Isotope production rates in a thick lead target in the reaction d+Pb at 2.52 GeV

O Yordanov¹, S Batzev¹, L Kostov¹, Ch Stoyanov¹, P Zhivkov¹, J Adam², V D Kovalenko², M I Krivopustov², A A Solnyshkin², V M Tsoupko-Sitnikov², K Katovsky³ and M Majerle⁴

¹ Institute for Nuclear Research and Nuclear Energy, 72 Tzarigradsko chaussee, Blvd. BG – 1784 Sofia, Bulgaria
² Joint Institute for Nuclear Research, Joliot-Curie 6, 141980 Dubna, Moscow region, Russia
³ Czech Technical University, Zikova 4, 166 36, Prague 6, Czech Republic
⁴ Nuclear Research Institute Rez plc, Husinec – Rez 130, 250 68 Rez, Czech Republic

E-mail: orliny@inrne.bas.bg

Abstract. The “Energy Plus Transmutation”-setup consists of a thick cylindrical lead target of 456 mm length and diameter of 84 mm, which is surrounded by a thick layer containing 206,4 kg of natural uranium. This small-sized target-blanket-assembly resembles the geometry and the constituents of an industry-sized subcritical system for power production and nuclear waste transmutation. The irradiations of the target-blanket-assembly are performed using deuteron beams of variable energies, delivered by the Nuclotron accelerator at the Joint Institute for Nuclear Research in Dubna. Using precision gamma-spectroscopy, the amounts of isotopes of bismuth, lead and thallium, produced in the lead target during the irradiation with a deuteron beam of 2,52 GeV [1,2] have been determined. The collected data on the isotope production are compared with the predictions of nuclear-reaction- and particle-transport-codes.

1. The “Energy+Transmutation” experimental program
In an accelerator-driven system a high-intensity, relativistic particle beam- most common of protons, is impinged on a thick target, made of a heavy material like lead or tungsten. The target nuclei, hit by the primary beam, undergo spallation reactions, in which in average a large amount of fast neutrons is released. Depending on their energies and among the wide variety of the possible reaction channels, the released neutrons are expected in particular to induce fission, or to be captured in nuclei from the material, surrounding the target. The neutron-induced reactions on long-living radioactive nuclides are expected to produce short-living ones, or to induce the fission of fissile nuclides.

In this respect the accelerator-driven systems show a technological possibility to meet the ecological and security problems, caused by the long-term radioactivity and the presence of fissile material, accumulated in the spent fuel of today’s nuclear power plants.

The transformation of hazardous nuclear materials by means of nuclear reactions is also referred to as transmutation.
The setup used in the experiments within the “Energy + Transmutation”-program consists of a thick lead target of 456 mm length and diameter of 84 mm, which is surrounded by an array of aluminium cans, containing $^{238}$U.

The light particles, mostly neutrons and protons, which are produced in the interaction of the primary beam with the lead target, are passing through thick layers of matter including the target itself and the massive uranium blanket. The particle transport through the setup is determined by diverse reaction mechanisms as scattering-, capture- knock-out-, fission- and fragmentation reactions.

2. The irradiation with 2.52 GeV deuteron beam

During the presently discussed experiment, the setup was irradiated with a deuteron beam of 2.52 GeV, a total amount of $6.5 \times 10^{12}$ particles was impinged on the target.

In order to understand the physical behaviour of the setup a large set of probes, including nuclear track detectors, samples for neutron activation analysis and samples containing $^{238}$Pu, $^{239}$Pu, $^{129}$I, and $^{237}$Np were placed at different positions within, and on the surface of the setup.

As important, as the detailed measurement of the neutron distributions in energy and intensity within and around the setup, are the experimental data on the interaction of the primary beam with the target. In order to collect experimental data also from the target of the setup, a set of samples, made of lead, as the target itself, have been placed on the surfaces of the target sections, slightly away from the symmetry axis of the setup.

3. Positioning and $\gamma$-spectroscopic survey of the lead samples

The interaction of the primary beam with the target was monitored with a set of six lead samples, placed in the vicinity of the symmetry axis of the setup.

The samples have masses of 609 mg, 590 mg, 574 mg, 599 mg, 574 mg and 605 mg, equal diameters of 15mm, and are composed of natural lead. It is expected, that the residual nuclei will be created in fragmentation-, proton-induced-, and neutron-induced reactions.

The six samples of lead were positioned during the experiment 2 cm away from the symmetry axis of the setup, as follows: samples 1 and 5 on the entrance- and exit sites of the target, samples 2, 3, 4 were placed in the slits between the sections of the lead target, the sample “0” was positioned away from the setup, behind the aluminium beam monitor.

After the end of the irradiation the samples have been removed from the setup and transported to the experimental area of the Laboratory of Nuclear Problems of JINR for $\gamma$-spectroscopy measurements.

In order to collect data on short- and long-living isotopes, the $\gamma$-spectra have been measured sequentially, whereas the time for collecting data was initially set to 30 min, with a time between two measurements in the order of hours. The last spectra of each probe where collected within six hours, with about one week of time between the last two subsequent measurements. A similar time schedule was applied also for the $\gamma$-spectroscopy of the other activation samples.

The $\gamma$-spectra of the lead samples were collected using an Ortec HPGe-detector coupled with an Ortec-Spectrum Master MCA. No $\gamma\gamma$-coincidences have been taken into account.

4. Isotope production in reactions of the primary beam

The determination of the amount of the isotopes produced in the samples during the irradiation proceeds in two steps. The first step is the analysis of the $\gamma$-spectra and the unambiguous assignment of the observed $\gamma$-lines to the corresponding decaying isotopes. The identification of the decaying nuclide is based on the energy of the detected transition and the beta-decay halflife, the latter being estimated after analyzing a sequence of spectra, in which the observed $\gamma$-line is present.

In the second step, based on the known beta-decay halflives of the identified nuclides and the measured number of decays, one calculates the amount of the decaying nuclide of interest, contained in the sample at the end of the irradiation. The final result has to be corrected with respect to
absorption of the outgoing $\gamma$-rays within the volume of the sample, beta-decays in the course of the irradiation, summing effects due to coincident detection of $\gamma$-transitions within a cascade. The R-value for the production of a certain nuclide in one lead sample is defined as:

$$R_{A,Z} = \frac{N_{A,Z}}{Y_{total} \cdot N_{nat,Pb}}$$  \hspace{1cm} (1)

- $R_{A,Z}$: production rate of $(A,Z)$-nuclides
- $N_{A,Z}$: total number of $(A,Z)$-nuclei produced during the irradiation
- $Y_{total}$: total number of projectiles (beam integral)
- $N_{nat,Pb}$: number of Pb-atoms in the sample

4.1. Bismuth isotopes

The $\gamma$-transitions, assigned to the beta-decays of $^{203}$Bi, $^{204}$Bi, $^{205}$Bi and $^{206}$Bi were observed. The beta-decay-halflife of $^{202}$Bi and all lighter Bi-isotopes including their isomers are of 1,72 h and less, therefore their decays were not observed during this experiment. All beta-decay-halflives of Bi-isotopes, heavier than $^{206}$Bi are in the order of years, consequently if even being produced, the activity of $^{207}$Bi and $^{208}$Bi is expected to be far below the observation limits of this measurement, $^{209}$Bi is a stable isotope. The isotope productions of bismuth isotopes are shown on figure 1.

![Figure 1](image)

**Figure 1.** Production rates of isotopes of bismuth and lead, determined in the six lead samples.

4.2. Lead isotopes

The isotope production rates of lead isotopes are shown on figure 1. The lightest observed isotope of lead is $^{198}$Pb with a beta-decay halflife of 2,40 h. The lead isotopes lighter than $^{198}$Pb have beta-decay halflives, which are below one hour, thus their decays were not
observed. The 209.3 keV transition, which coincides with the beta decay of $^{198}$Pb was observed only in the spectra of the samples “1”, “2” and “5”. The decays of the isotopes $^{200}$Pb with $T_1/2=21$ h and $^{201}$Pb with $T_1/2=9.33$ h were observed in all samples. The isotope $^{202}$Pb has a decay half-life of 5.25E4 years. The only $\gamma$-transitions could be observed in coincidence with the decay of the isomer $^{202m}$Pb having a lifetime of 3.53 h. The decay of the isomeric state ($I_p=9-$, $E=2169.83$ keV) proceeds together with the emission of $\gamma$-rays of 960.67 keV (92%), 786.99 keV (50%), 657.49 keV (32%), 422.18 keV (86%). The measured number of transitions of these energies will reveal the production rate of the isomer $^{202m}$Pb, but not the production rate of $^{202}$Pb itself. Thus the obtained result is expected to be much lower if compared to the production rates of the neighbouring isotopes $^{201}$Pb and $^{203}$Pb. The percentage of the produced $^{202}$Pb depends on the isomeric ratio of the reaction, e.g. how many of the residual nuclei have been produced in states with spins greater then 9 relatively to the total $^{202}$Pb production. The calculation of the isomeric ratio of $^{202}$Pb is beyond the scope of the present analysis, however it may serve as experimental result for the population of isomers in particle-induced reactions. The heaviest isotope of lead, whose decay was observed in the accumulated $\gamma$-spectra, is $^{203}$Pb, with a beta-decay half-life of 51.873 h.

4.3. Thallium isotopes

The thallium isotopes, whose decays are observed in the $\gamma$-spectra of the lead samples, are of two origins. A part of the thallium nuclides are formed as residual nuclei in the reactions of the primary beam or its residues with the lead nuclei of the samples. Additionally thallium isotopes are produced in the beta-decays of the lead isotopes obtained in the course of the irradiation. Therefore in order to determine the isotopic production-rates of thallium, the isotopic production rates of lead have to be known in advance. In the cases, where numbers on the production of lead isotopes were not available, the corresponding results for the production of thallium isotopes can be given only as cumulative values. The isotope $^{198}$Tl has a half-life of 5.3 (5) h. The isomers $^{198m1}$Tl and $^{198m2}$Tl have lifetimes of 1.85 h, and 32.1 ms respectively. $^{198m2}$Tl can be produced only in reactions, induced by the primary beam, since the isomeric states are not populated in the beta decay of $^{198}$Pb. $^{198m1}$Tl undergoes with 54% probability beta decay, the internal transition branch amounts 46%, while $^{198m2}$Tl decays only by internal transitions. The intensities of the accompanying $\gamma$-transitions are different for the beta-decays of $^{198}$Tl and $^{198m1}$Tl. The 587 keV transition has an intensity of 0,20(4)% for the decaying $^{198}$Tl and an intensity of 52% for the decaying $^{198m1}$Tl. Thus the 587 keV $\gamma$-line is expected contain mostly decay-events of $^{198m1}$Tl. The decay of $^{198}$Tl was analyzed using the 675,88 keV transition, having 11% intensity, while this transition is not observed in the decay of $^{198m1}$Tl. The decays of $^{199}$Tl and the isomer $^{199m1}$Tl are accompanied also by the strong 411.8 keV transition. The analysis of the 411.8 keV transition has shown, that a part of the intensity originates from the decay of $^{199}$Au. Therefore the 411,8 keV transition was excluded from the analysis. $^{199}$Tl has a decay half-life of 7.42 (8) h. Since no decays of $^{199}$Pb ($T_1/2=90$m) and $^{199m}$Pb ($T_1/2=12,2$m) were observed, the production rate of $^{199}$Pb remained unknown. Therefore the production rates of $^{199}$Tl are provided as cumulative values. $^{200}$Tl has a decay half-life of 26.1 (1) h. The beta-decay of $^{200}$Tl is accompanied by numerous $\gamma$-transitions, the strongest of which – 367,943 keV (Ig=83%), 1205,717keV (Ig=29,9%), 828.32 keV (Ig=10,8%), 579,278keV (Ig=13,8%) were analyzed. The production rates of $^{200}$Pb in the individual samples were obtained in a previous step of the analysis, the final results for the production rates of $^{200}$Tl were calculated as the weighted mean of the individual values, obtained within the analysis of the four $\gamma$-transitions. $^{201}$Tl has a decay half-life of 72.912 (17) h. Also in this case the production rates of the decaying predecessor $^{201}$Pb were previously determined. The strongest $\gamma$-transition occurring after the decay of
201Tl has 10% of intensity and an energy of 167.43 keV. The determination of the production rates of 201Tl was done using this transition.

202Tl has a decay halflife of 12.23 (2) d. To the production of 202Tl contribute the decays of 202Pb and the isomer 202mPb. Since the halflife of 202Pb is 5.25E4 years, no decays of 202Pb have been observed in this experiment. A relevant contribution to the production of 202Tl comes from the decay of 202mPb, which has a lifetime of 3.53h. 202mPb undergoes to 9.5% beta decay and to 90.5% internal transitions as well. The determination of the production rates of 202Tl was done using the 439.56 keV transition. The measured production rates if thallium isotopes are shown on figure 2.

![Figure 2. Production rates of isotopes of thallium, determined in the six lead samples.](image)

5. Numerical simulation and comparison with experimental data

The simulations of the experimental data of the measured production rates have been performed using the MCNPX-nuclear-reaction- and particle transport code. The simulation provided the fluxes of the deuterons, protons and neutrons, measured in units of particles/deuteron/cm². The production-cross-sections were also calculated within the code, except for reactions of less than 150 MeV energy of the incoming particle. For these cases the production cross sections have been calculated using the THALYS code. The production rate $R_{\text{Calc}}$ is calculated as follows:

$$R_{\text{Calc}} = \sum_{A(Pb)} p_{A(Pb)} \left( \phi_n(E_i) \sigma_{n,X(A,Z)}^{A(Pb)}(E_i) + \phi_p(E_i) \sigma_{p,X(A,Z)}^{A(Pb)}(E_i) + \phi_d(E_i) \sigma_{d,X(A,Z)}^{A(Pb)}(E_i) \right) \times 0.001 \times 10^{-24}$$

\(\phi_n(E_i), \phi_p(E_i), \phi_d(E_i)\): calculated neutron-, proton-, deuteron- flux at certain position and fixed energy, measured in [particles/cm²/incoming deuteron].

\(\sigma_{n,X(A,Z)}^{A(Pb)}(E_i), \sigma_{p,X(A,Z)}^{A(Pb)}(E_i), \sigma_{d,X(A,Z)}^{A(Pb)}(E_i)\): cross sections for the production of the nuclide X(A,Z) in neutron-, proton-, and deuteron-induced reactions on a lead isotope of mass A(Pb), for certain energies, measured in milibarns.
The final result is taken as the sum over the four naturally occurring lead isotopes, weighted by their natural abundances $p_{A_i}(Pb)$.

Except for the sample positioned outside the setup, the deviations between the calculated and the measured production ratios do not exceed one order of magnitude. The deviations observed in the sample “0” could result either from an incorrect description of the beam interaction with the sample, or from deviations in the calculations of the deuteron-induced reactions on lead.

The calculated and measured production rates of lead isotopes are in nearly perfect agreement for the positions 1 and 2, while for positions 3,4 and 5 a maximal three-fold difference was observed.

The deviations between the measured and calculated values production rates increase for the isotopes of thallium. The lowest deviation by factor of 1.96 between both values of the production rate was found for $^{202}$Tl in the sample, placed at position 2.

![Graph](image)

**Figure 3.** Ratios between the experimental isotope production rates $R_{exp}$ and the calculated isotope production rates $R_{calc}$ of isotopes of thallium and lead in the five lead samples, placed on the surface areas of the target sections.

The analysis of the simulation data has shown, that the neutron-induced reactions dominate the production of the lead- and thallium isotopes in the target. Based on the compared calculated and measured production rates, underestimated cross-sections for the outgoing proton-neutrons-emission-channels could explain the observed deviations for the isotopes of thallium. For the proper understanding of this effect additional simulations, complementary to MCNPX, of the interaction of the primary beam with the lead target are planned. The obtaining of isotope-production data in the lead target at additional energies of the primary beam are desirable.

**References**

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