Eur. Phys. J. A **23**, 257–264 (2005)
DOI 10.1140/epja/i2004-10095-7

THE EUROPEAN PHYSICAL JOURNAL A

On-line production of Rb and Cs isotopes from uranium carbide targets

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Received: 11 August 2004 / Revised version: 11 October 2004 /
Published online: 13 December 2004 –
Communicated by C. Signorini

Abstract. A series of on-line mass separation experiments have been performed at the IRIS facility to measure the yield and release of Rb and Cs neutron-rich isotopes produced by fission reaction of ^{238}U. A 1 GeV proton beam was used to bombard uranium carbide targets with the densities of 11 g/cm^3 and 1.5 g/cm^3 held at temperatures in the range (2000–2230) °C. The release curves of Rb and Cs long-lived isotopes were measured from both kinds of targets. The overall production efficiency was determined making use of experimentally measured cross-sections of that isotope production. Comparison of the experimental yields of Rb and Cs isotopes with the calculated ones after corrections for losses due to finite release times suggests that the diffusion is the dominating process reducing the efficiency for short-lived isotopes. When normalized to the same thickness, an enhancement for the high-density rod target of the measured isotope yields is observed when going far from stability. This is possibly explained by the reactions induced by secondary neutrons. A significant odd-even effect with higher yields of Cs even neutron isotopes has been observed, confirming a similar effect obtained in earlier experiments.

PACS. 25.85.Ec Neutron-induced fission – 25.85.Ge Charged-particle-induced fission – 29.25.Rm Sources of radioactive nuclei – 41.75.Ak Positive-ion beams

1 Introduction

The increasing interest for the study of exotic neutron-rich nuclei far from stability motivates the development of uranium carbide targets, which are well suited for production of n-rich isotopes of a large set of elements of the periodic table by the ISOL (Isotope Separation On-Line) method. In addition, these targets enable the production of neutron-deficient heavy nuclei from Tl to Fr by fragmentation reactions, the latter being of considerable interest for testing of fundamental interactions.

Although uranium carbide targets [1,2] have been used for many years at ISOL facilities, the question still remains of which target density is the best to produce a selected nuclide. It is thus useful to carry out on-line experiments comparing the yields of different nuclides with UC targets of different density. Release times measured in the same experimental conditions are needed to understand the mechanism of the processes taking place in the target material and in the volume of the target-ion source unit. Such experiments have been performed in the frame of the Collaboration PNPI-LNL-GANIL-Orsay at the IRIS facility, where a program on measurements of the production yields and delay times of a large variety of different on-line produced species is being carried out [3,4]. Here results for Rb and Cs isotopes are presented.

2 Experimental

A schematic drawing of the target-ion source unit construction [5] is shown in fig. 1. The construction holding the target material consists of two containers. The outer container of 0.25–0.30 mm wall thickness is heated by the ohmic heating. It is made of 5–6 layers of a rolled tungsten foil 50 μm thick. The use of tungsten for the outer cavity construction ensures a high mechanical thermo-stability

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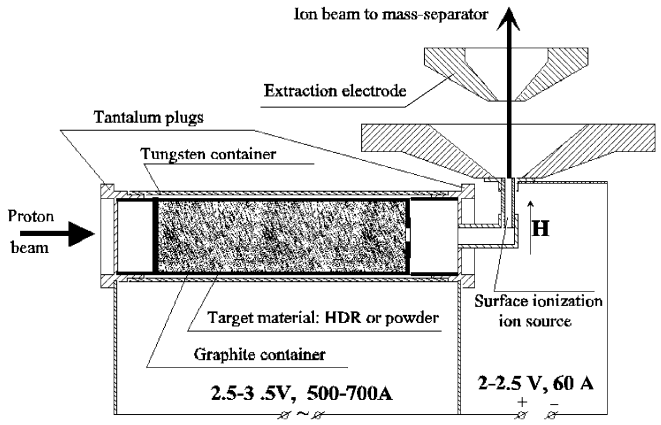

Fig. 1. Schematic drawings of high-density rod (HDR) and low-density powder (LDP) uranium carbide targets coupled with a high-temperature surface ionization ion source.

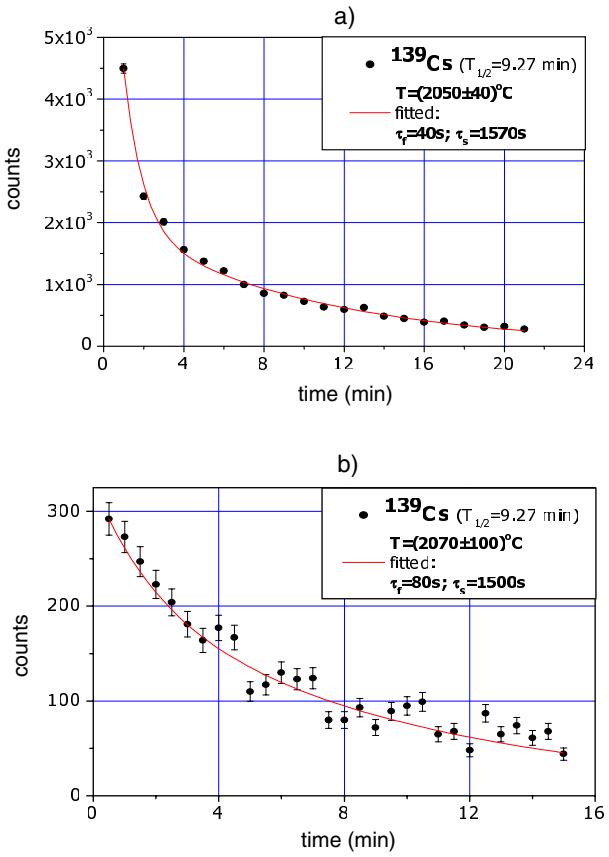

Fig. 2. Delay time curves of ^{139}Cs released from a) HDR and b) LDP targets. Generally the error of temperature measurement maintaining one target is 40 °C. When the delay curve from a LDP target was measured, the error of the temperature measurement was up to 100 °C. The error of defining $\tau_{\rm f}$ and $\tau_{\rm s}$ is up to 50% and 30%, respectively.

that is necessary for on-line experiments during which the target unit has been maintained at a very high temperature for a long period of time. The inner container in which the target material is placed is manufactured from a highly porous graphite. Construction of the target assembly has ensured the housing of the graphite container with a chemically aggressive target material being supported by the end tantalum plugs to avoid any touch of graphite to the tungsten in its most hot central parts. Two kinds of target materials, both uranium carbides, have been tested during the experiments: a low-density powder (LDP) ($\rho = 1.5$ g/cm^3, thickness 1.9 g/cm^2), and a high-density rod (HDR) ($\rho = 11$ g/cm^3, thickness 8.2 g/cm^2). The initial grain size of the powder target material was $\leqslant$ 10 microns. As to the HDR target, according to the information from a supplier, that can be considered as a preliminary one, the initial grain size of the target material was (20 $\pm$ 10) microns. The fission of uranium was induced by a 1 GeV proton beam with an intensity in the range 0.05–0.07 μA. A target-ion source unit with a high-temperature surface ionization ion source [5] was used for the production of isotopes of the alkaline elements Rb and Cs. The temperature of the ion source was kept in the range of (2000–2100) °C.

The measured neutron-rich isotopes of Rb and Cs were selected by their mass number, collected on a movable tape, transported to a counting position and identified by the characteristic γ-lines emitted in their β-decay. The γ-rays were recorded with a coaxial high-purity germanium detector of 1.2% efficiency at 1.3 MeV and 3 keV FWHM.

Peak areas were corrected for detection efficiency and intensities per decay according to the table of isotopes to obtain the number of decays of the nuclei of interest during the counting period. The yields (ions/s impinging on the collector tape) were finally deduced after a correction taking into account the measurement times and the radioactive half-lives [6]. In the case of an isomeric state presence (^{90}Rb) we measured the appropriate γ-line in order to introduce the necessary correction to the measured isotope yield.

3 Measurement results and discussion

3.1 Yields and delay times

Yields and release times have been measured in comparable temperature conditions for both the HDR and LDP uranium carbide targets. The detailed description of the yield and delay time measurement method is presented in [4]. Release time measurements have been carried out at a temperature of about 2050 °C for the high-density rod and the low-density powder target. A measured release time value is the time interval when activity released from the target drops two times after switching off the proton beam. In fig. 2 the delay curves of ^{139}Cs ($T_{1/2} = 9.27$ min) for a high-density rod and low-density powder are presented. Figure 3 demonstrates the ^{89}Rb ($T_{1/2} = 15.15$ min) delay curve from a HDR target measured in similar experimental conditions. Generally the error of temperature measurement maintaining one target was 40 °C, but when the delay curve of ^{139}Cs from a LDP target was measured it was up to 100 °C due to the target heating instability. The release curves have a fast ($\tau_{\rm f}$) and slow ($\tau_{\rm s}$) component of about one and twenty-five minutes

Table 1. Fission-residues Kr, Rb, Xe and Cs cross-sections (in mb) in the spallation reaction ^{238}U + p at 1 GeV [8].

Mass number	**89**	**90**	**91**	**92**	**93**	**94**	**95**
σ_{Kr}	6.03	5.14	3.26	1.74	0.66	0.23	0.07
σ_{Rb}	9.2	8.2	7.4	6.12	4.6	2.4	1.17
Mass number	**139**	**140**	**141**	**142**	**143**	**144**	**145**
σ_{Xe}	3.15	2.11	0.77	0.32	0.06	0.01[(a)]	0.001[(a)]
σ_{Cs}	3.55	3.34	3.4	2.05	1.04	0.4	0.12

[(a)] Cross-section obtained by extrapolation from light isotopes.

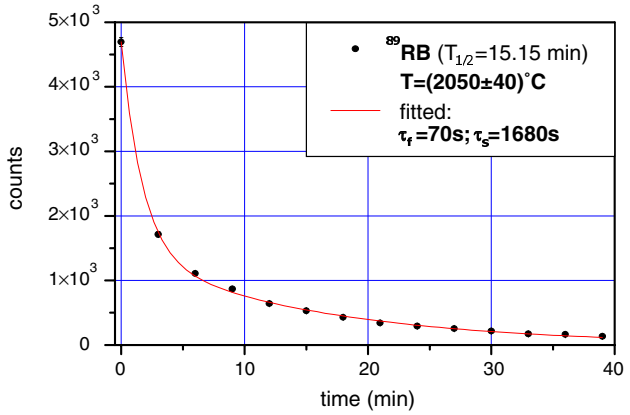

Fig. 3. Delay time curve of ^{89}Rb released from the HDR target measured at a temperature of 2050 °C. The error of defining τ_f and τ_s is up to 50% and 30%, respectively.

defined with typical errors up to 50% and 30%, respectively. The formula of the measured release curve fitting was given in [7]. One of the possible explanations of a complex character of the measured delay curves for ^{89}Rb and ^{139}Cs is that they can be disturbed by the contribution of decay in the target material of mother nuclei ^{89}Kr and ^{139}Xe with the half-life of 3.2 min and 41 s, respectively. As we have not measured the yield and delay time for Kr and Xe, we cannot correctly estimate their contribution to the measured Rb and Cs isotopes yield and release curves. Therefore we present the measured yields in two limiting cases, considering: a) the release time of Kr and Xe atoms produced in the target is very short and there is not any perturbation of the measured yield and b) the release time of Kr and Xe is very long and contribution of their decay to the measured yield has a maximum value. That contribution was calculated making use of known production cross-sections for isotopes of Kr and Xe [8]; those are precursors for measured Rb and Cs isotopes. In table 1 the corresponding cross-sections of production of Rb, Kr and Cs, Xe neutron-rich isotopes are presented. As one can see from table 1, the contribution from Kr and Xe isotopes decay to the measured yields for Rb and Cs lying close to stability can reach more than 50%, while for short-lived isotopes far from stability it is negligible. It should be pointed out that contribution can be much less in reality, as atoms of noble gases have much shorter diffusion and effusion release times than alkalis.

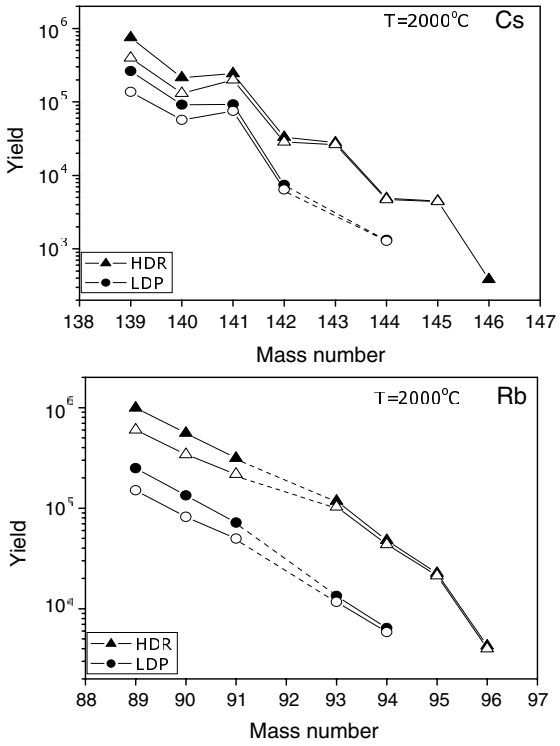

Fig. 4. The yields of Cs and Rb isotopes released from the HDR and LDP targets at 2000 °C. The yields are normalized to 1g/cm^{2}target thickness and 0.1 μA proton beam intensity. Black triangles and circles show the yields considering that there is not any perturbation of the measured yields by Xe and Kr decay in the target. Open triangles and circles show the yield suggesting that perturbation is maximal.

Figure 4 demonstrates the yields of Rb and Cs neutron-rich isotopes released from the LDP and HDR targets at 2000 °C considering the pointed-out possibilities. Black triangles and circles show the yields from HDR and LDP targets, respectively, considering that there is not any perturbation of the measured yields by decay of Kr and Xe in the target. Open triangles and circles represent the yields suggesting that the perturbation is maximal. Figure 5 shows Cs isotope yields measured at a temperature of 2230 °C accepting similar considerations. As one can see from figs. 4 and 5 a possible perturbation of the measured yields exceeding the measurement errors may occur

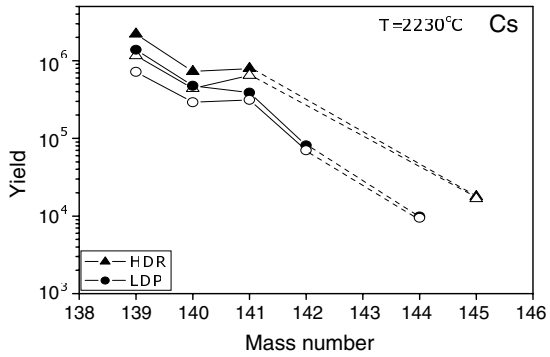

Fig. 5. The yields of Cs isotopes released from the HDR and LDP targets at 2230 °C. The yields are normalized to 1g/cm^2 target thickness and 0.1 μA proton beam intensity. Black triangles and circles show the yields considering that there is not any perturbation of the measured yields by Xe and Kr decay in the target. Open triangles and circles show the yield suggesting that perturbation is maximal.

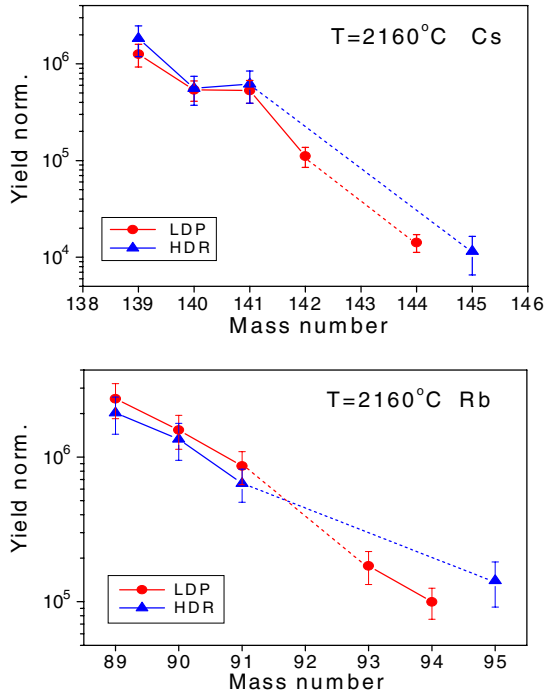

Fig. 6. The yields of Cs and Rb isotopes released from the HDR and LDP targets at 2160 °C. The yields are normalized to 1 g/cm^2 target thickness and 0.1 μA proton beam intensity. Black triangles and circles show the yields considering that there is not any perturbation of the measured yields by Xe and Kr decay in the target.

for ^{139,140}Cs and for $^{89-91}$Rb. In figs. 6 and 7 the yields of Rb and Cs isotopes from both targets are presented measured at temperatures 2160 °C and 2230 °C, respectively. All measured yields are presented for the target thickness and the proton beam intensity normalized to 1 g/cm^2 and to 0.1 μA.

To compare the obtained results with already published ones in table 2 the yields of long-lived Rb and Cs isotopes which are not influenced considerably by the tar-

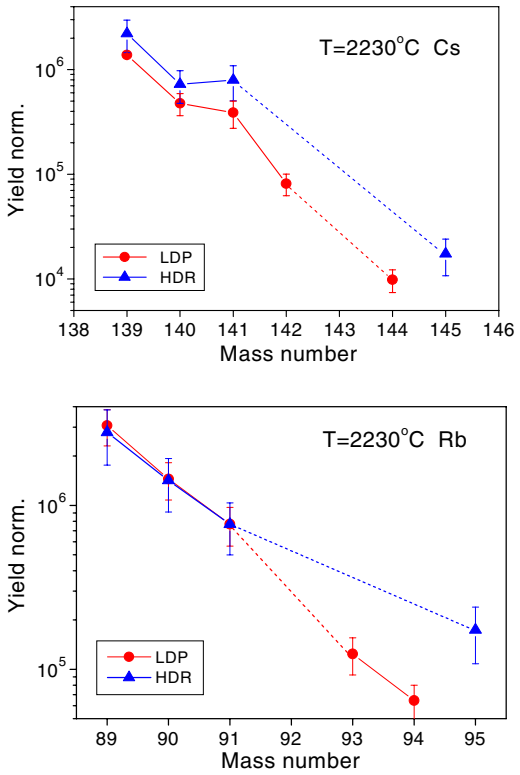

Fig. 7. The yields of Cs and Rb isotopes released from the HDR and LDP targets at 2230 °C. The yields are normalized to 1 g/cm^2 target thickness and 0.1 μA proton beam intensity. Black triangles and circles show the yields considering that there is not any perturbation of the measured yields by Xe and Kr decay in the target.

Table 2. The normalized yield of long-lived Rb and Cs isotopes measured at the ISOLDE and IRIS facilities.

Isotope	^{89}Rb	^{90}Rb	^{140}Cs
$Y_{\rm ISOLDE}$	2×10^8	1×10^8	9×10^7
$Y_{\rm IRIS}$	3×10^7	1.4×10^7	7.3×10^6

get temperature measured in the present work and at the ISOLDE facility [9] are presented. As one can see from table 2, the ratio $Y_{\rm ISOLDE}/Y_{\rm IRIS} = 7$ for ^{89}Rb, 7 for ^{90}Rb and 12 for^{140}Cs, where $Y_{\rm ISOLDE}$ and $Y_{\rm IRIS}$ are the yields measured at the ISOLDE and IRIS facilities, normalized to the target thickness of 1 g/cm^2 and proton beam intensity 1 μA. This discrepancy is rather significant and hardly can be explained by the fact that at ISOLDE a 4π-β counter was used for the collected activity detection, while at IRIS a γ-detector for the yield measurement was applied. To understand more clearly the reason of that discrepancy a close cooperation between ISOLDE and IRIS groups is needed. For the efficiency measurements further experiments at the IRIS facility will be carried out as well.

Summarizing the obtained results, it is worth noting that: a) the normalized yields of Rb and Cs isotopes from a HDR target are close or at some temperatures higher than from a LDP target that can be explained by the error in temperature establishing, when two targets have been

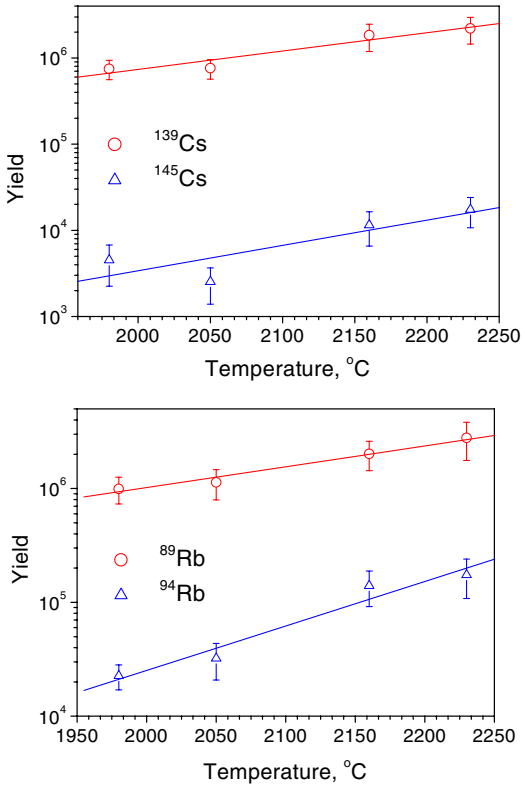

Fig. 8. Temperature dependence of the yields of ^{139}Cs ($T_{1/2}$ = 556.2 s), ^{145}Cs ($T_{1/2}$ = 0.6 s) and ^{89}Rb ($T_{1/2}$ = 909 s), ^{94}Rb ($T_{1/2}$ = 2.7 s) produced from the HDR target.

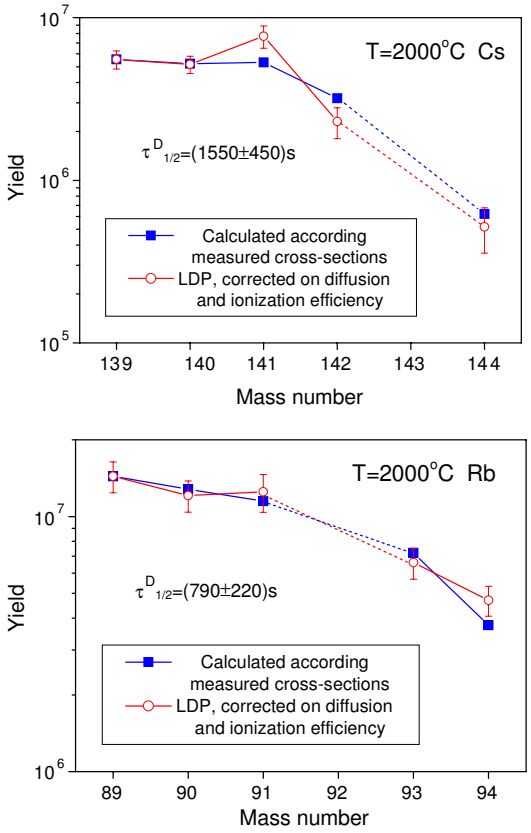

Fig. 9. Yields of neutron-rich Rb and Cs isotopes released from LDP measured at 2000 °C. The yields are normalized to 1g/cm^2 target thickness and 0.1 μA proton beam intensity. No perturbation by Xe and Kr isotope decay is supposed. The yields for the low-density powder target corrected for decay losses due to diffusion and the ionization efficiencies are adjusted to reproduce the calculated values for ^{89}Rb and ^{139}Cs. The good agreement suggests that diffusion is the process controlling the release of Rb and Cs.

compared; b) the trend of the yield curves *versus* mass number demonstrates higher yields of isotopes far from stability from a HDR target; c) experimental data for Cs isotopic distribution shown in figs. 4 and 5 demonstrate a noticeable odd-even structure with higher yield values for even neutron isotopes that is in a good agreement with the measured one in [10].

The production yields of Rb and Cs as a function of temperature are shown in fig. 8. The yield of short-lived ^{94}Rb increases with temperature faster than the yield of long-lived ^{89}Rb. For ^{145}Cs the spread of the points is too large to make a similar conclusion.

Assuming that the release time has been dominated by the diffusion process, the efficiency factor describing decay losses can be expressed as follows [11]:

$$\varepsilon_{\mathrm{D}} = 3\alpha^{1/2}[\coth(\alpha^{-1/2}) - \alpha^{1/2}]\,. \qquad (1)$$

In that expression $\alpha \equiv \tau_{\mathrm{N}}/\pi^2\tau_{\mathrm{D}}$, where τ_{N} is the nucleus lifetime and τ_{D} is the delay time. Correspondingly $\tau_{\mathrm{N}} = T_{1/2}/0.693$, $\tau_{\mathrm{D}} = \tau_{1/2}/0.693$, where $T_{1/2}$ is the value of the nucleus half-life and $\tau_{1/2}$ is the half-release time. In our experiments we measured the value $\tau_{1/2}$ which is the time interval when activity released from the target drops two times after switching off the proton beam. We have indicated $\tau_{1/2} \equiv \tau^{\mathrm{D}}_{1/2}$, emphasising that the obtained half-release time is the delay time determined by the diffusion process.

Figure 9 compares Rb and Cs isotope yields measured from a low-density target at a temperature of 2000 °C with the calculated ones based on experimental cross-sections [8] and including the decay losses by diffusion according to formula (1). The best agreement of measured and calculated yields in the case when there is not any perturbation by Kr and Xe decay have been achieved by inserting the delay time value $\tau^{\mathrm{D}}_{1/2} = (1550 \pm 450)$ s and ionization efficiency $\varepsilon_{\mathrm{i}} = 10\%$ for Cs and $\tau^{\mathrm{D}}_{1/2} = (790 \pm 220)$ s, $\varepsilon_{\mathrm{i}} = 2.5\%$ for Rb. The χ^2 value was 2 and 1.5 for Cs and Rb yield fitting, respectively. For the yield correction, when a maximal contribution of Kr and Xe isotopes decay was suggested, the best fitting was obtained inserting the delay time (320 ± 80) s and ionization efficiency of 4% for Cs and (200 ± 60) s, 1% for Rb (see fig. 10). We can conclude the variant suggesting the absence of a noticeable perturbation of the Rb and Cs measured yield by Kr and Xe decay in the target is much closer to reality, as it gives delay time values consistent within experimental errors with the measured ones.

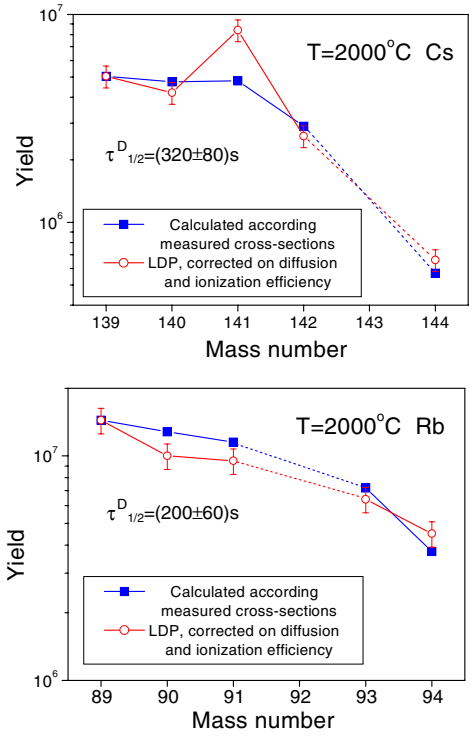

Fig. 10. Yields of Rb and Cs isotopes as in fig. 9, but maximal contribution from Xe and Kr decay in the target is supposed.

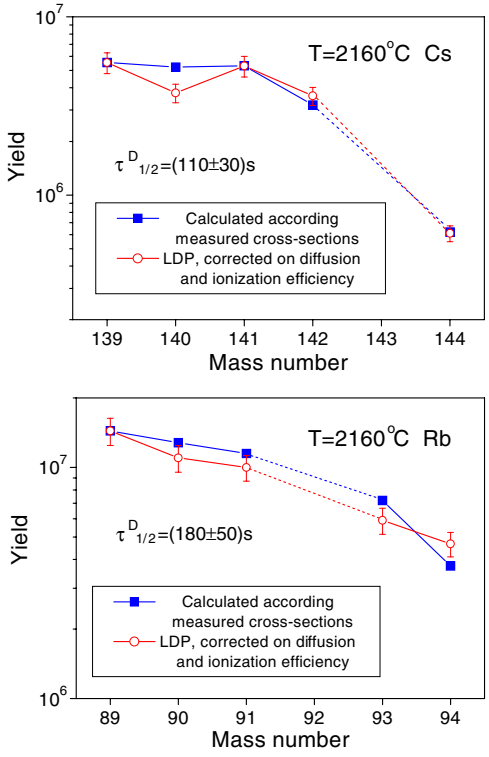

Fig. 11. Yield of Rb and Cs isotopes as in fig. 9, but measured at 2160 °C.

Figure 11 gives a comparison of measured and calculated yields at a temperature 2160 °C. When no perturbation was suggested the best agreement of measured and

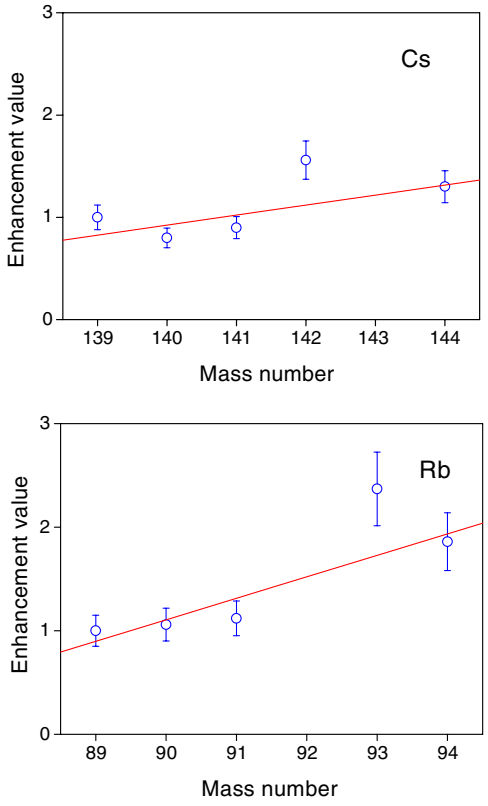

Fig. 12. The enhancement value of the yields of Cs and Rb isotopes with the HDR target. The yields are normalized at ^{89}Rb and ^{139}Cs to emphasize an excess of production of neutron-rich isotopes with the high-density target, presumably due to production of secondary neutrons.

calculated yields was achieved with the release time value (110 ± 30) s and ionization efficiency of 25% for Cs and (180 ± 50) s and 16% for Rb. The ionization efficiency rise with the target temperature can be explained by the fact that if the target temperature is higher than the temperature of the ion source, atoms of Rb and Cs are more effectively ionized on a hot inner surface of the tungsten target container than in the ion source. A lower than it was expected ionization efficiency for Rb and Cs can be explained by the ionizer inner surface carbonization that decreased considerably its work function. The off-line measured ionization efficiency of a similar tungsten ionizer connected to the oven for a calibrated sample evaporation was 45% for Rb and 55% for Cs atoms.

In general the yield trend of Rb and Cs isotopes measured at different temperatures shows that the normalized yields from the HDR target are higher than from a low-density powder target when going far from stability. It is possible to interpret that result as an indication of a larger chance for fission induced by secondary particles (mostly neutrons produced via (p, xn) or fission) in the high-density target since their reaction rates are scaling quadratically with the target density. In such a case, an enhancement of the most neutron-rich isotopes with respect to those closer to the valley of stability is anticipated to occur [12,13]. In order to facilitate the observation of

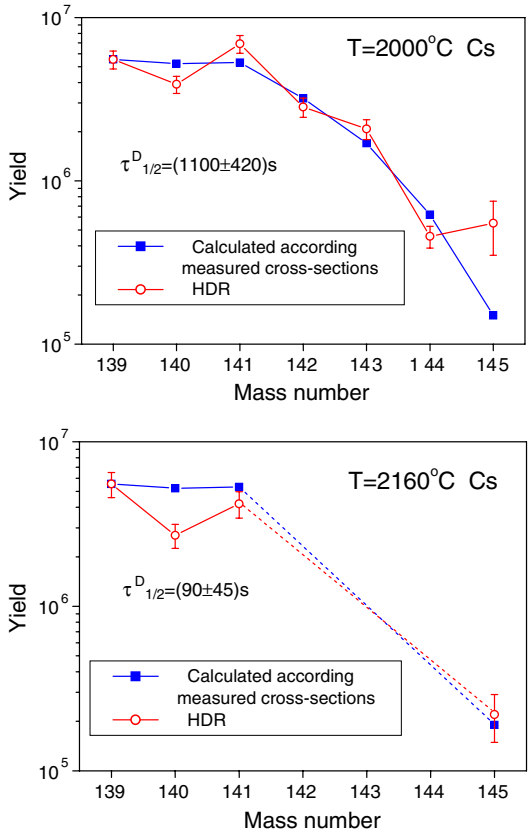

Fig. 13. Yields of neutron-rich isotopes from HDR target measured at 2000 °C and 2160 °C, corrected on the diffusion, ionization efficiency and on the enhancement factor.

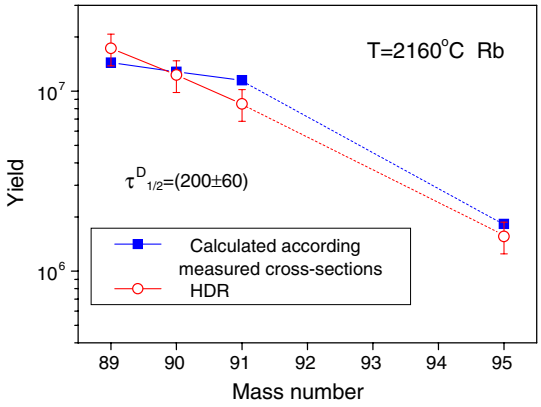

Fig. 14. Yields of Rb isotopes from HDR target measured at 2160 °C, corrected on the diffusion, ionization efficiency and on the enhancement factor.

such an effect the measured yield ratio for the HDR and LDP targets normalized at ^{89}Rb and ^{139}Cs has been put as a function of atomic number. That dependence is shown in fig. 12. There is a clear indication of a larger enhancement of the yield of Rb isotopes far away from stability with the high-density target. For Cs isotopes that effect was almost in the limits of experimental errors. To get more unambiguous conclusions the yield measurements of isotopes further from stability from HDR and LDP target are needed.

Figures 13 and 14 show the yields of Cs and Rb isotopes from a HDR target corrected taking into account the diffusion mechanism and the enhancement factor. As one can see, the evolution of measured yields is consistent with diffusion in the same way as for a low-density powder target. The release time values are equal within the limits of error bars for both kinds of targets.

3.2 Yield odd-even effect

As one can see from the measured yield curves, a large odd-even effect with higher yields of Cs even neutron isotopes has been observed, confirming a similar effect obtained in earlier experiments [10], and in our target tests, making use of low- and high-density powder uranium carbide targets [13]. Most clearly that effect is presented in fig. 13 by the Cs isotope yield trend measured at a temperature of 2000 °C, where all appropriate corrections have been introduced. As was demonstrated by the series of experiments carried out, that effect does not depend either on the way of neutron-rich isotope production (both proton- and neutron-induced fission reactions were used), or on the type of uranium carbide target utilized. The Cs isotope yields are very suitable for observation of that effect, as all measured isotopes have no isomer states and the obtained yield data have a high level of reliability. It should be pointed out that the appropriate cross-sections measured (see table 1) do not demonstrate any odd-even structure. From the other side the LAHET code calculations [14] give odd-even cross-section effect which is good consistent with the obtained experimental data.

4 Conclusions

The yields of on-line mass-separated Rb and Cs isotopes produced by fission of ^{238}U induced by 1 GeV protons have been studied for a high-density rod (11 g/cm^3) and a low-density powder (1.5 g/cm^3) uranium carbide targets. The yields of Rb and Cs neutron-rich isotopes from the HDR are higher or equal to those from the LDP (normalized to equivalent target thickness). That can be explained by the temperature defining errors, and possibly by the larger rate of fission reactions induced by secondary neutrons, in accordance with the observed enhancement of the production of the most neutron-rich isotopes. The measured release times of Rb and Cs long-lived isotopes in the temperature interval (2050–2160) °C had close values for the investigated target materials. This allowed the measured yields of Rb and Cs isotopes from a HDR and LDP targets to be corrected using the close values of the delay parameters.

The evolution of the experimental yields with respect to the calculated in-target productions *versus* mass number is consistent with diffusion, being the main process responsible for a finite release time of Rb and Cs. It is worth

pointing out that this applies to both the low-density powder and high-density rod targets to a large interval of radioactive half-lives of the measured isotopes (^{89}Rb–^{95}Rb, $T_{1/2}$ = 909 s–0.38 s and ^{139}Cs–^{145}Cs, $T_{1/2}$ = 556 s–0.6 s) in the temperature region of (2000–2160) °C.

The yield curves of Cs isotopes demonstrates a significant odd-even structure with higher yield of even neutron isotopes, confirming the same effect obtained in earlier experiments, making use of a high-density powder uranium carbide target for Rb, Cs and In isotopes production by proton- and neutron-induced fission. This effect has a universal character and does not depend either on the way of neutron-rich isotope production (both proton- and neutron-induced fission reactions were used), or on the type of uranium carbide target utilized. The appropriate cross-sections measured in the reaction ^{238}U + p at 1 GeV do not demonstrate any odd-even structure.

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