NUCLEI FAR FROM STABILITY USING EXOTIC TARGETS

J. B. Wilhelmy, G. E. Bentley, K. E. Thomas, Ronald E. Brown, E. R. Flynn, and
J. Van der Plicht Los Alamos National Laboratory, Los Alamos, N.M.

L. G. Mann, and G. L. Struble Lawrence Livermore National Laboratory, Livermore,
California

Abstract

The meson factories such as the Los Alamos Meson Physics Facility have made possible high fluence medium energy proton beams that can be used for spallation reactions to produce macro quantities of unstable isotopes. Targets of over 10 g/cm^2 can be exposed to total fluence approaching 1 A-hour resulting in spallation yields in the 0.01-10mg range for many isotopes of potential interest for nuclear structure studies. With the use of hot cell facilities, chemical processing can isolate the desired material and this coupled with subsequent isotope separation can result in usable quantities of material for nuclear target applicaton. With off-stable isotopes as target materials, conventional nuclear spectroscopy techniques can be employed to study nuclei far from stability. The irradiation and processing requirements for such an operation, along with the isotope production possibilities, are discussed. Also presented are initial experiments using a ^{148}Gd ($t_{1/2}$ = 75a) target to perform the (p,t) reaction to establish levels in the proposed double magic nucleus ^{146}Gd.

1. Introduction

Study of nuclei far from stability is usually accomplished using conventional targets with exotic reactions and/or sophisticated detection techniques. The major exception to these conventional targets has been in the study of the properties of the heavy actinides for which reactor-based production programs have provided sufficient materials for targets as heavy as ^{257}Fm[1,2]. With the advent of the high intensity meson factories such as LAMPF, a new possibility has emerged for the production of macro quantities of off-stability isotopes using spallation reactions to provide nuclear target materials. The Los Alamos Meson Physics Facility, on a routine basis, now provides beams of 500-600 μA of 800 MeV protons. The major purpose of the facility is to produce pions and muons in various production targets, but following this production a substantial amount of the primary proton beam is still available. Through irradiation facilities located at the beam stop area, spallation reactions can be performed on a variety of target materials. For this to be a viable operation, the capabilities and expertise for the handling and chemical processing of the intensely radioactive samples have to be available. To produce targets for further nuclear studies, the chemically separated samples have to be, in general, isotopically separated onto suitable backings for nuclear reaction studies. With these combined technology capabilities, it then becomes possible to reach nuclear regions far from stability by using conventional nuclear spectroscopy probes with their intrinsicly high efficiency and resolution. To date, we have begun initial steps to develop these capabilities and, as a demonstration of the technique, have produced a ^{148}Gd target ($t_{1/2}$ = 75a). With this target we have performed the ^{148}Gd (p,t) ^{146}Gd reaction to study the low lying levels in the interesting nucleus ^{146}Gd.

2. Irradiation and Processing

To make use of the large beam available in the LAMPF beam stop area, an extensive isotope production program was begun with the emphasis on medical applications[3]. There are clinical requirements for a large variety of specific radioactive isotopes for diagnostic and therapeutic application. Often it is desirable to produce large quantities of a relatively long lived isotope which can then serve as a "cow" or generator for some shorter lived isotope which is used in nuclear medical applications. With this procedure radioactive compound labeling can be obtained, if required, and a controlled specific radiation dose can be administered. For these purposes it is necessary to be able to produce macro quantities of the primary isotopes of interest. To this end an irradiation facility was constructed at the LAMPF beam stop and is schematically illustrated in Fig. 1. The constraints on this facility are that it be able to withstand the intense proton beam and provide convenient access to enable remote easy target insertion, exchange and servicing. This "stringer" facility allows the insertion of up to 9 target assemblies for simultaneous irradiation. Each of the targets is water cooled with a flow rate of ~60 ℓ/min to enable dissipation of the 30-50 kilowatts of power delivered by the beam. A photograph of a standard target assembly is shown in Fig. 2. The total thickness is 38 mm, and the primary housing is constructed of aluminum with a thickness in the irradiation region of 6 mm. A copper block is inserted into this irradiation window, and it contains the actual target material. In general the targets can be up to 12 mm thick and 25 cm^2 in area. The large area is required since the beam substantially diverges when it goes through the pion production targets and
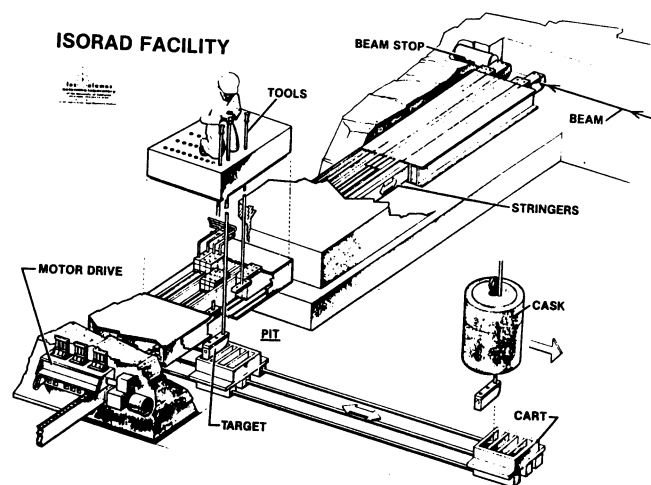

Fig. 1   Schematic representation of the remote irradiation facility at LAMPF. Up to nine targets can be irradiated at one time. Each is water cooled with a flow rate of ~60 liter/min.
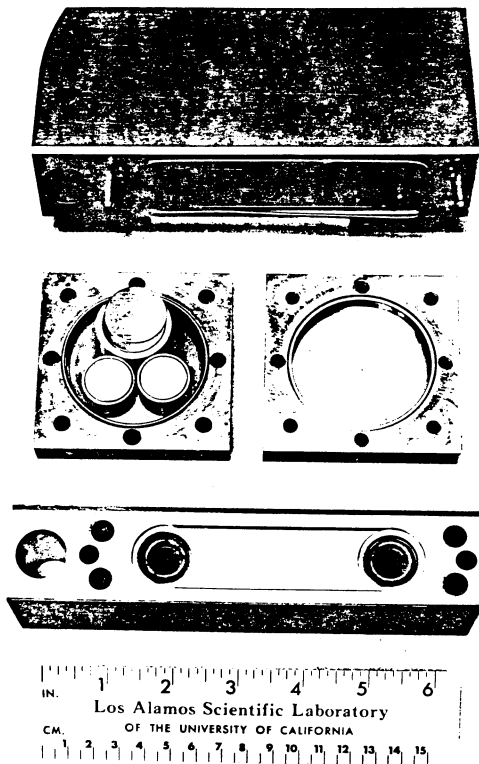
Fig. 2   Target assembly. Individual targets (in this case Mo) are placed inside a copper container – which has a receptive area of 25 cm^2 and is 1.25 cm deep.

continues to diverge through small angle scattering throughout the stringer target assembly.

A typical irradiation cycle of LAMPF is ~4 months during which time ½-1 A-hr of proton beam will be delivered to the stringer irradiation facility. During this period, targets will be inserted and removed as required for production of specific isotopes. In general, these irradiations result in intensely radioactive target assemblies with initial activity levels in the hundreds or thousands of Röntgen range. The assemblies are loaded into a special cask made of enriched ^{236}U and transported to a hot cell facility located some 5 km from LAMPF. In this facility the targets are removed from the cask and disassembled for chemical processing. All standard chemical procedures can be remotely carried out within the hot cell area. Once the desired isotopes are extracted, they can be packaged in a form suitable for shipment to medical facilities. This extensive capability also permits the possible isolation of desired isotopes for nuclear studies as well.

3.   Isotope Production

The initial feasibility studies of producing targets from isotopes made at LAMPF have concentrated on the spallation products formed with protons on Ta targets. Ta is an essentially monoisotopic metal and has excellent thermal properties for exposure to the high fluence beam. Since it is a heavy element, the spallation process concentrates production into a broad range of neutron deficient isotopes throughout the rare earth region. Figure 3 shows some experimental data and a calculated cumulative spallation yield cross section for 540 MeV protons on Ta using an inter-nuclear cascade model
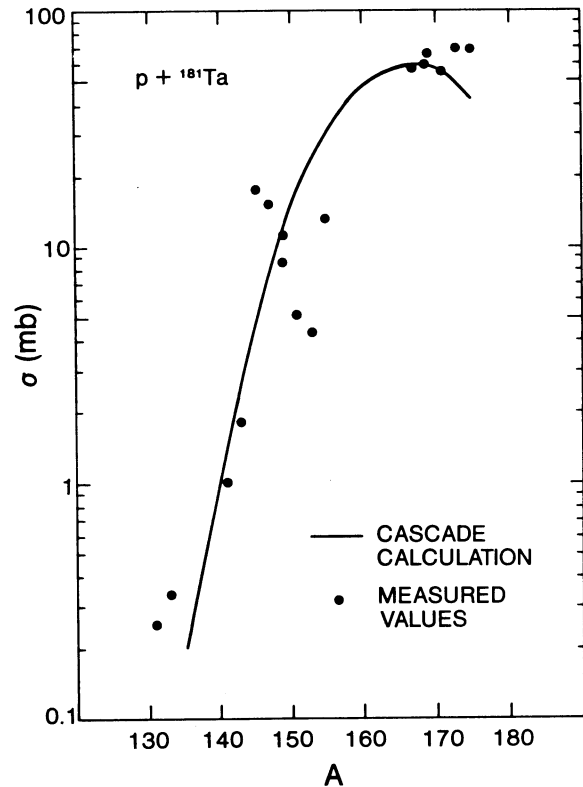

Fig. 3   Cross section for cumulative yields of mass chains for the reaction p+ ^{181}Ta. The solid curve is a cascade calculation followed by statistical evaporation[4]) for 540 MeV incident protons.

followed by statistical evaporation[4]). The model reproduces the general shape and magnitude of the measured distribution, but the important point to note is that there is substantial cross section for production of neutron deficient isotopes throughout the rare earth region. With an irradiation of a target of 10g/cm^2 with a total fluence of 0.5 A-hr of protons, a cross section of 1 mb results in production of ~100 μg of product.

The spallation yield is maximized for neutron deficient isotopes as indicated in Fig. 4. The average primary mass produced in the reaction is displaced by 5-10 neutrons from the line of beta stability toward the neutron deficient side. Of course, the distribution is quite wide having a σ of about 2.2. for a given mass chain. This provides independent yields to a substantial number of isotopes and thus, to some extent, can overcome shielding effects associated with beta stable isotopes blocking the cumulative chain. In this production region several isotopes are of potential interest for targets for nuclear structure studies. For ease of handling the initial efforts will concentrate on the long lived α emitting rare earths ^{146}Sm ($t_{1/2} = 10^8$a), ^{148}Gd ($t_{1/2} = 75$a) ^{150}Gd ($t_{1/2} = 1.8 \times 10^6$a) and ^{154}Dy ($t_{1/2} \sim 10^7$a). Of these the most interesting are ^{146}Sm and ^{148}Gd since the (p,t) pick up reaction can be used to study the closed N=82 shell. The nucleus ^{146}Gd has received considerable attention recently due to its apparent double closed shell structure and in the next section we discuss our first results with the radioactive target ^{148}Gd to produce and study this isotope.

Other isotopes of potential use are indicated in Fig. 4. Of these, two deserve special mention.
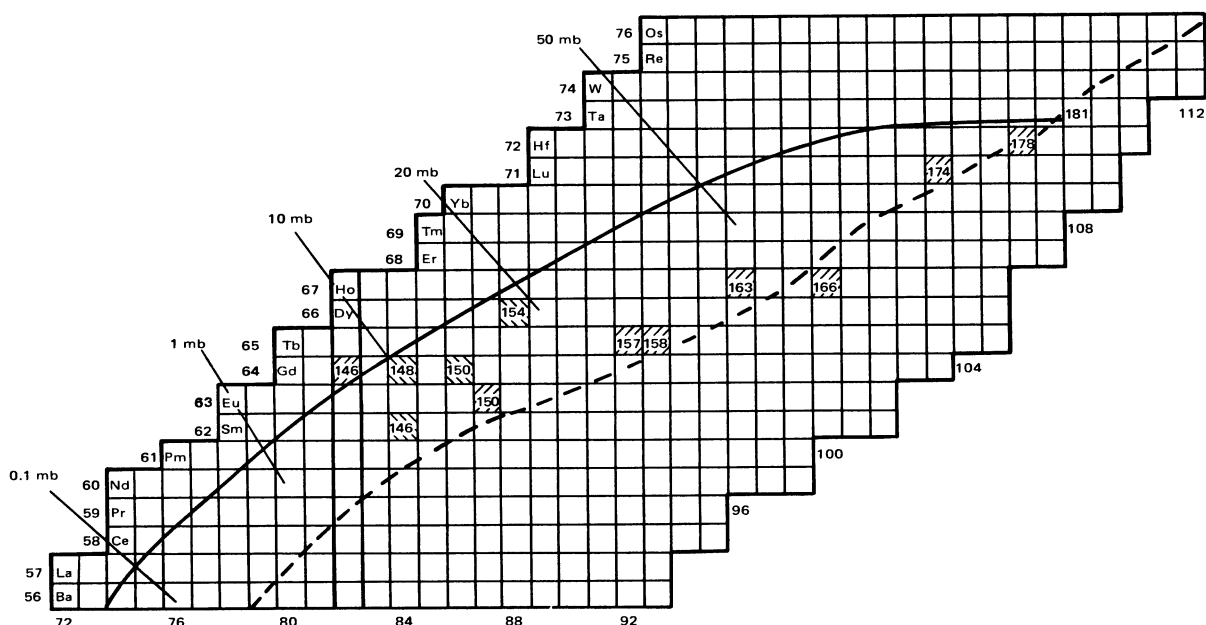
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Fig. 4. Portion of a chart of the nuclides. The dashed line is the line of beta stability, and the solid line indicates the average product for a given mass formed in the proton induced spallation of ^{181}Ta. The cross hatched boxes indicate isotopes of potential interest for target preparation. Also shown are production cross section intervals.

The first is ^{146}Gd which has a relatively short half life of only 48.3 days. Since this is a presumed doubly closed shell nucleus various inelastic and direct reaction probes should provide corroboration for this assignment. Adequate quantities of this isotope can in principle be isolated. The most recent Gd extraction from an irradiated Ta target (Table I) had over 100 μg of this isotope at the end of bombardment. However, even a target containing only 5 μg of the isotope will have a disintegration rate of 100 millicuries and, therefore, require adequate shielding for handling purposes. Another isotope of high interest is ^{178m2}Hf. This 31 year isomer is at an excitation energy of 2.4 MeV but, most importantly, has a spin of 16^+. The nuclear structure properties associated with high angular momentum are currently an active area of research. Theoretical speculation concerning yrast traps and shape changes for nuclei of high angular momentum[5]) has resulted in extensive experimental programs to study these regions. Our recent irradiation of 11.9 g/cm^2 of Ta with 0.565 A-hr of protons has resulted in the production of over 40 μg of the ^{178m2}Hf high-spin isomer. With this as a starting material, Coulomb excitation studies using heavy ion projectiles should be able to reach the highest identified nuclear spin states. There is substantial effort being expended on the development of the so called "spin spectrometers" or "crystal balls" for measuring γ-ray multiplicities utilizing large NaI arrays. The availability of ^{178m2}Hf would permit an extension in the use of this sophisticated equipment into unique areas.

TABLE I

Gd Assay

Mass	146	148	151	153
$t_{\frac{1}{2}}$ (a)	0.13	75.	0.33	0.66
Yield t=0 (μg)	115.	300.	206.	179.

There are still substantial problems to be overcome before ^{178m2}Hf becomes readily available for nuclear structure studies. To obtain a suitable target the chemically isolated Hf fraction has to be isotopically separated. Since Hf is a refractory element, the efficiency for this separation might be low unless extensive ion source development is performed. Also, even though the production yield of the isomer is acceptably large, the production of the stable ground state is much larger. The cascade calculations estimate that the ground state yield will be ~150 times as large as that measured for the isomer (Table II). Even at this impurity level it

TABLE II

Hf Assay

Mass	172	175	178m2	178
$t_{\frac{1}{2}}$ (a)	1.87	0.19	31.	Stable
Yield t=0 (μg)	6220.	5290.	41.	(6000)

may be possible to use this target material in Coulomb excitation studies by exploiting the higher total γ-ray energy released associated with the isomer when compared with the ground state. An attractive area of research for this isomer is exploring the possibility of performing multiple step selective photoionization with laser systems. A high resolution tuneable dye laser could be used to resonantly excite the unique hyper-fine level splitting associated with the isomer. This coupled with a high intensity laser could then be used to selectively photoionize the isomer permitting, in principle, isomer separation.

In addition to isotopes produced with spallation reactions on Ta there is interest in several light elements. Production targets of Cu, V and Si have been irradiated in the primary proton beam. For these lighter elements the lower charged particle Coulomb barrier results in more equivalent proton and neutron emission. The yields are, therefore,

not as skewed toward the neutron deficient region. Figure 5 presents this lighter element region and indicates isotopes of interest. Table III summarizes these isotopes and indicates the production yield expected in a typical LAMPF irradiation cycle (~4 months). In all cases 10's-100's of μg are
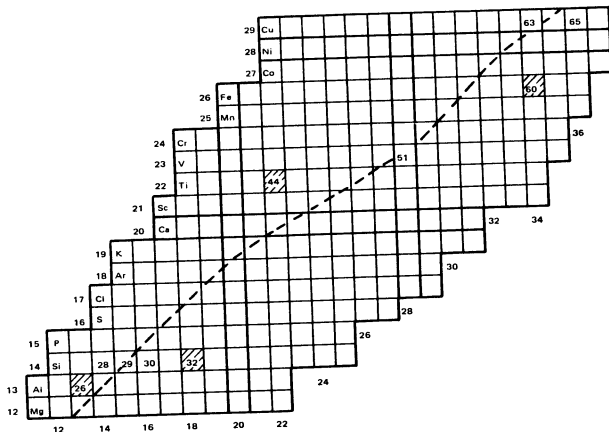

Fig. 5. Portion of a chart of the nuclides emphasizing the ligher element region. Dashed line is the line of beta stability. Labeled open boxes are primary irradiation targets for proton induced spallation reaction, while the cross hatched boxes represent isotopes of interest for secondary target preparation.

TABLE III

Production of Light Isotopes

Isotope	$t_{\frac{1}{2}}$ (a)	Target	Production/Cycle (μg)
^{60}Fe	3×10^5	Cu	20-200
^{44}Ti	47	V	20-200
^{32}Si	~650	V	30-300
^{26}Al	7×10^5	Si	50-500

expected, and this should be adequate feed material to produce usable nuclear targets. Properties associated with these isotopes can yield information regarding the nucleosynthesis process in stars and the processes involved in supernova explosions. Through the use of the (t,^{3}He) reaction information can be acquired on the low lying 1^+ levels [6]) and thus some insight for the Gamow-Teller decay strength, which is important for understanding isotopic production in nucleosynthesis and in supernova. The extension of these studies to the most neutron rich isotopes is important and, therefore, measurements on ^{60}Fe (3 x 10^{5}a) is of special interest. The (t,p) reaction on ^{60}Fe would provide an extension of available information on level structure for the nuclear synthesis codes.

The (p,γ) measurements on ^{26}Al (7.3 x 10^{5}a) are also of astrophysical interest. Recent measurements[7]) of the isotopic content of ^{26}Mg in meteorites has led to discrepancies in the theories of nucleosynthesis and the origin of the solar system. By directly measuring the (p,γ) cross section on this isotope the nature of this apparent discrepancy can be investigated. Studies on ^{44}Ti (47a) can provide insight into level structures in this region and are also important for nucleosynthesis systematics. Another unique isotope for study is ^{32}Si (~650a). This very neutron rich isotope permits

exmination of the nuclear region which bridges the gap between the deformed s-d shell and the closed N=20 neutron shell. Using (t,p), (t,α) and (t,^{3}He) reactions this region could be investigated in detail.

For each isotope to be studied, chemical processing techniques have to be perfected for operation in remote hot cells to efficiently extract microgram quantities from up to kilogram samples. In general these chemically isolated products have to be isotopically separated onto thin target backings. This requires development of ion source technology and the availability of a separator which can safely handle the radioactive feed material and be adequately decontaminated following the separation. As a long term development, shorter lived isotopes will become more attractive for potential target material. This will necessitate rapid handling throughout the processing and coordination with accelerator scheduling to maximize the usefulness of the target.

4. **Initial Experiment:** ^{148}Gd(p,t)^{146}Gd

The nucleus ^{146}Gd has received considerable attention recently because of its supposed doubly closed shell character. It has, like ^{132}Sn and ^{208}Pb, a high lying 3^- first excited state. Because ^{146}Gd lies too far from the line of beta stability to be examined by normal light ion particle transfer, it has been studied only through γ-ray and conversion electron measurements following xn reactions[8-14]) and by two proton transfers using heavy ions[15]) and the (^{3}He,n) reaction[16]). In order to complement the xn studies and to improve the resolution obtainable with the two proton transfer reactions we have prepared a ^{148}Gd (t$_{\frac{1}{2}}$ = 75a) target and performed the ^{148}Gd(p,t)^{146}Gd reaction.

The target material was produced by spallation reactions on a 22 g/cm^2 Ta target. The primary beam energy of 800 MeV protons was degraded to the 400-700 MeV range through various production targets placed before our target. This was a relatively early exposure at LAMPF with the irradiation carried out during 1978 when the average beam current was on the order of 100 μA. An integral beam fluence of ~5.6 x 10^4 μA-hr was seen by the target. The tantalum was transported to a hot cell facility for chemical processing. It was dissolved in an HF/HNO$_3$ mixture, and the rare earth group was isolated from the bulk soluton by adding a lanthanum carrier and precipitating the rare earth fluorides. The rare earths were placed back in solution and separated from each other by using standard cation exchange chromatography with α-hydroxyisobutyric acid as the column eluant. The chemically extracted gadolinium fraction was assayed to contain 9.8 μg of ^{148}Gd. A 1 mg carrier of natural Gd was added to the solution and then precipitated as a wet hydroxide. The precipitate was shipped to Lawrence Livermore National Laboratory where it was converted to an oxide and isotopically separated. The ^{148}Gd was collected on a 40 μg/cm^2 carbon foil onto a nominal spot size of 2 x 2 mm^2. The target was returned to Los Alamos where a thickness determination was performed using ^{14}C elastic scattering, which gave only a limit of <16 μg/cm^2.

Using the three stage capability at the Los Alamos Van de Graaff facility we used 24 MeV protons to perform the (p,t) reaction. Assuming the ground state cross section of the reaction on ^{148}Gd to be the same as the previously measured ^{142}Ce(p,t)^{140}Ce,[17]) a thickness of 6 μg/cm^2 was obtained. The triton spectra were recorded using a Q3D spectrometer

having a 1 meter position sensitive helical wound detector located at the focal plane. Figure 6 present a spectrum obtained at a scattering angle of $25°$. Even with this very thin target a nine point angular distribution was measured for all states below 2.5 MeV. Experimental results and the corresponding DWBA fits are shown in Fig. 7 for three different levels. One important result from these measurements is that no excited states lie below the 1.575 MeV level, thus corroborating previous studies and supporting the double magic character of this nucleus. The DWBA analysis of this level is consistent with an L=3 assignment, but we do not feel it is sufficiently unique to firmly establish the spin from this analysis. The level at 1.980 MeV is established as an L=2 transfer and is consistent with assignment as a 2^+ level made through difficult threshold analysis in an $^{144}Sm(\alpha,2n\gamma)^{146}Gd$ measurement[12]. In the normal γ-ray cascade this level is not a member of the yrast band and is thus not populated. With our light ion direct reaction technique the assignment of this important level is clearly established. The level at 2.173 MeV has the distinctive signiture of an L=0 transfer and is in agreement with the recent assignment based on xn, conversion electron measurements[14]. This level is, however, not the pairing vibration state since it contains less than 10% of the ground state strength. Figure 8 presents a level scheme for the low lying levels in ^{146}Gd as deduced from our (p,t) measurements along with previous assignments from xn studies.

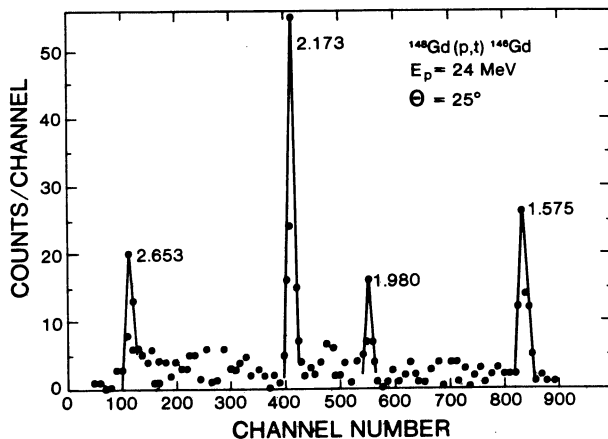The results of the present experiment clearly show that useful nuclear reaction results can be

Fig. 6   A portion of the triton spectrum from the reaction ^{148}Gd (p,t) ^{146}Gd.

obtained on targets as thin as ~ 10 $\mu g/cm^2$ using light ion reactions and a high resolution spectrometer. Irradiation using higher beam fluences can potentially provide orders of magnitude more material for target preparation. We anticipate that this will open new regions for significant physical measurements in previously inaccessible areas.
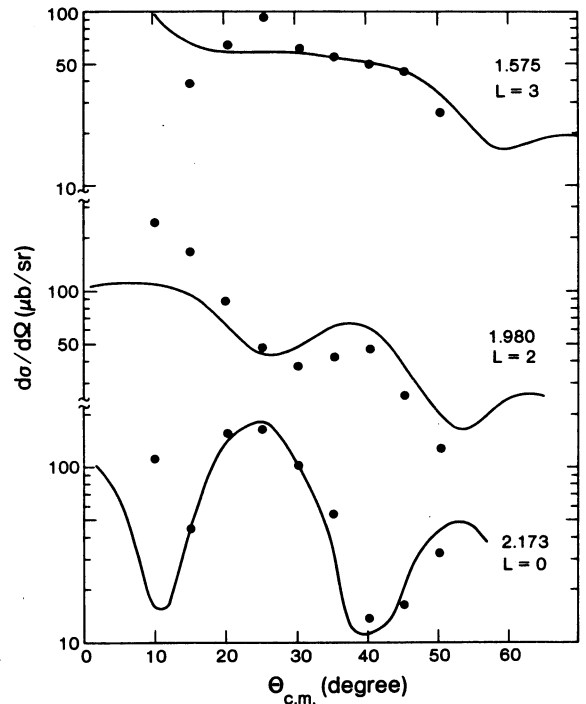
5. Acknowledgements

Fig. 7   Angular distribution for three levels in ^{146}Gd. The solid lines are DWBA fits with the indicated transferred angular momentum.

Fig. 8   Deduced level scheme for low energy states in ^{146}Gd. Assignments from the current (p,t) investigations are indicated on the right, while previous assignments based on (xn,μ and e$^-$) are on the left.

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DISCUSSION

E. Roeckl: What are the overall efficiencies you obtained in mass separating gadolinium and hafnium?

J.B. Wilhelmy: For the gadolinium efficiencies as high as 60% have been obtained. However, in the case of the preparation of our target on a thin carbon backing the collection was $\sim$ 15%. For hafnium we have not determined an efficiency as yet but for the chemical homolog Zr we have obtained $\sim$ 5-10% collection efficiencies.

W. Benenson: Can you tell us how dangerous and radioactive the actual targets are?

J.B. Wilhelmy: For the ^{148}Gd target we have used, the activity was $\sim$ 20-30 μCi and since this is a low energy alpha emitter it is relatively easy to handle. We do however hope to make a target of ^{146}Gd which has only a 38-day half life. In this case a target containing 5 μg of material will have an activity level of $\sim$ 100 mCi and this requires more caution in handling. We do have experience in working with similar activity levels in actinide element targets where we have not encountered undue difficulties.

R.A. Naumann: As an indication of the feasibility of using radioactive targets of moderately short half-life for triangular spectroscopy, I can report that we have been carrying out experiments at Princeton using 300-day berkelium 249 in collaboration with the Livermore National Laboratory. The targets were isotope separated at Livermore and transported and used at Princeton without problems.

J.B. Wilhelmy: We have had similar experience at Los Alamos.

P. Kleinheinz: The 2.16 MeV 0^+ state in ^{146}Gd as discussed by Julin et al. [Phys. Lett. 94B (1980) 123] is predominantly the proton 0^+ level, and therefore quite expected to be only weakly seen in (p,t). It also would be most attractive to determine the masses of ^{146,147,149,150}Gd from transfer studies with this target, and to obtain neutron single particle strengths.