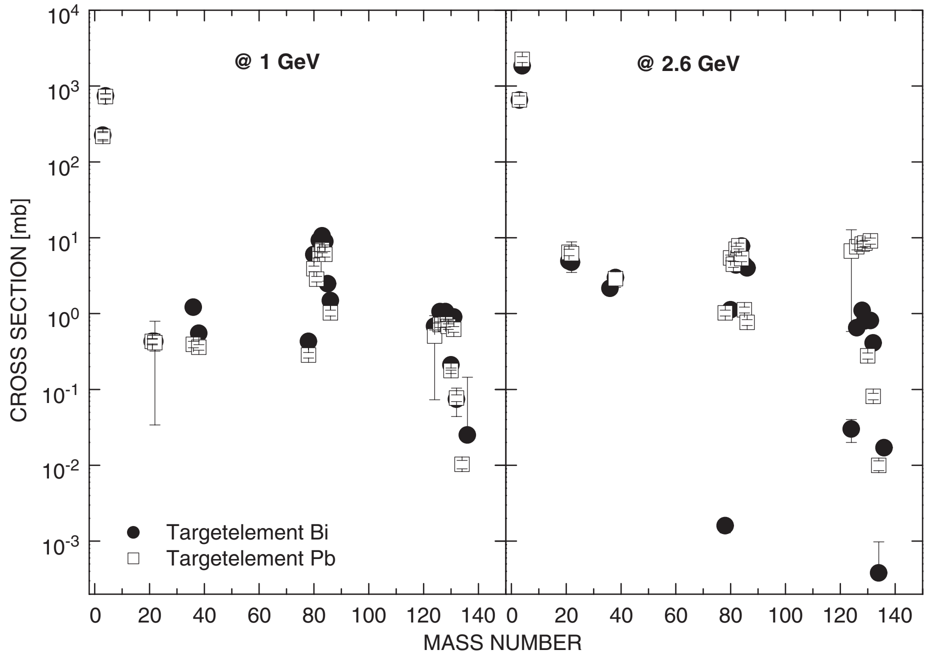

Fig. 3. Isotopic cross-sections for noble gas isotopes produced in natural lead and bismuth at 1 GeV (left panel) and 2.6 GeV (right panel).

and ^{4}He, whereas at higher energies the production of ^{4}He becomes 3–4 times larger than the production of ^{3}He. The excitation functions for the production of ^{21}Ne and ^{22}Ne are shown in Fig. 1(b). Cross-sections for ^{21}Ne could be determined for energies down to 50 MeV, i.e., at energies significantly lower than the Coulomb-barrier for the emission of a ^{21}Ne fragment, which is about 100 MeV. Since interfering reactions from impurities can be neglected, e.g. the required impurity of $>0.1\%$ is considerably higher than allowed by the nominal purity of the foils (99.99 + %), the data for ^{21}Ne and ^{38}Ar (not shown) clearly demonstrate that the effective Coulomb-barrier in a nuclear reaction might only be half the value of the nominal Coulomb-barrier. This finding, although the process not yet understood, is in agreement with earlier studies [10].

The cross-sections for Kr isotopes enable us to distinguish two different reaction mechanisms. The n-rich isotopes, e.g. ^{85}Kr (Fig. 1(d)) are dominantly (only?) produced via *hot symmetric fission*, i.e. a process occurring early in the reaction process while the excited nucleus is still hot. Therefore, hot symmetric fission would be associated to times early in the evaporation phase. The n-poor isotopes, e.g. ^{81}Kr (Fig. 1(c)) are produced via cold symmetric fission, i.e. fission after the nucleus has almost completely de-excited. (Note the change in definition compared to Ref. [4]). For nuclides with intermediate masses the excitation function can be interpreted as being a mixture of the two.

Xenon isotopes are produced via *asymmetric fission* and (for some n-rich isotopes) via deep spallation. For ^{130}Xe, ^{132}Xe and ^{134}Xe asymmetric fission is the dominant (only?) reaction mechanism in the energy range studied by us. For the other (more n-rich) Xe isotopes a steep rise in the excitation function above 600–1000 MeV is observed. We interpret this finding as due to deep spallation, which has a threshold energy of about 600 MeV.

4. Results for bismuth

Some of the (preliminary) data for the proton-induced production of noble gas isotopes from bismuth are shown in Figs. 2 and 3. The cross-sections for the production of ^{4}He and ^{82}Kr from lead (grey symbols) and bismuth (black symbols) are compared in Fig. 2. Since both elements are very similar with respect to proton and neutron numbers, one would expect the excitation functions for the same product nuclides to be very similar. Surprisingly, the data obtained so far clearly indicate some substantial differences. For example, while the cross-sections for the production of ^{4}He from bismuth below 200 MeV are higher than the lead data, the latter are slightly higher above 1 GeV. As a consequence, the excitation function for ^{4}He production from bismuth is less steep than for the production of ^{4}He from lead. In contrast to the experimental finding, INCL4/ABLA predicts the same shape for both excitation functions, the cross-sections for bismuth being slightly higher than for lead.

Significant differences between lead and bismuth are also observed for the production of ^{82}Kr. As discussed elsewhere [4], the excitation function for the production of ^{82}Kr from lead has a steep rise at low energies and reaches a plateau above 1 GeV. Such a shape has been interpreted as being a mixture of hot and cold symmetric fission. In contrast, production of ^{82}Kr from bismuth is characterised by increasing cross-sections at low energies, a local maximum at about 1 GeV and a high-energy decrease. Such a shape has been interpreted as representing a relatively hot symmetric fission mode, i.e. fission after a short intranuclear cascade early in the evaporation phase [4,7]. The data therefore indicate that the production of ^{82}Kr from bismuth is by hot symmetric fission and from lead by a mixture of hot and cold symmetric fission.

Fig. 3 compares the residual noble gas distributions from lead and bismuth at 1 GeV (left panel) and 2.6 GeV (right panel). At first glance, the distributions for both target elements (at each energy) look very similar. Nevertheless, significant differences can be observed. For example, the production of n-poor Kr-isotopes is higher in lead than in bismuth. In contrast, the cross-sections for n-rich Kr-isotopes are lower in lead than in bismuth. The total cross-section for Kr-production, i.e. the sum of the isotopic cross-sections, is similar for lead and bismuth.

For Xe-isotopes no such systematics are observed. For example, the production of n-poor Xe-isotopes is higher in lead than in bismuth. In contrast, the cross-sections for even Xe-isotopes are higher for bismuth than for lead. Interestingly, the total cross-section for Xe-production, i.e. the sum of the isotopic cross-sections, for lead is about 10 times higher than for bismuth. For the lighter residual nuclides no such big effects are observed. The cross-sections for ^{3,4}He-, ^{21,22}Ne-, and ^{36,38}Ar-production for lead and bismuth agree to within about 30%.

The partly large differences for the noble gas production from lead and bismuth are rather unexpected and not yet understood. If the observed effects are due to the fact that ^{208}Pb, which is the major lead isotope, is a double magic nucleus while ^{209}Bi has one *excess* proton compared to a magic number remains to be seen and further studies are needed.

5. Comparison with model calculations

The INCL4/ABLA model describes most of the experimental data reasonably well, i.e. within a factor of 2. Notable exceptions are the production of ^{21,22}Ne, ^{36,38}Ar and the Xe-isotopes above 1 GeV by deep spallation and/or multifragmentation. For the Xe-isotopes this is due to the fact that we are close to the reaction threshold, i.e. close to the limit of applicability of INCL4/ABLA. Note that the model predicts similar cross-sections for lead and bismuth whereas the experimental data indicate some substantial differences.

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