Excitation functions for(d,x)reactions on $^{133}$Cs up to $E_d = 40$ MeV

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Abstract

In the frame of a systematic study of excitation functions of deuteron induced reactions the excitation functions of the $^{133}$Cs(d,x)$^{133m,133ng,131mg}$Ba, 134, 132Cs and $^{129m}$Xe nuclear reactions were measured up to 40 MeV deuteron energies by using the stacked foil irradiation technique and $\gamma$-ray spectroscopy of activated samples. The results were compared with calculations performed with the theoretical nuclear reaction codes ALICE-IPPE-D, EMPIRE II-D and TALYS calculation listed in the TENDL-2014 library. A moderate agreement was obtained. Based on the integral yields deduced from our measured cross sections, production of $^{133}$Cs via the $^{133}$Cs(d,2n)$^{131}$Ba $\rightarrow$ $^{133}$Cs reaction and $^{133}$Ba via $^{133}$Cs(d,2n) reactions is discussed in comparison with other charged particle production routes.

Keywords: $^{133}$Cs targets, proton induced reactions, experimental cross sections, model calculations, $^{133m,133ng,131mg}$Ba, 134,132Cs, and $^{129m}$Xe activation products, $^{133}$Cs and $^{133}$Ba radionuclide production

1. Introduction

Cross sections of charged particle induced reactions in Cs play an important role for production of radioisotopes of Ba and Cs, used in different applications. The nuclear data are needed for production of long-lived $^{133}$Ba, used as standard source for calibration of gamma spectrometers and for many other applications (van der Meulen et al., 2010; van der Walt et al., 2008). Shorter-lived $^{133m}$Cs was used for myocardial imaging and for investigation of transport and metabolism process of cesium and as a substitute for potassium (Eybel et al., 1979). The pure EC decay of $^{134}$Cs (X-ray emitter) gained application in inter-

9.4-28.5 MeV energy range (Mocoroa et al., 1966). Nactis products, $^{133}$Cs and $^{133}$Ba radionuclide production

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deuterons (Mukhammedov et al., 1984).

2. Experimental

The excitation functions were measured up to 40 MeV via the activation technique by bombarding stacked CsCl targets with a low intensity deuteron beam at the AVF-930 cyclotron of the Tohoku University (Sendai). Reactions induced on Al monitor foils were used to refine the parameters (energy, intensity) of the incident beam. Special care was taken in preparation of uniform targets with well-known thickness, in determination of the energy and intensity of the bombarding beam along the target stack and in determination of the activities of the samples. Self-supporting pellets (120 mg/cm$^2$) of CsCl targets were made by pressing. The pellets were then stacked with 100 µm thick Al backings and 10 µm thick Al protecting covers. To minimize the water content of the targets, the stock CsCl powder (99.9 %) was dried under vacuum and the pellets were sealed in a plastic bag before irradiation. The targets were irradiated in a He gas atmosphere. For determination of the beam intensity and energy, the complete excitation function of the monitor reactions were re-measured simultaneously by using the cover (10 µm) and the 100 µm backing Al-foils. The target stack was irradiated with 40 MeV incident energy for about a 40 min, at 25 nA beam current. The target integrity after the irradiation was checked. Final flux data were determined from the monitor reactions. The energy of the extracted beam was determined by a calibrated magnetic bending set-up. Possible effect of secondary neutrons was experimentally checked by inserting an additional pellet beyond the charged particle stopping range. The activity of the irradiated samples was measured without chemical separation by using well-proved gammaspectroscopy. Due to the high initial dose rate the first activity measurements were started around one day after the end of bombardment, which resulted in the complete decay of most of the short-lived radioisotopes. The second series of gamma spectra measurement started four days after EOB. The energy degradation along the stack was determined by calculation and by comparison with the re-measured $^{27}$Al($d,x)^{24}$Na monitor reactions (Andersen and Ziegler, 1977; Tarkanyi et al., 1991, 2001) (Fig. 1). The uncertainty of the energy scale was estimated by taking into account the energy uncertainty of the primary beam, the possible variation in the target thickness and the effect of beam straggling. The cross sections were deduced by using the standard activation formula, so called isotopic cross sections were calculated taking into account that Cs is monoisotopic. The decay data, the contributing reactions and the reaction Q-values for production of the measured radioisotopes of Ba, Cs, Xe and their longer-lived isomeric states, taken from (Kinsey et al., 1997; Pritychenko and Sonzogni, 2003; Sonzogni, 2005), are summarized in Table 1. The uncertainties of the cross-sections were estimated by using the error propagation formula used for calculation (of-Weights-and-Measures, 1993). The uncertainties of the contributing factors were: number of bombarding particles (7 %), gamma intensity data (1-3 %), detector efficiency (5 %), peak area (0.1 to 10 %), number of the target nuclei (5 %). Typical overall uncertainties of the cross-sections are around 12-15 %.

3. Theoretical calculations

The cross sections of the investigated reactions were calculated using the pre-compound model codes ALICE-IPPE (Dityuk et al., 1998) and EMPIRE-II (Herman et al., 2007) modified for deuterons by Ignatyuk (D versions) (Ignatyuk, 2011). The theoretical curves were determined using one recommended input data-set (Belgya et al., 2005) without any optimization or adjustment of parameters to the individual reactions or stable target isotopes. Independent data for isomers with ALICE-D code was obtained by using the isomeric ratios calculated with EMPIRE. The experimental data are also compared with the cross section data reported in the TENDL-2014 (Koning et al., 2014) nuclear reaction data library. The TENDL library was developed on the base of the TALYS nuclear model code system (Koning and Rochman, 2012) for direct use in both basic physics and applications (default TALYS calculations).
4. Experimental results

The numerical values of the recently measured cross-sections of the investigated reactions are collected in Table 2. The excitation functions are shown in Figs. 2-8.

4.1. The $^{133}$Cs(d,2n)$^{133m}$Ba, $^{133}$Ba(m+) reactions

No cross section data have been earlier reported for production of the $^{133m}$Ba ($T_{1/2} = 38.9$ h) metastable state (Fig. 2). The values in TENDL-2014, the calculated results of ALICE-D and EMPIRE-D all overestimate the experimental data in different proportions. We could find only one earlier experimental data set for the cumulative activation cross section of the ground state of $^{133}$Ba ($T_{1/2} = 10.52$ a) after the decay of the isomeric state, reported by (Pement and Wolke, 1966). There is an acceptable agreement with our results in the overlapping energy region (Fig. 3). There are large differences in magnitude between the TENDL-2014 theoretical results and the experimental data. The predictions of EMPIRE-D are rather well describing the experimental values over the whole studied energy range, while the ALICE-D values are decreasing too fast above 12 MeV.

Figure 2: Excitation function of the $^{133}$Cs(d,2n)$^{133m}$Ba reaction

Table 1: Decay characteristic of the investigated reaction products $^{133m,133mg,131mg}$Ba, $^{134,132}$Cs, and $^{129m}$Xe
4.2. The $^{133}\text{Cs}(d,2n)^{131}\text{Ba}(m^+)$ reaction

Due to the long cooling time we could not assess cross section data for the short-lived $^{131m}\text{Ba}$ metastable state ($T_{1/2} = 14.6$ min). The cross sections for $^{131}\text{Ba}$ ($T_{1/2} = 11.5$ d) obtained after total decay of the isomeric state (m+) are shown in Fig. 4. The model calculation results in TENDL-2014 support our result both in shape and in magnitude. The EMPIRE-D and ALICE-D are overestimating the experimental values and show opposite energy shifts of the maximum position.

4.3. The $^{133}\text{Cs}(d,p)^{134}\text{Cs}(m^+)$ reaction

The excitation function for production of $^{134}\text{Cs}$ (Fig. 5) was measured after the total decay of the short-lived meta-stable state ($T_{1/2} = 2.912$ h, IT = 100\%) to the long-lived ground state ($T_{1/2} = 2.0652$ a). The agreement with the rather scattered data of Wolke, 1967 is reasonable, where it is possible to compare. The deep valley around 10 MeV in Natowitz results could be reproduce neither with our measurement (no data in this region) nor with the model codes. The significant underestimation of the experimental (d,p) cross section data in the TENDL-2014 library can be observed also in this case. The values predicted by ALICE-D and EMPIRE-D are overestimating the maximum by a factor of two. In Fig. 5 we also present the predictions for (d,p) reactions coming from systematics in this mass region.

4.4. The $^{133}\text{Cs}(d,p2n)^{132}\text{Cs}$ reaction

The measured cross section data of $^{132}\text{Cs}$ ($T_{1/2} = 6.479$ d) are shown in Fig. 6 in comparison with the theoretical predictions of TENDL-2014, ALICE-D and EMPIRE-D. While agreement for the TALYS based result is acceptable, the two other codes strongly underestimate the experimental data. No earlier cross section data are available.

4.5. The $^{133}\text{Cs}(d,p3n)^{131}\text{Cs}$ reaction

We could not measure the direct production of $^{131}\text{Cs}$ ($T_{1/2} = 9.689$ d) as this radionuclide decays without emission of gammas and can only be quantified by a dedicated series of measurements of the complex X-ray spectrum. To estimate the possibilities for direct formation of the medically important $^{131}\text{Cs}$ we present in Fig. 7 the predictions of our ALICE-D and EMPIRE-D calculations together with the TALYS results in the TENDL-2014 library.
4.6. The $^{133}$Cs(d,2p6n)$^{129m}$Xe reaction

The radio isotope $^{129}$Xe has two longer-lived isomeric states: the $T_{1/2} = 8.88$ d half-life metastable state and the stable ground state. The practical threshold of 20 MeV shows that $^{129m}$Xe is produced at lower energy directly via a $^{133}$Cs(d,$\alpha$2n) reaction and not by emission of independent nucleons. The measured excitation function for production of $^{129m}$Xe is shown in Fig. 8 in comparison with the theoretical results. Here the results of ALICE-D and TENDL-2014 describe well the shape and values of our few experimental points. EMPIRE-D seems to be too large, but it reflects the effect of alpha-channels, description, which differs very strongly in ALICE and EMPIRE (different pre-equilibrium models).

5. Integral yields

Based on the experimentally determined cross sections we have calculated the differential and integral yields of the produced radio-isotopes. The yields are so called physical yields, calculated for an instantaneous irradiation (Bonardi, 1987; Otuka and Takács, 2015). Figs. 9 and 10 show the integral yields for the production of the investigated radionuclides of Ba, Cs and Xe in comparison with the few experimental integral yield data in the literature (see literature review in the introduction). The experimental thick target yield are in good agreement in the case of $^{133m}$Ba and $^{134g}$Cs with our yield data deduced from experimental cross sections. In the case of $^{133g}$Ba the previous data are somewhat higher, and there are no previous measurements for the rest of the radioisotopes.
6. Production of $^{131}$Cs and $^{133}$Ba

6.1. $^{131}$Cs

As it was discussed in more detail in our previous paper (Tárkányi et al., 2009b) for commercial use $^{131}$Cs is presently produced at high flux nuclear reactors by radioactive decay of its $^{131}$Ba mother obtained through neutron capture on naturally occurring $^{130}$Ba (natural Ba contains only 0.106 % $^{130}$Ba) or on enriched $^{130}$Ba targets. As it was mentioned, we have studied already a few charged particle induced production routes for $^{131}$Ba/$^{131}$Cs and we compared the $^{131}$Xe(p,n), $^{127}$I(α,γ), $^{132}$Ba(p,x) nat$^{132}$Ba(p,x), $^{133}$Cs(3n), $^{129}$Xe(α,2n), natXe(α,xn) reactions. The yields of the most practical low and medium energy charged particle production routes (Tárkányi et al., 2009b), completed with our new experimental results on $^{133}$Cs(d,4n) reaction are summarized in Fig. 11. According to Fig. 11 out of the possible charged particle production routes the indirect production through the $^{132}$Ba(p,x) reaction is the most productive, but the isotopic abundance of the $^{132}$Ba is only 0.101 %. The $^{133}$Cs(p,3n)$^{131}$Ba reaction also has high yield and allows using targets with natural (mono)isotopic composition. The situation is similar in case of relying on the $^{133}$Cs(d,4n) reaction, but a higher energy accelerator is required. The direct production of $^{131}$Cs through the $^{131}$Xe(p,n) reaction has also reasonable yield and this production route starts at the lowest energy.

6.2. $^{133}$Ba

The production of $^{133}$Ba via a (n,γ) reaction on $^{132}$Ba (which has a 0.101 % natural abundance) in a nuclear reactor has been reported, but the product is not carrier free. The cyclotron production of no carrier added $^{133}$Ba is possible using charged particle induced reactions: via (p,n) or (d,2n) reactions on monoisotopic $^{133}$Cs, via $^{132}$Xe(α,3n), nat$^{132}$Xe(α,xn), $^{131}$Xe(α,He,n) and nat$^{132}$Xe(α,He,xn) reactions on gas targets using less frequently available and nowadays very expensive $^{3}$He.

Table 2: Experimental cross sections of the investigated reaction products:

| Deuteron energy (MeV) | $^{131}$Ba | $^{133}$Ba | $^{132}$Ba | $^{130}$Cs | $^{132}$Cs | $^{133}$Xe | $^{131}$Ba | $^{133}$Ba | $^{132}$Ba | $^{130}$Cs | $^{132}$Cs | $^{133}$Xe |
|-----------------------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| 5.0                   | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 6.0                   | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 7.0                   | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 8.0                   | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 9.0                   | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 10.0                  | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 11.0                  | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 12.0                  | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 13.0                  | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 14.0                  | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 15.0                  | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 16.0                  | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 17.0                  | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 18.0                  | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 19.0                  | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |
| 20.0                  | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       | 1.0       |

Figure 9: Integral yields for production of radioisotopes of Ba by bombarding $^{133}$Cs with deuterons

Figure 10: Integral yields for production of radioisotopes of Cs and Xe by bombarding $^{133}$Cs with deuterons
physical yield (GBq/C) of 133Cs, the out of the possible charged particle production routes, experimental data are missing. According to Fig. 12, section data, or TENDL-2014 theoretical results when yield calculations are based on fitted experimental cross sections become significantly higher. The possible use of our experimental data for production 131Ba(131Cs) and 133Ba (section 6.) was discussed in comparison with other production routes, showing the importance of deuteron induced reactions in production of the above mentioned medically important radioisotopes.

7. Summary and conclusions

We present experimental cross sections for the nuclear reactions 133Cs(d,x), 133Cs(3n), 133Cs(4n), 134Ba, 134Cs, and 129mXe up to 40 MeV deuteron energies. The new data are first experimental activation data sets, except for 133Ba and 134Cs. The comparison with the recently published TENDL-2014 library shows only moderate agreement in few cases, and completely missing in other cases. Also the D-versions of the ALICE-IPPE and EMPIRE-II codes (specially adapted for deuteron induced reaction) are still largely deviant from the experimental data. The possible use of our experimental data for production 131Ba(131Cs) and 133Ba (section 6.) was discussed in comparison with other production routes, showing the importance of deuteron induced reactions in production of the above mentioned medically important radioisotopes.

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