NEUTRON CAPTURE RATES OF LIGHT ISOTOPES FOR INHOMOGENEOUS BIG BANG NUCLEOSYNTHESIS*

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The quantitative analysis of scenarios of nonstandard big bang nucleosynthesis requires the detailed knowledge of capture reaction rates on light isotopes, especially for the build-up of nuclei with $A \geq 12$. With a new setup for fast cyclic activation, capture reaction rates of a variety of isotopes have been measured, especially for the nuclei ^{22}Ne, ^{14}C and ^{18}O. The technique was checked on isotopes of Kr, Ag, Xe and Pt, isomeric ratios were determined and some of the cross sections served as cross section standard for the measurements on ^{22}Ne and ^{18}O.

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1. Introduction

The analysis of the inhomogeneous big bang nucleosynthesis requires the determination of the reaction rates of the involved nuclei [1]. In this nonstandard big bang nucleosynthesis nuclei with $A \geq 12$ may be formed *via* reaction sequences like $^{7}\mathrm{Li}(n,\gamma)^{8}\mathrm{Li}(\alpha,n)^{11}\mathrm{B}(n,\gamma)^{12}\mathrm{B}(\beta)^{12}\mathrm{C}(n,\gamma)^{13}\mathrm{C}(n,\gamma)^{14}\mathrm{C}(\alpha,\gamma)^{18}\mathrm{O}(n,\gamma)^{19}\mathrm{O}(\beta)^{19}\mathrm{F}$. The capture reactions of the relevant stable or long-lived isotopes on these pathways can be studied experimentally in spite of the small size of the cross sections. An experimental method, the fast cyclic activation techniques [2–6], has been developed at the Karlsruhe 3.75 MV Van de Graaff accelerator to measure some of the capture cross sections of these nuclei. This method is an extension of the previous activation technique [7] to the counting of activities with half lives of only a few seconds. Because of its sensitivity and selectivity (number of target

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atoms N times capture cross section $\sigma \approx 0.1$ mol μbarn) it allows for the determination of isomeric cross section ratios.

2. Fast cyclic activation technique

A normal activation measurement is subdivided into two parts: (1) the irradiation of the sample, (2) the counting of the induced activity [7]. The irradiation time of the sample cannot be extended arbitrarily to gain statistics because of the occurrence of a half life dependent saturation effect. This limits the activation measurements to isotopes with half lives greater than about 5 minutes. An irradiation time corresponding to four times the half life generates already 94% of the obtainable activity. In many cases the count rate achieved in such a measurement is statistically not acceptable, and an increase of the neutron flux or the sample amount to improve the situation is normally only a limited remedy for smaller and smaller half lives. The way out of this dilemma, the cyclic activation method, is the repetition of the irradiation and activity counting procedure many times. Especially for nuclei with half lives of only seconds a large number of irradiation and counting cycles is needed. As the characteristic time constants of a cycle have to be adjusted to the short half life, a fast sample changer is also indispensable for the experiment. The time constants for each cycle, adjusted to the decay rate λ of the investigated, isotope are: the irradiation time, t_b, the counting time, t_c, the waiting time, t_w (the time to switch from irradiation to counting phase), and the total time, $T = t_b + t_w + t_c + t'_w$ (t'_w the time to switch from the counting to the irradiation phase). The accumulated number of counts from a total of n cycles, $C = \Sigma_i^n C_i$, where C_i are the counts after the i-th cycle, is calculated for a chosen irradiation time, t_b, which is short enough compared with the fluctuations of the neutron flux [5]:

$$C = \varepsilon_\gamma K_\gamma f_\gamma \frac{1}{\lambda}\left[1 - \mathrm{e}^{-\lambda t_c}\right]\mathrm{e}^{-\lambda t_w}\frac{1-\exp(-\lambda t_b)}{1-\exp(-\lambda T)} N\sigma\Sigma\Phi_i\left\{1 - f_b \mathrm{e}^{-\lambda T}\right\}, \quad (1)$$

with $f_b = \sum_{i=1}^{n} \Phi_i \exp[-(n-i)\lambda T] / \sum_{i=1}^{n} \Phi_i$.

The following additional quantities have been defined: ε_γ: Ge-efficiency, K_γ: γ-ray absorption, f_γ: γ-ray intensity per decay, N: the number of target nuclei, σ: the capture cross section, Φ_i: the neutron flux in the i-th cycle. The quantity f_b is calculated from the registered flux history of a ^{6}Li glass monitor.

The activities of nuclides with half lives of several hours to days can be also counted after the end of the cyclic activation consisting of n cycles.

$$C_n = \varepsilon_\gamma K_\gamma f_\gamma \tfrac{1}{\lambda}[1-\exp(-\lambda T_M)]\exp(-\lambda T_W)[1-\exp(-\lambda t_b)] N\sigma f_b \Sigma\Phi_i\,. \quad (2)$$

Here T_M is the measuring time of the Ge(Li)-detector and T_W is the time elapsed between the end of cyclic activation and beginning of the new data acquisition. With the equations (1) and (2), the accumulated number of counts C and the final counts C_n after several previous cyclic activations with remaining activities can be formulated as well [5]. From Eq. (2) it is also obvious that the common activation technique is a special case of cyclic activation.

Equations (1) and (2), respectively, contain the unknown quantities σ and the total neutron flux $\Sigma\Phi_i$. Therefore, cross section ratios can be formed for different isotopes exposed to the same total neutron flux. This is the basis for the determination of the cross section of an isotope ^{A}Z, if a well-known standard isotope, ^{197}Au, is chosen as a reference material [8]. In general, the sample to be investigated is characterized by a finite thickness so that it is desirable to sandwich the sample by two comparatively thin gold foils for the determination of the effective neutron flux at sample position. These Au foils should be counted individually after termination of the cyclic activation according to equation (2). The effective count rate is calculated from these individual rates.

3. Experimental arrangements

The measurements are carried out at the Karlsruhe pulsed 3.75 MV Van de Graaff accelerator. In Fig. 1 a scheme of the experimental setup is shown. Using the special properties of the ^{7}Li(p,n) reaction near the reaction threshold, a Maxwellian neutron spectrum with a thermal energy $kT = 25$ keV can be generated for the irradiations [5, 7, 8]. The required properties for the ^{7}Li(p,n) reaction are adjusted before the start of the activation. The proton energy conditions are determined in a time-of-flight (TOF) measurement using the accelerator in pulsed mode. The proton beam is wobbled initiated by magnetic deflection to cover the area of the Li target. The beam profile formed is studied on a quartz target. To switch back and forth between sample irradiation and activity counting, a fast sample changer operating with compressed air was used. Close to the beam line, where the neutrons have been generated, the Ge-detector for activity counting well shielded by lead and Li loaded paraffin, has been installed (Fig. 1). During the irradiation phase, the analog to digital converter is gated to prevent data acquisition. The relative neutron flux is recorded continuously with a ^{6}Li glass detector. During the activity counting phase, neutron generation is interrupted by a beam stop for the proton beam. This is essential to reduce all prompt accelerator dependent γ-rays. The beam stop is installed in the beam line at the accelerator hall, so that in the activity counting periods the experimental hall is free of prompt background radiation.

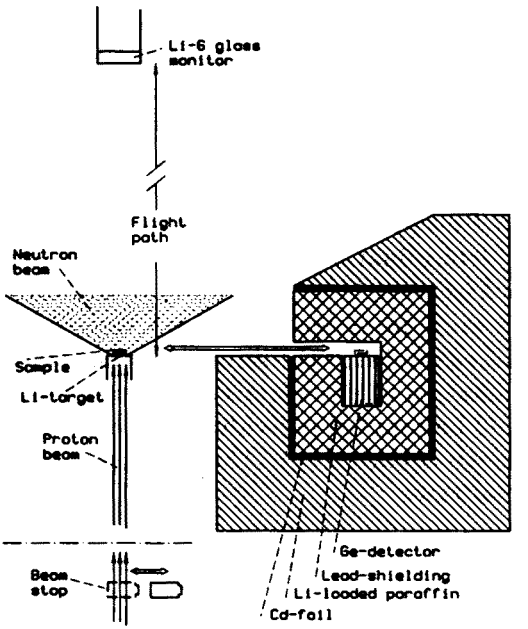

Fig. 1. Scheme of the experimental setup for fast cyclic activation. The irradiation phase is shown. In the counting phase the proton beam is blocked by the beam stop in the accelerator hall, and the sample is switched to the detector position. The positions of the beam stop and the sample are indicated.

4. Measurements

4.1. $^{107,109}Ag(n,\gamma)$

In order to perform test measurements to check the properties of the present setup, natural silver with two isotopes of about equal abundances seems to be a most suitable element. The generated activities of ^{108}Ag and ^{110}Ag have half lives of 2.37 m and 24.6 s, respectively. The activation cross sections are large enough that it is easy to obtain sufficient statistics. The characteristic γ-ray lines lie at convenient energies and the metallic samples are easy to machine. The capture cross sections for ^{107,109}Ag are the cross sections to the respective short lived ground states of ^{108,110}Ag. To obtain the total capture, a less than 10% correction for the cross section to the corresponding long lived isomeric states has to be applied. The results are in reasonable agreement [5].

4.2. Capture cross sections to isomeric states

In an investigation of natural Kr and Xe samples (pressurized gas in stainless steel spheres or Kr loaded zeolite) the cross sections to short lived isomers of Kr and Xe isotopes ($^{79}Kr^m$, $^{81}Kr^m$, $^{85}Kr^m$, $^{125}Xe^m$, $^{127}Xe^m$, $^{129}Xe^m$, $^{131}Xe^m$, $^{135}Xe^m$) were measured [2]. As in some cases also the capture cross section to the respective ground state was determined, isomeric ratios could be formed and compared with corresponding isomeric ratios at 25.3 meV. The relative constancy of the isomeric ratios as a function of energy formed a basis to estimate the total capture cross sections of the pure *s*-process nuclides ^{128}Xe and ^{130}Xe [2] from the measurements of the cross sections to the isomeric states. The Kr measurements also served to carefully determine the capture cross section of ^{86}Kr *vs* the ^{197}Au standard, so that in later measurements Kr could be used as a second standard for pressurized gas mixtures.

Isomeric ratio measurements were also carried on Pt. The $^{196}Pt(n,\gamma)$ $^{197}Pt^{m+o}$ reaction yielded for $kT = 25$ keV: $\sigma^m = 14.0 \pm 1.5$ mbarn, $\sigma^{m+o} = 216 \pm 25$ mbarn, the $^{198}Pt(n,\gamma)^{199}Pt^{m+o}$ reaction: $\sigma^m = 2.95 \pm 0.18$ mbarn, $\sigma^{m+o} = 90 \pm 13$ mbarn. For ^{194}Pt only the cross section to the isomer in ^{195}Pt at $kT = 25$ keV was determined as $\sigma^m = 30 \pm 2$ mbarn, but a total cross section $\sigma^{m+o} = 400 \pm 90$ mbarn estimated. In spite of the small abundance of ^{190}Pt, the total capture cross section at $kT = 25$ keV could be measured to be 742 ± 200 mbarn, demonstrating the abilities of the method. The systematic trends of the total capture cross sections of the even-even Pt isotopes suggest for the pure *s*-process isotope ^{192}Pt a capture cross section at $kT = 25$ keV of 570 ± 130 mbarn.

4.3. $^{22}Ne(n,\gamma)Ne$

After a Hauser–Feshbach calculation [9] and a measurement [10] has shown that the $^{22}Ne(n,\gamma)$ cross section could easily be as big as 900 μbarn, *s*-process scenarios, based on the $^{22}Ne(\alpha,n)$ reaction as neutron source, suffered from the neutron poisoning effect of the $^{22}Ne(n,\gamma)$ reaction. The cyclic activation measurement of $^{22}Ne(n,\gamma)$ using pressurized Kr–^{22}Ne gas mixtures solved this problem. A capture cross section for ^{22}Ne of only 66 ± 5 μbarn [3] was found at $kT = 25$ keV, which restores the expected behaviour of the ^{22}Ne neutron source in the stellar models. Our result is also in good agreement with calculations applying the direct capture mechanism [11] which in this case appears to be dominant compared with compound capture. The $^{22}Ne(n,\gamma)$ cross section is also of relevance for the analysis of inhomogeneous big bang nucleosynthesis.

4.4. $^{14}C(n,\gamma)$, $^{18}O(n,\gamma)$ and $^{15}N(n,\gamma)$

For the synthesis of nuclei heavier than $A \geq 12$ in nonstandard big bang scenarios ^{14}C may act as an important bottle neck. Because of a high available neutron density, $^{14}C(n,\gamma)$ is expected to compete strongly with the alpha, proton and deuteron reactions, triggering a sequence of reactions to heavier isotopes [12, 13]. The $^{14}C(n,\gamma)$ measurement [4], however, resulted in a value by a factor of 5 [12] or 100 [13] lower than expected ($\sigma(kT = 23.3\ \text{keV}) = 1.1 \pm 0.28\ \mu\text{barn}$), so that the breakthrough to higher mass nuclei occurs mainly *via* $^{14}C(\alpha,\gamma)^{18}O$. It turns out that further build-up is dominated by (n,γ) reactions and beta decays [1]. In our measurement (Fig. 2) we found for the $^{18}O(n,\gamma)$ reaction $\sigma(kT = 23.3\ \text{keV}) = 8 \pm 1\ \mu\text{barn}$ [6]. This relatively small cross section limits the production of heavier isotopes with $A \geq 20$. For the flow *via* $^{14}C(n,\gamma)^{15}C(\beta)^{15}N(n,\gamma)^{16}N(\beta)^{16}O(n,\gamma)$ the $^{15}N(n,\gamma)$ reaction has been measured recently and the analysis is under way. Both measurements, $^{18}O(n,\gamma)$ and $^{15}N(n,\gamma)$, were performed using as samples pressurized gas mixtures of Kr–^{18}O, and Kr–^{15}N, respectively.

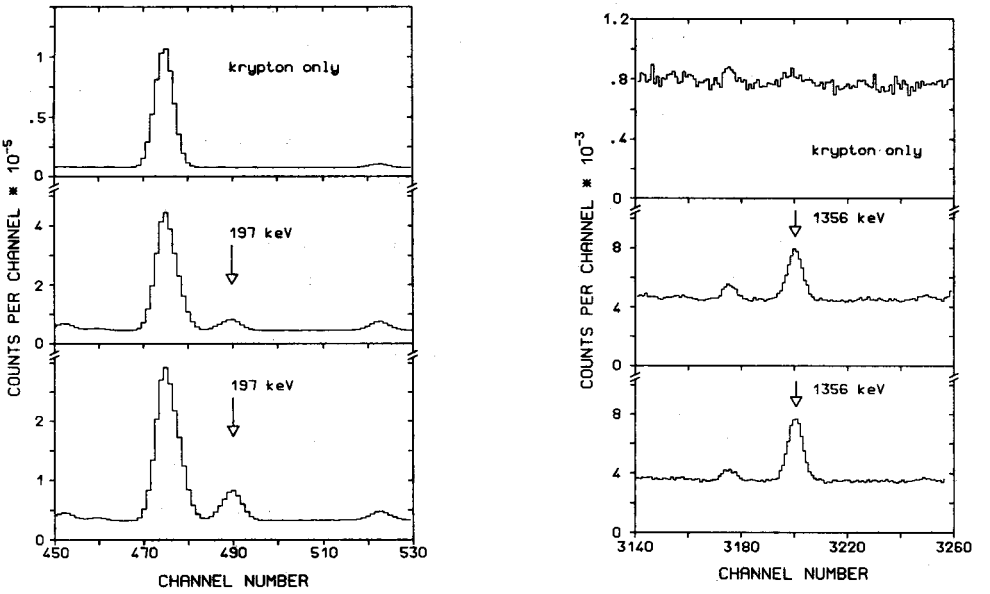

Fig. 2. Accumulated γ-line intensities of the measurements with samples of two different Kr-^{18}O gas mixtures and Kr gas only.

5. Conclusions

Fast cyclic activation can be applied to the measurement of cross sections where product activities with half lives from a few seconds to many

days are involved. In the measurements, where the very short residual activities occur, the optimum of sensitivity of the method is reached, because all radioactive nuclei produced in the capture reaction are with a short time available for detection. This leads to an optimum signal to background ratio. The longer the half life, the longer the counting time to cover a significant part of the activity. In the experiments [3, 4, 6] it was shown that cross sections of a few μbarn can be measured. The product of a number of target nuclei times capture cross section characterizes the possibilities of the setup. From the above mentioned measurements we derive for this quantity 0.1 mol·μbarn. This means either the possibility to measure a μbarn cross section with several hundred mg target material or to measure a barn cross section with a μg target. Further examples to apply the fast cyclic activation technique for light isotopes are $^{10}\mathrm{Be}(n,\gamma)^{11}\mathrm{Be}$ (13.8 s), $^{19}\mathrm{F}(n,\gamma)^{20}\mathrm{F}$ (11 s), $^{26}\mathrm{Mg}(n,\gamma)^{27}\mathrm{Mg}$ (9.46 m) and $^{36}\mathrm{S}(n,\gamma)^{37}\mathrm{S}$(5 m). From these cases a ^{36}S measurement to study the s-process synthesis of this rare isotope will be performed next.

The present activation technique cannot, of course, account for reactions where the product nucleus is stable. One must use then the time-of-flight technique and record the prompt emitted γ-ray lines. The primary γ-ray lines of the $^{7}\mathrm{Li}(n,\gamma)$ and $^{12}\mathrm{C}(n,\gamma)$ reactions were measured by Nagai *et al.* [14, 15], and of the 434 keV p-wave resonance of $^{16}\mathrm{O}(n,\gamma)$ by Igashira *et al.* [16], with a Van de Graaff accelerator using the $^{7}\mathrm{Li}(p,n)$ reaction for neutron production and a setup with short flight path (150 mm) and well shielded γ-ray detector. The γ detection system applied was an anti-Compton NaI(Tl) detector but, to improve the system, a Ge-crystal for γ-ray energy detection [17] should be tried because of the much better energy resolution.

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