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Special Targets for Nuclear Reaction and Spectroscopic Studies

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Strongly focused and monoenergetic charged-particle beams from modern accelerators and targets fabricated from quantities of isotopically enriched and stable materials are the essential components from many current nuclear physics experiments. Although a large body of this kind of experimental work requires substantial amounts of target material, an important subset of such experiments can be done with as little as a few μg of material. Experiments where charged particles or electrons can be focused on or transported to a detector are examples of accelerator-based studies which can be made with targets that contain relatively small amounts of material. For these kinds of studies, it then becomes possible to extend the domain of potential target materials to species which are very rare or which are unstable and undergo radioactive decay. At our laboratory during the last ten years, we have made targets for nuclear spectroscopy studies of ^{152}Eu (13.4y), ^{154}Eu (8.5y), ^{249}Bk (320d), ^{151}Sm (90y), and ^{148}Gd (75y). We will report our experience with fabricating these and other kinds of stable targets and discuss our plans for preparing additional targets which offer interesting and exciting prospects for future nuclear research studies.

Although there are always a variety of technical difficulties associated with doing any kind of nuclear science research, the problem of obtaining a reasonable target for various accelerator or reactor based experiments is the most pervasive. In principle, the idea of bringing together focused particle beams and stationary target nuclei is easily conceptualized. In practice, however, once an interesting physics study is identified, the preparation of a suitable target usually involves significant technical challenge and ingenuity. In this report, we describe some of our experience in our Nuclear Properties Group at the Lawrence Livermore National Laboratory (LLNL) with preparing special targets for nuclear research studies.

Our studies have involved the use of both neutron and charged-particle beams and Table I presents a partial list of the targets prepared. The Nobel gas targets were prepared for in-beam γ -ray studies which used an external thermal neutron beam. Large amounts of target material were required for these experiments and a special device - a cryostat - was constructed to isolate and contain a large quantity of gas (in the solid state) so that the measurements could be done. The details of the construction of this device, as well as the various studies performed with it have been described previously.^{1,2,3}

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The ^{176}Lu target is unique because this material has an extremely low natural isotopic abundance (2.61%). A sample of several tens of milligrams of Lu_2O_3 was obtained from the Oak Ridge National Laboratory and had an isotopic enrichment of $\sim 70\%$ in ^{176}Lu . An additional isotope separation step⁴ was done on this sample at LLNL. By this procedure we were able to make a target of isotopically pure ^{176}Lu ($\sim 99.9\%$) that had a thickness of $\sim 22 \text{ mg/cm}^2$ and which was supported on a Th substrate. This target was subsequently used for ^{16}O Coulomb-excitation experiments.⁵

The remainder of the list in Table I involves radioactive targets which were prepared for charged-particle spectroscopy experiments. These targets were generally thin ($10\text{--}40 \text{ }\mu\text{g/cm}^2$) and were used for studies where light-ion reaction products were measured with a magnetic spectrograph. In general, the procedure for preparing such targets involves three steps: (1) production of the target material, (2) material purification and (3) depositing the material on a suitable substrate. We describe here in detail our preparation of a ^{148}Gd target. A similar procedure was used for the remaining radioactive targets. The results from the various studies performed with these targets have been published elsewhere.^{6,7,8}

Table I. Targets for nuclear studies prepared by the LLNL Nuclear Properties Group.

Isotopic Material	$t_{1/2}$	Material Production Technique	Approx. Thickness	Nuclear Studies (reference)
^{86}Kr	--	--	2 g/cm^2	2
^{136}Xe	--	--	2 g/cm^2	3
^{148}Gd	75y	Ta + P spallation	$25 \text{ }\mu\text{g/cm}^2$	10
^{151}Sm	90y	$^{150}\text{Sm} + n$	$35 \text{ }\mu\text{g/cm}^2$	7
^{152}Eu	13y	$^{151}\text{Eu} + n$	$30 \text{ }\mu\text{g/cm}^2$	6
^{154}Eu	8.6y	$^{153}\text{Eu} + n$	$15 \text{ }\mu\text{g/cm}^2$	6
^{176}Lu	--	--	22 mg/cm^2	5
^{249}Bk	311d	$^{247}\text{Bk} + 2n$	$30 \text{ }\mu\text{g/cm}^2$	8

Raw material for the ^{148}Gd target was produced by spallation reactions in a tantalum metal target by $\sim 750\text{-MeV}$ protons at the Isotopes Production Facility at LAMPF. The details of the irradiation and the chemistries associated with separating the hafnium and lanthanide fractions have been reported previously.⁹

The source material we obtained had been separated from the lanthanide fraction and contained gadolinium dissolved in an acidic solution. An assay of this material showed that it contained ~ 170 μg of ^{148}Gd and high levels of europium, samarium, and terbium contamination. In addition, a high level of lead contamination was present in ~ 20% of the sample because of leakage into a shielded container during shipping. We used standard ion-exchange chemistries to remove the lead and rare-earth contaminants and obtained a clean gadolinium fraction that contained ~ 132 μg of ^{148}Gd .

We prepared samples for the isotope separator from the clean gadolinium fraction by electrodeposition. The pure gadolinium fraction was evaporated to dryness, converted to the nitrate, and dissolved in ~ 200 mL of 0.1N HNO_3 . A 20- μL aliquot was taken from this solution and added to about 1 mL of isopropyl alcohol in a small-volume (2 mL) quartz electrodeposition cell. A small platinum disk was sealed to the base of the cell and served as the anode. The cathode, which acted as the collector electrode, was a small strip of tungsten (10 mm x 2 mm x 0.03 mm); it was immersed to a depth of ~ 3 mm in the nonaqueous plating solution. We began the deposition by using an applied voltage of ~ 200 V and typically observed initial cell currents in the range 0.2 to 0.4 mA. We concluded the plating procedure when the current dropped below ~ 0.1 mA with an applied voltage of ~ 400 V. The time to complete the deposition was ~ 1 h. During plating, we adjusted the voltage to maintain cell current in the range 0.2 to 0.5 mA, but did not allow it to exceed 400 V. The deposit on the cathode was not chemically identified; it had a white powdery appearance and may have been a hydrated oxide of gadolinium. Eight tungsten strips coated with gadolinium were prepared in this way, and the total amount of ^{148}Gd plated was ~ 99 μg .

We prepared thin targets by collecting a ^{148}Gd beam from the Nuclear Chemistry Isotope Separator⁴ on thin (~ 50- $\mu\text{g}/\text{cm}^2$) carbon foils. The gadolinium-coated tungsten foils each contained 10 to 15 μg of ^{148}Gd . We placed the individual tungsten foils in the isotope separator ion source and collected ^{148}Gd on the carbon foil until the beam became depleted. Approximately two coated tungsten foils were required to deposit ~ 1 μg of ^{148}Gd on a single carbon foil. We prepared four targets in this manner. We collected the ^{148}Gd over an area of about 3 mm x 1.5 mm to get target thicknesses ranging between about 20 and 30 $\mu\text{g}/\text{cm}^2$. We estimate that the total amount of ^{148}Gd finally recovered as usable targets was about 3 to 4 μg or 2.1% of the original material.

We used the ^{148}Gd targets for a number of charged-particle reaction experiments for nuclear structure studies.¹⁰ For one of the targets, a sample triton spectrum from the $^{148}\text{Gd}(p,t)^{148}\text{Gd}$ reaction is shown in Fig. 1 which is a computer reconstruction of the original data on a linear energy scale and combines the results of five separate experiments. Each experiment revealed only a portion (~ 20%) of the total spectrum. The spectrum is of excellent quality and, in particular, is free from contaminating peaks caused by other rare-earth materials.

Fig. 1

Table II summarizes the losses we experienced at each step of the target preparation procedure. As the table shows, the most inefficient aspect of our technique is the isotope separation step. Normally, we expect $\sim 30\%$ recovery with isotope separation of gadolinium, but this level was obviously not obtained in this particular case. With other rare-earth materials, such as europium and samarium, the expected isotope separation recovery is $> 50\%$ and can reach levels as high as 70% . Therefore, allowing for similar chemistry losses, we expect that in those selected cases where isotope separation recoveries are $\sim 50\%$, the amount of initial material required to make a reasonable number of usable targets can be as low as $\sim 100 \mu\text{g}$. The amount of initial material required can also be reduced somewhat by paying more attention to increasing the efficiencies of the wet-chemistry yields.

Proposed future work in our group will continue to involve the need for special targets. In particular, we are investigating techniques to prepare radioactive targets of ^{150}Eu (30y) and ^{227}Ac (22y) for charged-particle spectroscopy work. Each of these targets present special problems which are outside our range of experience and offer considerable technical challenge. Beyond that, we have begun a program to measure light-ion charged-particle cross sections by activation and in-beam techniques. Activation studies require the use of many separate target foils. Moreover, each foil must be homogeneous and each must have a thickness known accurately to a few percent.

Table II. Yield summary for ^{148}Gd target preparation.

	Original material ^a	Initial cleanup ^b	Electro- deposition ^b	Isotope separation ^c
Mass (μg)	170	132	99	4
Relative yield (%)	--	78	75	4
Cumulative yield (%)	--	78	58	2

^aThe initial amount of ^{148}Gd was determined by alpha-counting a small aliquot from the new sample.

^bThe chemical yields were determined by gamma-counting techniques. The sample contained a number of radioactive isotopes of gadolinium prior to isotope separation.

^cThe assay of the final targets was obtained by back alpha-counting and x-ray fluorescence techniques.

We are experimenting with two schemes to prepare targets for cross-section measurements. The first is forming a self-supporting "ceramic" target which can be prepared from a metal oxide. This procedure has been used by Quinby,¹¹ who prepared self-supporting oxide targets with thicknesses in the range 150-2500 $\mu\text{g}/\text{cm}^2$ from several materials. An alternate technique involves producing thin Kapton¹² plastic sheets (1-2 mil) which contain accurately known amounts of any material. The only requirement for the target material is that it is in the form of a powder and that the natural particle size is small enough so that it can be homogeneously mixed (coated) with the liquid plastic base before spreading and curing. Both techniques show considerable promise for making cheap, high quality targets for light-ion studies when low-Z element contamination can be tolerated.

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