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AUTHOR(S): Paul E. Koehler, Harold A. O'Brien, and Charles D. Bowman, P-3

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Los Alamos National Laboratory
Los Alamos, New Mexico 87545

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MEASUREMENTS OF ASTROPHYSICAL REACTION RATES FOR RADIOACTIVE SAMPLES*

P.E. Koehler, H.A. O'Brien and C.D. Bowman
Physics Division, Los Alamos National Laboratory, MS-D449,
Los Alamos, New Mexico 87545, USA

ABSTRACT

Reaction rates for both big-bang and stellar nucleosynthesis can be obtained from the measurement of (n,p) and (n,γ) cross sections for radioactive nuclei. In the past, large backgrounds associated with the sample activity limited these types of measurements to radioisotopes with very long half lives. The advent of the low-energy, high-intensity neutron source at the Los Alamos Neutron Scattering CEnter (LANSCE) has greatly increased the number of nuclei which can be studied. Results of (n,p) measurements on samples with half lives as short as fifty-three days will be given. The astrophysics to be learned from these data will be discussed. Additional difficulties are encountered when making (n,γ) rather than (n,p) measurements. However, with a properly designed detector, and with the high peak neutron intensities now available, (n,γ) measurements can be made for nuclei with half lives as short as several weeks. Progress on the Los Alamos (n,γ) cross-section measurement program for radioactive samples will be discussed.

INTRODUCTION

Over the years, many reaction rates of importance to both big-bang and stellar nucleosynthesis calculations have been measured in the laboratory. These rates have then been integrated into the reaction network used in the calculations and have improved the general understanding of several types of nucleosynthesis events. At present, the rates for several important reactions have not been measured, necessitating the use of theoretical estimates[1]. This may lead to large uncertainties in the isotopic yields from nucleosynthesis calculations. Many of the unmeasured rates involve neutron-induced reactions on radioactive nuclei. Previous measurements[2,3] of this type have been limited to nuclei with very long half lives due to potentially large backgrounds associated with the sample activity. The advent of pulsed spallation neutron sources, such as LANSCE[4], has opened up the possibility of making cross-section measurements for neutron-induced reactions on nuclei with short half lives. In this paper, we will give some examples of recent measurements of this type and briefly discuss the nuclear astrophysics to be learned. We will then discuss our plans for additional measurements, briefly outlining the techniques involved and the expected results. We expect that these measurements will greatly aid in the understanding of the astrophysical environments in which nucleosynthesis occurs, and will aid in the calculation of the expected nucleosynthesis yield of isotopes of interest to gamma-ray astronomy.

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EXPERIMENTAL TECHNIQUE

The experimental technique[5] for these measurements requires a large peak neutron intensity and a properly designed detector so that the detected rate for the reaction of interest is larger than the background rate associated with the decay of the sample under study. At LANSCE, the high peak neutron intensity is obtained by bombarding a tungsten target with an intense burst of protons. The protons are accelerated by LAMPF, and compressed into an intense pulse by the newly commissioned Proton Storage Ring (PSR). At the design intensity of the PSR (100 μA, at 12 Hz), the water moderated neutron intensity at 1 eV for a flight path of 7 m is 4×10^6 neutrons/(eV cm^2 sec), and the neutron spectrum of this "white" source is approximately proportional to $1/E_n$. The relatively long pulse width (250 ns) from the PSR limits the useful upper energy to about 50 keV at which point the energy resolution is about 25% for the 7 m flight path used in our measurements. The "white" nature of the neutron source means that measurements at all neutron energies are obtained simultaneously. This high neutron intensity allows measurements to be made with sample sizes in the 100 ng to a few hundred μg range. The small sample size means that the necessary radioactive samples are easier to produce and that only relatively modest activities must be handled. Even these small samples can still present some rather large background problems, but a properly designed detector can reduce the sample-related backgrounds to acceptable levels. Because the requirements for the detectors differ, we will discuss (n,p) and (n,γ) measurements separately below.

A^*(n,p) MEASUREMENTS

For A^*(n,p) measurements, where A^* is a radioactive nucleus, the sample-related backgrounds can be reduced to manageable levels by choosing a charged-particle detector of thickness no greater than that needed to stop the protons from the reaction of interest. The detection efficiency for radioactive decay emissions from the sample can thus be reduced to order 10^{-6} of the proton detection efficiency. Also, very few nuclei emit charged particles under bombardment by slow neutrons. Hence, the sample can be of relatively low specific activity, and can even be a chemical compound. Several unstable nuclei (^{22}Na, ^{26}Al, ^{56}Co) have been considered as candidates for observation by gamma-ray telescopes[6], and at least one[7] (^{26}Al) has been detected. Perhaps at this workshop we will hear of other successful observations. In certain astrophysical scenarios, A^*(n,p) reactions play an important role in the calculations of the production of these isotopes. For example, the relatively large ^{26}Al(n,p)^{26}Mg cross section is given as evidence that ^{26}Al may not be produced in sites where neutrons are important.[8] Also, some A^*(n,p) reactions may play an important role in the nucleosynthesis in explosive environments of very rare stable isotopes.[9]

We will discuss ^{7}Be(n,p)^{7}Li ($t_{1/2}$=53 days) as a first example of an important A^*(n,p) reaction rate. In this case, the reaction is of importance to the big-bang nucleosynthesis of ^{7}Li. Our measurements were made with 90 ng (≈30 mCi) of ^{7}Be, in less than one week of beam time with the PSR operating at only one-tenth of its design intensity. Prior to our measurements[5], only the thermal cross sections had been measured directly[10]. The rate[12] for this reaction used in calculations was based on this rather imprecise thermal value and on some also rather imprecise ^{7}Li(p,n)^{7}Be measurements[11] converted to (n,p) using detailed balance. Our new measurements have substantially reduced (by a factor of almost ten at thermal energy) the uncertainty in the reaction rate. The reaction rate determined primarily from our new data is compared to the old rate in fig. 1. The rate based on our data is only 60% to 80% of the old rate in the temperature range of interest[13] in the big-bang calculations ($T_9 \approx 0.3 - 1$). Calculations[14-16] have shown that this difference can lead to as much as a 20% increase in the amount of ^{7}Li produced in the big bang.

A second example of our recent $A^*(n,p)$ measurements[17] is ^{22}Na(n,p)^{22}Ne ($t_{1/2}$=2.6 years). In this case, the measurements were made with 360 ng (≈2.25 mCi) of ^{22}Na. This reaction may play a role in the nucleosynthesis of ^{22}Na and ^{22}Ne, and may aid in the interpretation of the neon-E anomaly[18] in meteorites. The astrophysical reaction rate calculated from our data is compared to the theoretical rate[1] in fig. 2. At our energies, most of the rate is due to protons emitted to the first excited state of ^{22}Ne. The theoretical rate is about a factor of ten lower than the experimentally determined one at very low temperatures. However, due to a resonance at E_n=170 eV, the two rates cross at T_9=0.05. We are currently exploring the astrophysical implications of this result.

A final example of $A^*(n,p)$ measurements is our recent data on the ^{36}Cl(n,p)^{36}S reaction[17]. These measurements were made with 410 μg (9 μCi) of ^{36}Cl. Because the half life for this sample is long ($t_{1/2}$=3x10^5 years), a high peak neutron intensity is not essential to the measurements. However, the relatively high average neutron intensity available from LANSCE is still important to measuring this comparatively small cross section within a reasonable time. Our preliminary measurements are displayed in fig. 3. Because the thermal cross section has not yet been measured, we display yields rather than cross sections. The data reveal several resonances for E_n>900 eV. This reaction is denoted by a * in Howard et al[9], a mark which they reserve for rates important to the nucleosynthesis of rare nuclei (^{36}S in this case) in explosive carbon burning. It remains to be seen how our measurements will affect the results of future calculations.

$A^*(n,\gamma)$ MEASUREMENTS

Because many nuclei have sizable (n,γ) cross sections at low neutron energy, $A^*(n,\gamma)$ measurements require a separated isotope on a low mass backing as a sample. This requirement should not be too difficult to meet for the very small samples sizes required. For these types of measurements the decay radiation from the sample is often of the same type as that from the reaction of interest. Pileup of the low-energy decay γ-rays can result in a signal the same size as that from a neutron-capture event. To overcome this potentially large background one can make use of the fact that the gamma-ray decay energy, E_d, is almost always much lower in energy than the total energy, E_c, of the neutron capture cascade. Hence, a detector which registers all of the energy from the capture cascade, and which has a very short output pulse width ,τ, can effectively overcome this background. Of course, the size of this background is a very strong function of the ratio, E_d/E_c, and of τ, and one can always think of very difficult cases for which measurements are still not possible. Our calculations show that measurements many on interesting samples with half lives as short as a few weeks can be made. A few of these cases[19] are summarized in table 1. One other source of potentially serious background is from neutrons scattered into the detector and subsequently captured by one of the detector nuclei. This could conceivably result in a signal indistinguishable from the reaction of interest.

One obvious choice for a detector is a large tank of liquid scintillator such as that used in previous measurements on stable samples. The tank would be almost 100% efficient for the capture cascade, and by loading the liquid with ^{10}B it can be made much less sensitive to background neutrons. However, we feel that the relatively new scintillator barium fluoride (BaF_2) is a better choice. Monte Carlo calculations we have made using the computer code CYLTRAN[20] indicate that a thickness of roughly 15 cm of BaF_2 is adequate to make an approximately 100% efficient detector in essential agreement with the calculations of Wisshak et al.[21] In our application, the small sample size allows us to have a very small neutron beam (0.5 cm diameter). Hence, a 30 cm cube of BaF_2 with a 4 cm beam hole is sufficient to make a detector which is approximately 100% efficient. A detector of this size, composed of eight 15 cm cubes is relatively inexpensive to build compared to more complex designs such as the Karlsruhe sphere[22]. The fast component of BaF_2 ($\tau \approx 20$ ns for 15 cm crystals) provides for effective pileup rejection. Pileup of the slow component of BaF_2 contributes essentially only a constant offset signal which can be cancelled with the proper electronics.

A BaF_2 scintillator has several advantages over a large liquid scintillator tank. First, the total detector size is much smaller so that shielding from room background is much easier to accomplish. Second, the output pulse width can probably be made shorter for BaF_2 than for a tank of liquid scintillator of the required size ($\approx$1.5 m diameter). Third, the pulse-height resolution of BaF_2 can be varied by accepting more or less of the signal from the slow decay component. When using only the fast component, the expected resolution should be about equal to that obtained with a large tank of liquid scintillator (20-30%). However, for those stable samples which are available only in very small quantities as separated isotopes, or for radioactive samples of very low specific γ-ray activity, the opportunity exists with BaF_2 for improving the pulse-height resolution dramatically and, hence, for improving the accuracy of the measurement, perhaps to as good as 1% (ref. 21). The main disadvantage of BaF_2 is its greater sensitivity to the background from scattered neutrons. We have made Monte Carlo calculations using the code MCNP[23] to estimate the size of this background. The calculations indicate that our detector would be about 1% efficient to scattered neutrons at 10 keV. This can be reduced even further (to 0.3%) by lining the central beam hole with a 1 cm thick layer of $^{10}B_4C$. Hence, for almost all cases the background due to scattered neutrons can be reduced to acceptable levels for the range of energies possible in our measurements.

The resulting $A^*(n,\gamma)$ measurements will have a large impact on our understanding of the dynamics of s-process nucleosynthesis - a point where current models fail to reproduce the observations.[24] The first few measurements at selected s-process branch points should help reveal more about the mean properties of the s-process environment, the competition between radioactive decay and neutron capture helping to place more stringent limits on quantities such as the mean neutron density.[25] A larger series of measurements will allow an examination of the dynamics of the s-process. For example, the measurements will allow for a better understanding of the time dependence of the neutron flux and the peak neutron intensity. We hope that this in turn will spur the development of better models of the s-process environment.

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TABLE 1

A list of a few of the many isotopes of importance to the s-process of nucleosynthesis for which $A^{*}(n,\gamma)$ measurements should be possible at LANSCE.

Isotope	Half life	Motivation
^{85}Kr	10.7 y	Very important branch point.
$^{115}Cd^{m}$	44.8 d	Causes some of mass flow to bypass s-only ^{116}Cd.
^{151}Sm	93 y	Branch responsible for ^{152}Gd. Half life depends on temperature.
^{152}Eu	13 y	Same branching as ^{151}Sm.
^{154}Eu	8.8 y	Part of branch causing mass flow to bypass s-only ^{154}Gd. Half life depends on temperature.
^{155}Eu	5 y	Important branch point.
^{153}Gd	241 d	Same branching as ^{151}Sm.
^{163}Ho	33 y	Branch at ^{163}Dy, ^{164}Er abundance, and yields estimate for matter density.
^{170}Tm	129 d	Causes mass flow to bypass ^{170}Yb.
^{171}Tm	1.9 y	Continuation fo branch at ^{170}Tm.
^{179}Ta	665 d	Origin of ^{180}Ta, nature's rarest stable isotope.
^{185}W	75 d	Important brach point.
^{204}Tl	3.8 y	Important to ^{205}Pb - ^{205}Tl clock for early solar system.

FIGURE CAPTIONS

Fig. 1. The $^{7}Be(n,p)^{7}Li$ reactivity verses temperature. The solid curve is the rate calculated from our data, while the dashed curve is the theoretical rate of ref. 12.

Fig. 2. The $^{22}Na(n,p)^{22}Ne$ reactivity verses temperature. The solid curve is the rate calculated from our measurements, while the dashed curve is the theoretical rate of ref. 1.

Fig. 3. Preliminary yield verses neutron energy from our $^{36}Cl(n,p)^{36}S$ measurements. The yield has not been corrected for the variation with energy of the neutron flux.

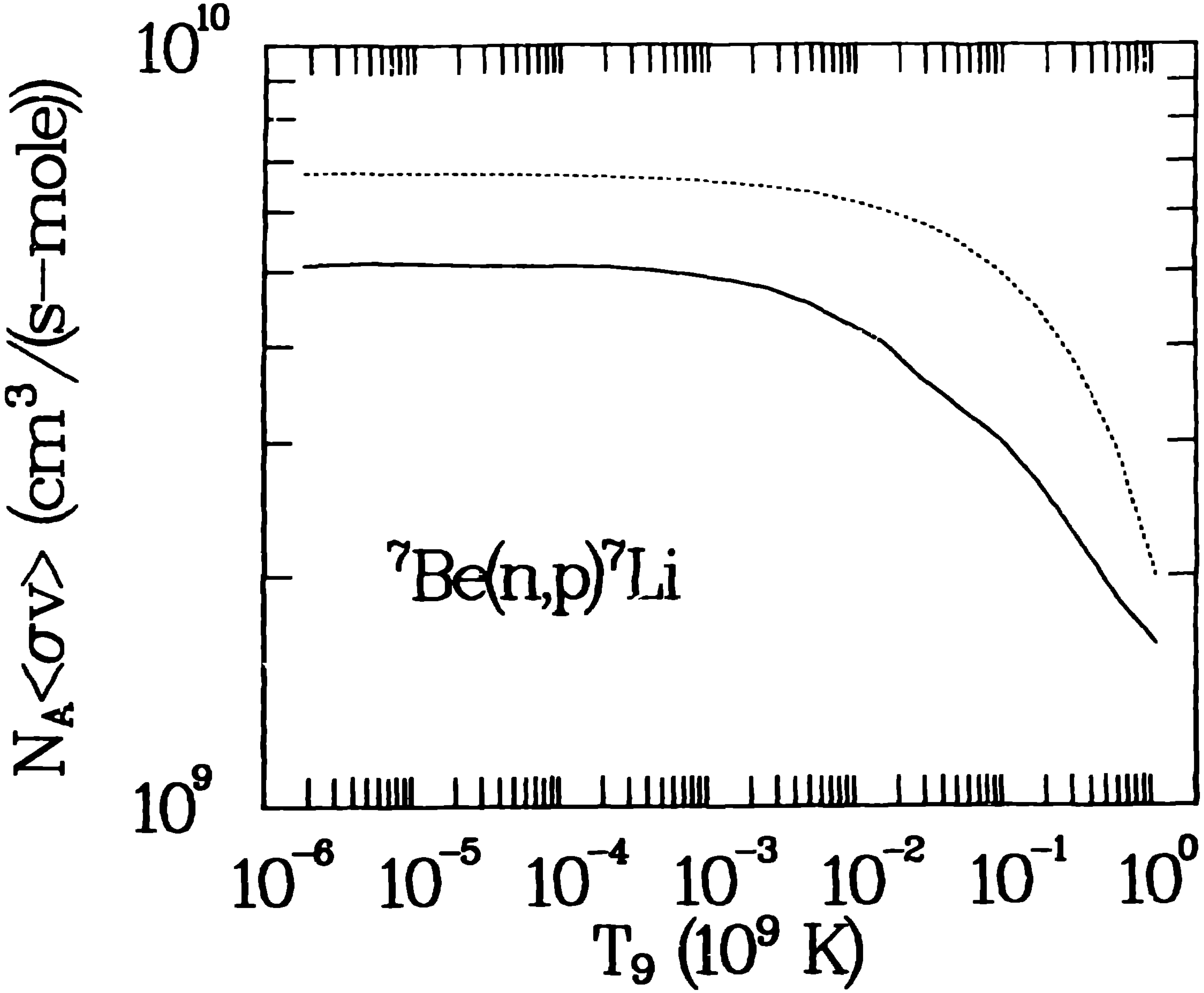

Figure 1. Koehler et al

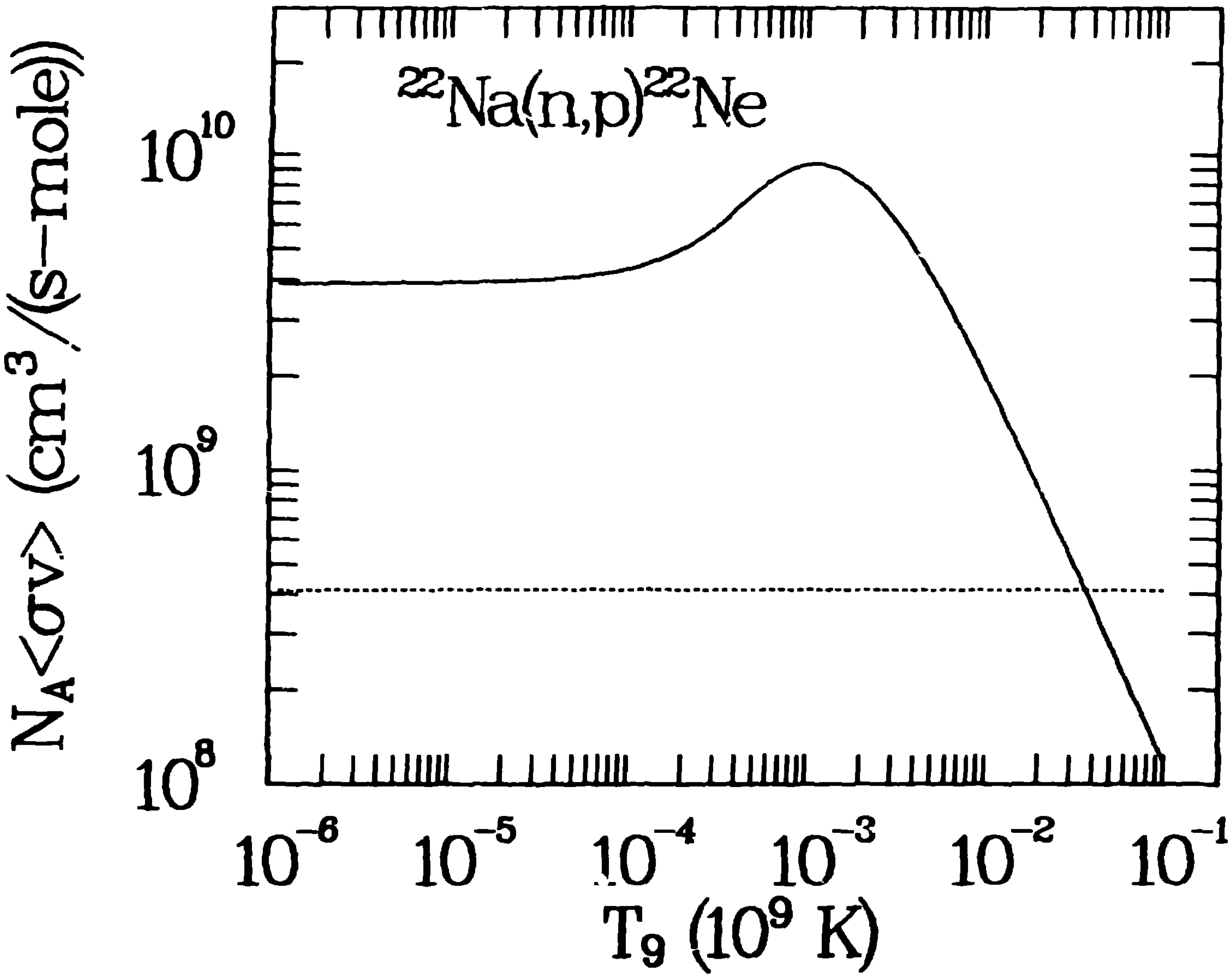

Figure 2. Koehler <u>et al</u>.

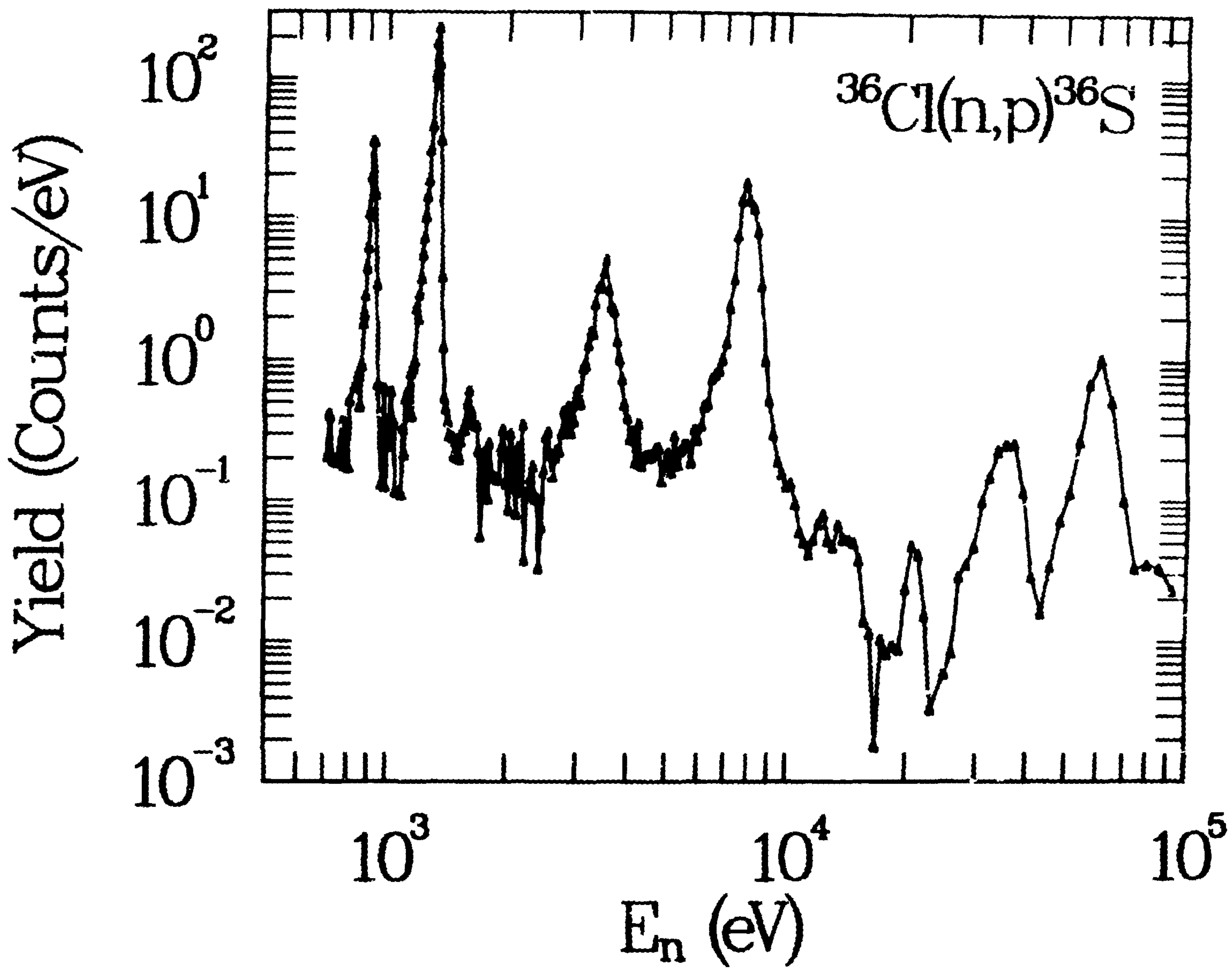

Figure 3. Koehler *et al.*