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S. A. Karamian[1], J. J. Carroll[2]

CALCULATED YIELD OF ISOMER DEPLETION DUE TO **NEEC** FOR ^{93m}Mo RECOILS

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[1]E-mail: karamian@nrmail.jinr.ru
[2]US Army Research Laboratory, Adelphi, Maryland, USA

Карамян С. А., Кэрролл Дж. Дж.
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Расчет выхода разрядки изомера вследствие NEEC для ядра отдачи ^{93m}Mo

В современных публикациях уделяется внимание возможностям разрядки ядерных изомеров за счет атомно-ядерных кооперативных процессов, таких как NEET и NEEC, при использовании накопительных колец и электромагнитных ловушек. Недавно была предложена новая схема, требующая применения только стандартных методов для получения изомера и торможения ядер отдачи в газе. Производящая реакция и конкретное ядро-изомер должны быть выбраны специально, чтобы удовлетворить требованиям резонанса для NEEC, обеспечивающего трансмутацию изомера в основное состояние через известный промежуточный уровень возбуждения. В данной работе проведены количественные расчеты и характеризованы процессы получения и разрядки изомера ^{93m}Mo в относительно простом эксперименте с использованием пучка ионов ^{91}Zr. Такие опыты могут быть осуществлены на существующих и работающих ускорителях в GSI (Дармштадт) или в ОИЯИ (Дубна). Ядро ^{93m}Mo должно быть получено в газовой мишени He в реакции ^{4}He (^{91}Zr, $2n$). Затем это ядро продолжает двигаться в газе с высокой скоростью и может трансмутировать в результате NEEC при захвате электрона на атомную вакансию, созданную за счет глубокой обдирки атомных оболочек.

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Karamian S. A., Carroll J. J.
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Calculated Yield of Isomer Depletion due to NEEC for ^{93m}Mo Recoils

Possibilities for nuclear isomer depletion due to atomic-nuclear cooperative processes such as NEET and NEEC with application of storage rings and electromagnetic traps are of interest in modern literature. Recently, a newer scheme requesting only standard techniques for isomer production in nuclear reactions combined with stopping of recoils in gas has been proposed. The isomer sample and the producer reaction must be chosen specially to meet the requirements for NEEC resonance in transmutation of the isomer to ground state via definite intermediate level. At the present work, quantitative calculations were carried out for production and depletion of the ^{93m}Mo isomer at relatively simple experiment using ^{91}Zr ions. Such studies could be arranged at already existing and operating accelerators, for instance, in GSI, or in Dubna. The ^{93m}Mo nuclei produced in He-gas target due to the ^{4}He (^{91}Zr, $2n$) reaction continue to move in gas with high velocity being then depleted due to NEEC in highly-ionized species.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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INTRODUCTION

For production of isomers in nuclear reactions, charged particle beams and solid targets are normally used. A significant recoil momentum is transferred to the reaction products by the bombarding projectiles. The atomic excitation and deep ionization take place with a high probability over the transmission of swift ions (recoils) in solid, or in gas matter [1]. At high velocity, near 10% of the light velocity, the heavy-element ions could be stripped to a charge state of (40–50)+ that is comparable to the charge state considered for detection of NEEC in electron-beam ion trap (EBIT). The electron impact energy providing NEEC is also supplied by the ion velocity in the stopper material. A gas absorber may supply better conditions for detection of NEEC radiation and also for stability of an atomic configuration over a time needed for electron capture.

Depletion of ^{242m}Am isomer by means of NEET was described in [2] attracting empirical and theory knowledge. Relatively high probability could be expected for ions confined in ECRIT trap. Another process, namely NEEC, may be also productive for this isomer depletion, as was mentioned in [2]. A possibility to discover NEEC process in experiment on confinement of the ^{242m}Am isomeric ions in EBIT trap was proposed in [3, 4]. In calculations [3], the depletion yield was deduced to be relatively low, being still at a detectable level, and this was confirmed in [2] for the same case of NEEC in EBIT, despite another approach in estimates.

A new scheme for NEEC detection has been proposed and tested by calculations for the ^{242m}Am isomer depletion in [2]. The scheme is based on the idea of swift reaction products (isomers) stripped to high charge states under transmission in matter and excited due to electron capture to the intermediate nuclear level. In application to ^{242m}Am the quantitative characteristics are as follows: an electron collision with I-like ^{242m}Am^{42+} ion at 2.658 keV corresponds to the resonance conditions for NEEC [3, 4]. An electron impact of similar velocity takes place in the absorber at ion energy of about 4.9 MeV/u in laboratory system (l.s.). The ^{242m}Am ions could be produced in inverse kinematics irradiating the deuterium target by 5 MeV/u ^{241}Pu ions. The velocity of the recoil products is enough for resonance NEEC conditions with excitation of an intermediate level located at 4.1 keV above the isomer. The case of ^{242m}Am is characterized in [2–4] as one of the best for experimental observation of isomer depletion due to NEEC. But

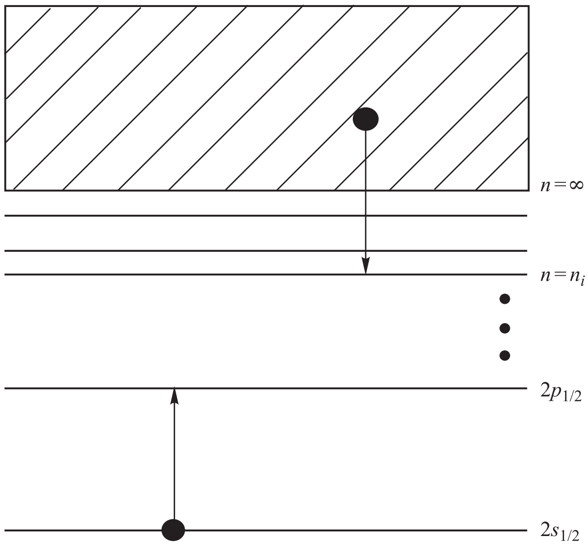

Fig. 1. General scheme of the di-electronic recombination (DR) process

this was not yet detected in real irradiations, either in EBIT [4], or at the recoil stopping scheme [2].

In the present work, we describe the case of ^{93m}Mo isomer as a candidate for depletion via stopping in gas. This isomer is attractive for NEEC because the potential intermediate level is located only at 4.8 keV above the isomer, similarly as in ^{242m}Am case, but the released energy past depletion is much higher — of about 2.42 MeV, i.e., by 45 times higher as compared to the ^{242m}Am case. The ^{93m}Mo isomer was mentioned earlier in the context of possible experiments using the EBIT trap [4]. But newer scheme of depletion under stopping must be developed now for this nucleus.

For swift ion, an electron capture resulted in NEEC, is an analogy of the known atomic process — di-electronic recombination DR. Very simplified illustration of DR is shown in Fig. 1. NEEC and DR processes are similar in general scenario. The only difference is that the energy released in electron capture is spent for nuclear excitation at the case of NEEC, but for excitation of additional electron to higher orbit due to DR, as is shown in Fig. 1.

1. RESONANCE CONDITIONS FOR NEEC IN A COURSE OF ION STOPPING

The currently proposed experimental scheme for detection of NEEC is displayed in Fig. 2. The recoil nucleus transmits through the gas degrader and its

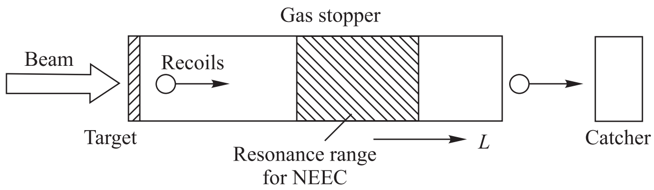

Fig. 2. Layout of a setup for excitation of the reaction-recoil products due to NEEC by stopping

velocity is systematically decreasing. At some distance, the resonance conditions for NEEC are reached, and it is shown as a resonance range within the gas stopper. The resonance coordinate distribution might be wide due to the velocity spread and possible variation of the charge state proper for definite NEEC transition. For instance, in the case of ^{242m}Am, NEEC to the $5p_{3/2}$ orbital in $42+$ charged Am ion may happen at another velocity in the higher-charged states where this orbital remains vacant.

Resonance conditions are schematically explained in Figs. 3, *a–c*. When ion is retarded in the gas absorber, its velocity is reducing and simultaneously the equilibrium charge is also decreased. Respectively, the kinetic energy of bombarding electron in the ion rest frame E_{kin} is diminished. The electron binding energy B_e is also decreased because the charge is reduced as a function of the velocity $q(v)$. A resonance takes place when the total energy of recombination $E_e = (E_{\mathrm{kin}} + B_e)$ becomes equal to the energy of the excited nuclear level E_n. The velocity v and $q(v)$ functions are shown in Fig. 3, *a*, the kinetic, potential and total energies of an electron — in Fig. 3, *b*, and resonance coordinate distribution — in Fig.3, *c*, all as a function of the stopping range. The resonance conditions may even appear multiple times for a single ion because of charge state fluctuations up and down in regular charge-exchange interactions of an ion with medium.

A nuclear radiation detector should be installed near the «resonance range» coordinate to measure prompt emission accompanying events of the isomer depletion by NEEC. On today, no experimental attempts are known aimed to explore the survival of isomers under stopping of the reaction products. The depletion rate could be negligible for many isomers, yet manifested at some special cases. The scheme is advantageous because each ion at some point in the degrader will obtain a resonance velocity required for NEEC, and the velocity spread does not cancel that.

An object for concrete experiment (within a stopping scheme) could be selected among the following isomers: ^{242m}Am, or ^{93m}Mo. Both could be produced in a gas target irradiated by accelerated heavy ions: ^{2}H (^{241}Pu, n) and ^{4}He (^{91}Zr, $2n$). The isomeric recoils possess enough velocity for NEEC reso-

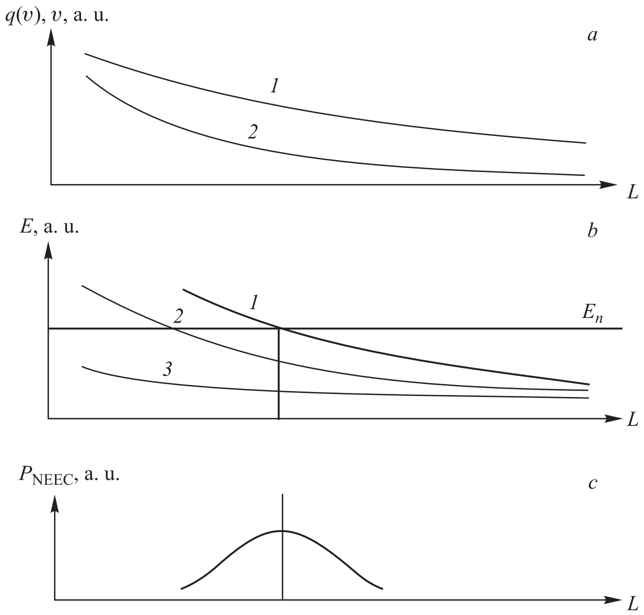

Fig. 3. Schematic illustration of the following parameters dependent on the stopping range L: *a*) of the ion velocity v and of equilibrium charge $q(v)$ shown by curves *1* and *2*, respectively; *b*) of the electron capture sum energy E_e (curve *1*), of the electron binding energy B_e (curve *2*), and of the electron kinetic energy $E_{\rm kin}$ in the ion rest frame (curve *3*); *c*) resonance in the NEEC probability due to matching of E_e energy and of the nuclear-transition energy E_n

Resonance parameters for nuclear excitation by NEEC

Reaction	Product nuclide		Activation level	Energy above isomer	Atomic state		Resonance energy, MeV/u	Electron impact energy
	Isomer	Spin, parity			Charge	Vacancy		
^{4}He (^{91}Zr, $2n$)	^{93m}Mo	21/2$^+$	17/2$^+$	4.8 keV	36+	$3p_{3/2}$	4.91	2.67 keV
^{2}H (^{241}Pu, n)	^{242m}Am	5$^-$	3$^-$	4.1 keV	42+	$5p_{3/2}$	4.89	2.66 keV

nance under stopping in the gas absorber. In the Table, resonance parameters are given for definite nuclear and atomic transitions in these cases. Such conditions provide NEEC in greatly stripped ions of the recoiled isomeric species. The level schemes of both nuclei are shown in Fig. 4, they look in principle similar, but the rate of nuclear transitions and other characteristics are different.

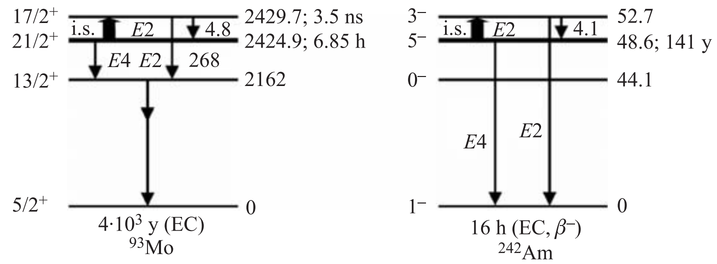

Fig. 4. Nuclear level schemes for ^{93}Mo and ^{242}Am according to Nuclear Data Sheets. The level and transition energies are given in keV

Let us give more details for the ^{93m}Mo case. The resonance matching is needed between the projectile energies required for the nuclear reaction and for the proper electron-collision energy at NEEC. The excitation function of the producer ^{4}He (^{91}Zr, $2n$) reaction is peaked near 6 MeV/u for Zr ion energy in l.s. The corresponding energy of the isomer recoil — 5.5 MeV/u exceeds the NEEC resonance value of 4.9 MeV/u. However, the produced recoil nucleus reaches the required resonance velocity due to energy loss in the gas absorber. The charge-state distribution is also wide, as is known from [1], and a 6% decrease in the ion velocity does not exclude the 36+ charge-state required for NEEC according the Table.

The satisfactory resonance conditions could also be found for the 35+ charge-state and for others nearby. The wide nuclear excitation function — with half-width of about 1.5 MeV/u, together with wide charge distribution of ions create some freedom for resonance matching. Regular electron capture and loss processes generate many options for NEEC within some range of ion energy under stopping. Thus, the discussed case of ^{93m}Mo isomer depletion is one of the best options because the requirements of different kinds could be satisfied, including the production yield, the isomer-depletion energy matched to the NEEC resonance, and the charge-state distribution of ions.

The successful detection of NEEC process is restricted by presence of the backgrounds, the same as in the sophisticated experiments with a storage ring, or with EBIT trap. Assume that in some hypothetical case, the isomer depletion probability for each stopped ion is as high as tens per cent. Then, the activation method could be applied to isolate this probability by comparison of the yields of the same reaction, but performed in the direct and inverse kinematical scheme [2]. For instance, it is possible to irradiate a heavy target by deuterons, or by ^{4}He ions collecting the produced activity and to compare that with the yield of the same isomer in the irradiation of the deuterium, or He gas targets by the heavy element ions. If the isomer depletion probability is much lower unity, then the activation method is not applicable and only the direct detection of prompt gamma radiation past depletion is possible.

It was mentioned above that regular charge-exchange events happen under stopping of an ion in matter. This looks positive for NEEC because of a high cross section of electron capture. However, that means also a high yield of radiative recombination RR photons, and correspondingly, high background for NEEC detection. Past excitation of a nucleus from a ground state to some excited level, the nuclear and recombination photons will possess practically the same energy, and a great physical background is created. Another situation is typical for isomer excitation by NEEC, the depletion photons are emitted with higher energy than the recombination quanta because the initial energy of an isomeric state contributes additively to the energy of a nuclear transition from the intermediate level to the ground state. Thus, the experiment for NEEC with isomer is advantageous even in the version of prompt radiation detection, not only within a scheme of the activation method.

2. ESTIMATED RATE OF ^{93m}Mo ISOMER DEPLETION AT STOPPING

Many years ago, the nuclear excitation by NEEC under stopping was discussed in [5, 6]. But that was devoted to other domains of physics as compared to our proposals for depletion of isomers [2, 7]. There were considered in [5] a high-velocity (13–93) MeV/u bare ions of the heavy elements from Ho to Pu those could capture an electron to K, or L shell with excitation of a nuclear level within 50 to 140 keV above the ground state. The evaluated cross section reaches tens millibarn in these examples. The same process under channeling of swift ions may be more promising because channeled ion moves near the channel median line (plane) and collides mostly with weakly bound valence electrons. Therefore, the spectrum of impact kinetic energy becomes relatively narrow that is affected in the enhanced yield of the NEEC resonance. Nevertheless, the rectified theoretical analysis of [6] is resulted in much lower cross sections at similar ion velocities compared to [5] despite expected growth due to channeling in Si crystal. The cross section of about microbarn was deduced in [6].

In the present article and in [2, 7], we describe the depletion of isomers requiring much lower excitation energy < 5 keV for nuclear transition to the intermediate state. Due to that, the NEEC probability could be higher with account also of much lower ion velocity and lower grade of atom ionization in this case, as compared to [6]. The electron stripping and recombination are the multiply repeated processes over the ion range and they take place as many times as $3 \cdot 10^4$ when the velocity is still enough for NEEC. Strictly speaking, one has to create the Monte Carlo transport code and to simulate the consequence of atomic collision processes over the ion trajectory. Such a progress in theory was not yet reported. A successful NEEC experiment also is in expectation for future. The simplified estimate of the ^{93m}Mo depletion probability is described below.

It was mentioned above that NEEC in general properties is similar to the di-electronic recombination DR, only nuclear excitation instead of atomic transition serves to absorb the energy released in recombination. Therefore, it would be possible to estimate NEEC cross section via DR. The latter process was successfully observed as one of the supplementary effects to more general radiative recombination RR process. Both NEEC and DR are the resonant processes. At definite ion velocity, the resonance conditions could be fulfilled for NEEC, but not for DR, and vice versa. However, one can use the $\sigma_{\rm DR}$ cross section as a scale for estimate of $\sigma_{\rm NEEC}$ at comparable velocities. Similarly, the yield values are linked by the expression:

$$Y_{\rm NEEC} = \frac{Y_{\rm DR} \cdot \Gamma_n}{\Gamma_a}. \qquad (1)$$

We reasonably assume that in real experiments, the resonance width will be completely covered due to the modest energy resolution. Therefore, in calculations of the yield $Y \sim \sigma$, the σ values correspond to the total cross section for definite resonance unlike to the peak $d\sigma/dE$ magnitude. Resonance widths are given by Γ symbols in (1), and they define the absolute rate, not only the energy dependence of the processes.

At depletion of ^{93m}Mo by NEEC, the recombination energy must be equal to the nuclear transition energy, means to 4.8 keV. At the case of DR, this energy may be enough to excite an electron from L to M shell in the high-charge state Mo ion. Then, the atomic level width in Eq. (1) effective for DR could be estimated from the known [8, 9] radiative width of the L vacancy being numerically of about 0.1 eV in Mo atom. However, this corresponds to the neutral atom unlike to our case when radiative transitions of unbound electrons to the vacancy could happen because of almost completely stripped atomic shells in the ion. Then, Γ_a in Eq. (1) must be replaced by $\Gamma_a^{\rm eff} \approx 0.01$ eV.

For nuclear activation level at 2429.7 keV, the half-life $T_{1/2} = 3.5$ ns corresponds to a width $\Gamma = 1.7 \cdot 10^{-7}$ eV. That is defined by the 268 keV $E2$ transition to the level located at 2161 keV. But for NEEC we are concerned by the transition of low energy — 4.8 keV back to the isomer at 2424.9 keV. At the same multipolarity, the γ-decay width is reduced by many orders of magnitude.

Furthermore, it was already mentioned that NEEC is an inverse process to electron conversion (IC), and the estimated width for NEEC must include the width of inverse process, i.e., of IC. The conversion coefficient for the 4.8 keV $E2$ nuclear transition could be calculated using the code described in [10]. The conversion from $M_{\rm III}$ subshell must be specified. In highly stripped ion the remaining electrons are strongly bound and $M_{\rm III}$ orbit takes the location replacing $L_{\rm III}$ orbit. Therefore, the effective for NEEC nuclear width was calculated using the $L_{\rm III}$ — conversion coefficient equal $2.6 \cdot 10^5$ according [10]. It was finally obtained $\Gamma_n^{\rm eff} = 0.8 \cdot 10^{-10}$ eV. Thus, effective values of widths for substitution to Eq. (1) are ready, remains only to define reasonably the $Y_{\rm DR}$ magnitude.

The ion-velocity active range corresponds to the kinetic energy gap from 5.5 to 4 MeV/u, that is the stopping range of 15 cm in He gas at normal conditions. Many electron-capture and electron-loss events happen over this range. The cross section of about $3 \cdot 10^{-17}$ cm^2 for elementary electron collision could be deduced for RR basing on the Mo ion stopping power [11]. Combining that with the He gas layer thickness of $4 \cdot 10^{20}$ atoms/cm^2, one obtains immediately $2.5 \cdot 10^4$ charge exchange collisions. Some of them will be manifested in a form of DR and some are accompanied by NEEC. The resonance for NEEC may appear for different charge states at corresponding velocities. The stopping from 5.5 to 4 MeV/u involves 15% regular decrease of the ion velocity and may cover many NEEC resonances corresponded to different ionization states of the swift ion. Even some definite charge of an ion may appear several times due to fluctuations. Electron states in medium may also be perturbed by the wake potential and because of multiple scattering by attractive Coulomb center. Capture of electrons from perturbed states allows additional options for the NEEC resonance.

Charge state distribution given in [1] supports an assumption that 6 different charges near the equilibrium charge are involved in the interplay of ion charge fluctuations. They are potentially active for NEEC. The resonances will take place for each ion and not as a single event. The experimental energy resolution makes no significance because each ion transmits through the resonances along regular decrease of the velocity. Similar scheme is realistic also for DR. The di-electronic recombination yield is assumed at a level 20% of the total RR yield, i.e., $Y_{\rm DR} = 5 \cdot 10^3$ for each ion under stopping from 5.5 to 4 MeV/u in He gas. Combining all together in Eq. (1), one deduces the yield of NEEC at a magnitude of 10^{-4}, i.e., each isomeric ^{93m}Mo nucleus has a chance for depletion with a probability of about $10^{-2}\%$.

This is a moderately low yield, but one has to involve the isomer production rate as high as of 10^5 s^{-1} with the standard cross section assumed for the $(\alpha, 2n)$ reaction. Then, one deduces a great effect of the isomer depletion by NEEC. In the rate estimate, we took a relatively low beam current nearby 1 particle nA. At higher intensities, the yield of isomer is growing proportionally, but an intense beam may create plasma in the gas stopper. That generates some uncertainties difficult for accounting. Plasma could disturb the resonance processes, or oppositely, may produce a positive influence. In [12], there was predicted that stellar plasma at temperature above $5 \cdot 10^6$ K provides acceleration of the ^{93m}Mo isomer decay by a factor of 10^6 due to NEEC. In our scheme, the bombardment by 2.7 keV electrons is equivalent to the effective temperature of $3 \cdot 10^7$ K and the NEEC rate must be enhanced even stronger.

Our estimates above are yet inaccurate because some parameters are taken only within the order of magnitude. The NEEC yield could be significantly higher than $10^{-2}\%$. At a high NEEC probability, the background-less method could be applied for detection of the process. As is proposed in [2, 7], the ^{93m}Mo recoils must be collected at solid catcher past transmission through the He gas volume. The yield of reaction products in this inverse kinematics should be compared to the direct scheme when ^{4}He ions impact the ^{91}Zr target. The ratio of yields contains a NEEC probability and the backgrounds are excluded in this scheme. The activation method is applicable only when $P_{\mathrm{NEEC}} \geqslant 0.05$, otherwise inaccuracies do not allow a successful measurement of the yield ratio taken in the direct to inverse kinematics.

At low probability, the NEEC radiation must be detected «in-beam» because the activation method becomes insensitive. The in-beam experiment is more complicated due to the presence of γ radiation of different origins. In literature, there were discussed the backgrounds created by Coulomb excitation of nuclei, or by radiative recombination of electrons. Positive aspect of the experiment with isomer is manifested in the special energy of photons emitted past NEEC that is not identical to the background γ lines of CE and RR radiation. Thus, backgrounds are suppressed also in the scheme of direct in-beam NEEC detection. The latter scheme requires nevertheless the best experimental equipment at a top level of modern possibilities.

SUMMARY

Isomer depletion under stopping of nuclear-reaction products in a gas absorber looks attractive for exploration of stimulated isomer decay. Relatively straightforward experiments could be carried out using common technique without great complications. Such options make significance also for discovery of the atomic-nuclear cooperative NEEC process. Low background conditions could be created for NEEC detection at the case of isomeric species excitation by electron capture. The yield of ^{93m}Mo isomer depletion was estimated using atomic data for charge-state distributions and for stopping power of swift heavy ions in matter. The moderate probability of NEEC combined with a standard nuclear-reaction rate promises relatively high yield of isomer depletion.

REFERENCES

1. *Shima K., Kuno N., Yamanouchi M., Tawara A.* // At. Data Nucl. Data Tables. 1992. V. 51. P. 173.
2. *Karamian S. A., Carroll J. J.* // Laser Phys. 2010. V. 20. P. 977.
3. *Palffy A., Evers J., Keitel C. H.* // Phys. Rev. Lett. 2007. V. 99. P. 172502.
4. *Bernstein L., Bleuel D., Marrs R. et al.* // Proc. of the ECT Workshop on Atomic Effects in Nuclear Excitation and Decay, Trento, Italy, 2009. http://www.ect.it

5. *Cue N., Poizat J. C., Remilliex J.* // Europhys. Lett. 1989. V. 8. P. 19.
6. *Yuan Z. S., Kimball J.* // Phys. Rev. C. 1993. V. 47. P. 323.
7. *Karamian S. A., Carroll J. J.* // Proc of Intern. Conf. on Current Problems in Nuclear and Atomic Physics. Kiev, Ukraine, 2011. P. 181.
8. *Keski-Rahkonen O., Krause M. O.* // At. Data Nucl. Data Tables. 1974. V. 14. P. 139.
9. *Campbell J. L., Papp T.* // At. Data Nucl. Data Tables. 2001. V. 77. P. 1.
10. *Kibedi T., Burrows T. W., Trzhaskovskaya M. B. et al.* // Nucl. Instr. Meth. A. 2008. V. 589. P. 202.
11. *Ziegler J. F.* Stopping Cross Sections for Energetic Ions in All Materials. Handbook, V. 5. N. Y.: Pergamon Press, 1980.
12. *Gosselin G., Morel P.* // Phys. Rev. C. 2004. V. 70. P. 064603.

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141980, г. Дубна, Московская обл., ул. Жолио-Кюри, 6.
E-mail: publish@jinr.ru
www.jinr.ru/publish/