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,*]
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. 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. 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.

Key words: Noble gases, Irradiation experiments, Nuclear reactions
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* IRSN/DPHD/SDOS/LRDE, Fontenay-aux Roses, France
Email address: `Ingo.Leya@phim.unibe.ch` (I. Leya).

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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. However, a key issue for the design and construction of such devices is the reliable modelling of the production of the radioactive inventory. Here we present thin-target cross sections for the proton-induced production of He-, Ne-, Ar-, Kr-, and Xe-isotopes from lead. Some of the important reaction mechanisms are discussed and the experimental data are compared to INCL4/ABLA model calculations. The complete cross section database for proton-induced production of noble gas isotopes from lead together with detailed discussions of the various reaction modes has already been published [4]. The data are also available from the NEA DATA Bank or other members of the International DATA Center Network under EXFOR number O0839. The cross sections for the target element bismuth are currently measured and will be published in a subsequent paper. First results will be presented at the conference. Most of the samples studied by us have already been analysed for residual radionuclides [5].

2 Experimental

The irradiation experiments were performed at the SATURNE synchrocyclotron of the Laboratoire National Saturne at Saclay, France, The Svedberg Laboratory at Uppsala, Sweden and the Paul Scherrer Institute at Villigen, Switzerland. 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. For further information see [4]. The noble gas isotopic concentrations were measured by static noble gas mass spectrometry. The samples were degassed in a Mo crucible and 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. Further details about the mass spectrometric measurements, the blank corrections and the calibrations are given in [4]. The errors shown in Fig. 1 include the uncertainties of the mass and the thickness of the target foil and the uncertainty of the noble gas concentrations (blank corrections, calibrations). The monitor cross section yield an additional uncertainty of 7% that effects all data in the same way.

3 Results

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. Since Helium diffusive losses during irradiation and/or storage (if any) should only be very minor [4], the data indicate that low energetic reactions do not distinguish between ^{3}He 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. 1b. 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 of about 100 MeV. Since interfering reactions from impurities can be neglected [4], 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; see also [4]. The results for ^{81}Kr and ^{85}Kr are shown in Fig. 1c,d. In general, the cross sections for the Kr isotopes enable us to distinguish two different reaction mechanisms; *hot symmetric fission* for n-poor isotopes, e.g. ^{81}Kr, and *cold symmetric fission* for n-rich isotopes, e.g. ^{85}Kr. For intermediate masses the excitation functions can be interpreted as being a mixture of the two. The cross sections for the production of Xe isotopes are interpreted as a result of *asymmetric fission*. 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 Xe isotopes a steep rise in the excitation function above 600-1000 MeV is observed. We interprete this finding as due to deep spallation, which has a threshold energy of about 600 MeV.

The obtained cross section database enables one to distinguish various reaction mechanisms. The production of Ne- and Ar-isotopes is via deep spallation and/or multifragmentation (or very asymmetric fission). Krypton isotopes are produced either via hot or cold symmetric fission; the isotopic yields follow a simple gaussian distribution. Superimposed to the gaussian are odd-even effects, which disappear at higher excitation energies. The production of Xe-isotopes is via asymmetric fission below 600 MeV and via a combination of asymmetric fission and deep spallation above 600 MeV. The ratio of symmetric to asymmetric fission increases with increasing proton energy.

The predictability of the INCL4/ABLA model strongly depends on the type of reaction mechanism. A reasonably good description is achieved for the production of ^{4}He, for the hot symmetric fission modes (n-poor Kr-isotopes) and for the asymmetric fission modes (Xe-isotopes below 600 MeV). For the cold symmetric fission modes (n-rich Kr-isotopes) the agreement between model predictions and experimental data is worse. A special problem of the model is obvious for deep spallation and multifragmentation, simply because production of Xe above 1 GeV and of ^{21,22}Ne and ^{36,38}Ar is underestimated by up to

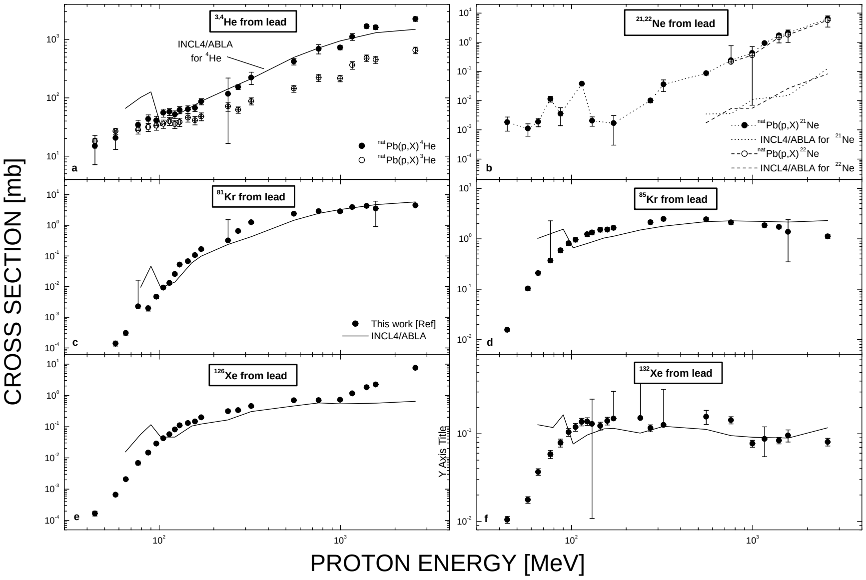

Fig. 1. Excitation functions for the proton-induced production of ^{3}He and ^{4}He (panel a), ^{21}Ne and ^{22}Ne (panel b), ^{81}Kr (panel c), ^{85}Kr (panel d), ^{126}Xe (panel e) and ^{132}Xe (panel f). The lines are results of INCL4/ABLA calculations. Error bars smaller than symbol sizes are suppressed.

two orders of magnitude. In general, the model calculations for most of the nuclides studied here are accurate to within a factor of 2 at best.

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