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RADIOACTIVE TARGETS FOR NUCLEAR ASTROPHYSICS RESEARCH AT LANSCE

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Abstract: During the past few years we have made measurements of (n,p) and (n,α) cross sections on several radioactive nuclei of importance to nuclear astrophysics. The measurements were made at the Manuel Lujan, Jr. Neutron Scattering Center (LANSCE) from thermal neutron energy to approximately 100 keV. Successful measurements have been completed on the radioisotopes ^{7}Be, ^{22}Na and ^{36}Cl while preliminary data have been taken on targets of ^{54}Mn and ^{55}Fe. Similar measurements have also been made on the stable isotopes ^{14}N, ^{17}O and ^{35}Cl. We are currently assembling a 4π barium fluoride (BaF_2) detector which will allow us to expand our program to (n,γ) measurements. The (n,γ) (and in some cases future (n,p)) measurements will require targets with higher specific activity and greater chemical purity than we have so far been able to use. We discuss the fabrication techniques used for the samples produced so far, the requirements the future (n,γ) targets must meet and our current plans for producing them, and the physics motivations for the measurements.

1. Introduction.

The advent of the large peak intensity available from the LANSCE white neutron source [1] makes possible measurements on small samples (approximately 100 ng to 100 μg) of short-lived radioactive isotopes [2]. Such measurements were difficult if not

impossible in the past due to the potentially large background from the radioactive sample. At LANSCE, measurements of (n,p) and (n,α) cross sections for radioactive samples have been made using straightforward techniques employing solid state detectors or ionization chambers. Measurements of (n,γ) cross sections are more difficult, and require a more elaborate and more costly 4π scintillation detector. For most of the planned measurements, the largest remaining difficulty is obtaining a suitable radioactive sample. Because they affect the target requirements, we will first give a brief outline of the motivations behind the measurements, followed by a description of our experimental facilities. Then, we will discuss the requirements a suitable radioactive target must meet, the methods we have so far used for producing them, and our future plans.

2. Motivations for the measurements

Measurements of (n,p), (n,α) and (n,γ) cross sections for radioactive targets have applications in nuclear physics, nuclear astrophysics, and in many applied programs. The major basic physics motivation for our measurements has been the nuclear astrophysics to be learned. For example, the $^{7}Be(n,p)^{7}Li$ reaction [3] is important in calculations of the big-bang nucleosynthesis of ^{7}Li, cross sections for the $^{22}Na(n,p)^{22}Ne$ and $^{22}Na(n,\alpha)^{19}F$ reactions [4] may aid in the explanation of the neon-E anomaly in meteorites [5] or in the possibility of observing nucleosynthetic ^{22}Na with future gamma-ray telescopes [6], and the $^{36}Cl(n,p)^{36}S$ reaction may be important in the nucleosynthesis of the very rare stable isotope ^{36}S in both explosive nucleosynthesis [7] and in the s-process [8]. There are a few other (n,p) and (n,α) reactions on radioactive nuclei of importance to nucleosynthesis calculations [7,9], and even some stable targets of importance [10]. In addition, there are a number of (n,γ) cross sections on radioactive targets which if measured would provide an almost model independent understanding of the dynamics of the s-process of nucleosynthesis [11]. This is important because current

stellar models [12] of the dynamical s-process environment fail to reproduce the experimental data.

The measurement of fission cross sections on radioactive targets is another area of interest. We have recently completed (n,f) measurements on ^{250}Cf, ^{254}Es and ^{247}Cm [13]. Finally, cross sections on radioactive targets are important in the study of the feasibility of using an accelerator driven neutron source to transmute nuclear waste [14].

3. Experimental setup

At LANSCE, neutrons are produced by spallation when the 800 MeV proton beam from LAMPF strikes a tungsten target. The neutrons are then slowed down in a water moderator and are collimated by layers of iron, brass, borated polyethylene, and lead to form a beam spot 0.5 cm in diameter at a position 7 m from the moderator. The flux at 1 eV was measured [15] to be 2.3×10^6 neutrons/(s eV cm^2) for a proton current of 57 μA, and hence, at the 100 μA design intensity, the flux is expected to be about 4×10^6 neutron/(s eV cm^2). The flux follows an approximately 1/E shape for energies between 0.2 eV and 100 keV. The small beam spot size was chosen so that a large solid angle could be covered with a relatively small detector. This is important in (n,p) and (n,α) measurements where, to minimize the background, solid state detectors of minimum thickness, and hence relatively small area (typically 50-450 mm^2), are used. Having a compact detector is important for (n,γ) measurements because the best scintillator for this use, BaF_2, is relatively expensive. For example, a BaF_2 detector of about 15 cm thickness is required for the (n,g) measurements [16]. Because of our small beam size, we can obtain this thickness by using a detector in the form of a 30 cm cube as shown in figure 1.

4. Target requirements

For (n,p) and (n,α) measurements, there are three major requirements that a target must satisfy. First, the target must be thin enough to allow the charged particles to emerge without too much straggling. Typical Q-values for reactions of interest range from about 0.5 to a few MeV. Hence acceptable target thicknesses range from a few tens of $\mu g/cm^2$ to as high as about 1 mg/cm^2. Because of our small beam spot size, this is usually the hardest requirement to satisfy, and often results in the need for a fairly high specific activity. Second, impurities with positive Q-values for (n,p) and/or (n,α) reactions and having comparatively large cross sections must be kept to a minimum. In practice the most troublesome impurities are ^{10}B and ^{6}Li, and targets made from chemical compounds are almost always acceptable provided that the target is not too thick. Third, the target backing should be as thin as practical, and should have a low neutron scattering cross section. In most cases this requirement is easy to satisfy by using a backing of aluminum foil as thick as 25 μm.

For (n,γ) measurements, potentially everything on the target (including the isotope under study) is a source of background caused by neutrons scattered from the target and subsequently capturing in the BaF_2 detector. Hence the ideal target would be monoisotopic and self supporting. First, this means that chemical compounds are usually not acceptable. Second, the specific activity must be high. The lower the specific activity, the more time must be spent in measuring and subtracting backgrounds, and the better the "impurities" in the target must be known. Finally, the target backing foil, if needed, should be as thin as possible. Calculations indicate that carbon foils of about 30 $\mu g/cm^2$ or thinner will be needed in most cases. Alternatively, aluminum foil as thin as 0.8 μm is available commercially, and should yield about the same neutron scattering background as 30 $\mu g/cm^2$ carbon.

5. Previous targets

The radioactive targets prepared so far have been made by depositing the target material from a water solution into a shallow depression in an aluminum backing foil. In addition to its simplicity, the main advantage of this "evaporation" method of preparing radioactive targets is the high efficiency of transfer of the target material to the backing foil. The major disadvantages are the lack of target uniformity, and the relative fragility of the targets. With careful handling, the latter disadvantage is not too serious for our work where the targets do not receive significant heating from the incident neutron beam.

The ^{7}Be was produced via spallation reactions by bombarding a zinc oxide target with 750 to 800 MeV protons in the Isotope Production Facility (IPF) at the Clinton P. Anderson Meson Physics Facility (LAMPF) [17]. The beryllium was chemically separated from the target and recovered as BeF_2. The specific activity was not measured. Aqueous BeF_2 was deposited into a shallow depression in a 8.5 μm thick aluminum foil. A heater placed beneath the foil facilitated the evaporation of the solution. This technique resulted in a fairly uniform deposit roughly 3 mm in diameter that contained approximately 90 ng of ^{7}Be. This target was employed for successful measurements [3].

The ^{22}Na was produced in an ultrapure aluminum target that was bombarded in the IPF at LAMPF. Chemical separation and purification of the ^{22}Na were performed by scientists at Dupont's Pharmaceutical and Imaging Agents Division (formerly New England Nuclear) utilizing special procedures and materials to minimize introduction of stable sodium and to eliminate contaminants [18]. The ^{22}Na was purchased as sodium chloride in water with a specific activity of 1200 mCi/mg (Equivalently, the Na was enriched to 19% ^{22}Na). The target was made by depositing the solution into a shallow depression in a 8.5 micrometer thick aluminum foil. A heat lamp shining on the sample served to speed evaporation of the water. The resulting

deposit was fairly uniform and approximately 3 mm in diameter and contained about 75 ng of ^{22}Na. The target was employed in successful measurements of the (n,p) and (n,α) cross sections for ^{22}Na [4].

Five ^{36}Cl targets were prepared by the evaporation technique. Although successful measurements of the cross section relative to ^{6}Li have been made [2], an absolute measurement has been hampered by the small size of the thermal cross section, and by a ^{10}B contamination in the sample material. The targets were made using vendor supplied solutions having specific activities in the range of 14 to 18 mCi/g (or an enrichment of 42-54%). The first target contained 0.75 μCi and showed no visible residue when examined with a magnifying glass. However, the sample proved to contain too little ^{36}Cl for a successful measurement in a reasonable time. The second target contained approximately 9.0 μCi of ^{36}Cl. A white crystalline ring was noted around the edge of the depression following evaporation. Using this target, the proton peak from the $^{36}Cl(n,p)^{36}S$ reaction proved to be too broad, with too long a tail for a successful absolute measurement. In an effort to decrease the amount of solids in the target, a third target was prepared using half the aliquated amount used in preparing the second target. No water rinses of the micropipette were added to the depression. Even with these measures a white crystalline deposit was observed on the outer edge of the depression and the target was again too thick for an absolute measurement.

In an effort to reduce target thickness while maintaining an adequate amount of ^{36}Cl, the next two ^{36}Cl targets were prepared using a depression measuring 3 mm wide by 1.5 cm long in the backing foil. Ten lambda drops of the active solution were evaporated in ten different steps along the depression. The first "linear" target contained 40 μCi of ^{36}Cl, while the second contained about 12 μCi. Once more proton peaks that were too broad were observed, indicating that the sample was too thick. The major problem appears to be that the deposit is very nonuniform. All except the first sample show a crystalline ring around the edge of the deposit and several large crystals inside

this ring. Some experimentation has been done using wetting agents in an attempt to obtain more uniform deposits, but a suitable ^{36}Cl target has so for not been made.

Preliminary measurements have been made on targets of ^{54}Mn and ^{55}Fe. In both of these cases, the targets were too thick and contained too little ^{54}Mn or ^{55}Fe, and too much ^{10}B, for successful measurements. The ^{54}Mn was obtained from a commercial source and had a specific activity of 296.1 mCi/mg (or an enrichment of 3.8%). Most likely it was produced by a (n,2n) reaction on stable manganese in a nuclear reactor. It was provided as manganese dichloride in 0.5 M HCl. This was treated with an excess of NH_4OH, evaporated, and placed in a furnace at 370 C for five minutes. Ammonium chloride sublimes at 340 C. The manganese chloride was dissolved in distilled water and transferred to a 5 mm depression in a 8.5 μm thick aluminum foil with a micropipette. A heat lamp was used to speed the evaporation. Following evaporation and firing for three minutes at 360 C, a small quantity of a light grey colored deposit was observed in the depression. The bare target, containing approximately 6 mCi of ^{54}Mn and 20 micrograms of manganese gave a radiation reading of 4 R/hr at about 1 cm.

According to ref. [19], electrodeposition from organic solutions can produce targets that are in most cases uniform and have good adhesion. Also, because of the high efficiency and easy handling, the method is especially well suited for the preparation of radioactive targets. Using the method for the deposition of iron described in their paper, 2 mCi of ^{55}Fe (with a specific activity of 43.27 mCi/mg, or an enrichment of 1.8%), containing 46.2 micrograms of iron as ferric nitrate in a volume of 0.1 mL, was added to 2 mL of isopropanol in an electrodeposition cell. An aluminum foil on a stainless steel base served as the cathode while a platinum wire was used as the anode. Electrodeposition was carried out for a period 60 minutes at a voltage of 120 V and a current ranging from 0.39 to 0.77 mA. About 6% of the iron activity deposited on the platinum with the remainder depositing on the Al foil. Microscopic examination showed

deposits resembling coral and some minerals appeared similar to iron pyrite. This was definitely a non-uniform deposition and no successful measurements could be made.

6. Future Targets

A number of targets we have studied in this program have suffered from a low specific activity that prevented measurements of the emerging protons and alpha particles. To solve this problem and that of radionuclide unavailability from other sources, a target irradiation station (see Fig. 2) was constructed and installed in the neutron therapy beam line of the M. D. Anderson Hospital Cyclotron Facility. The target can be remotely inserted into the beam line for irradiation during periods between patient treatments. The first target selected for bombardment in this facility is ^{26}Mg (enriched to 99%) which will be used to make ^{26}Al. This target will be irradiated with protons in the energy range of 5 to 20 MeV in order to sweep the peak of the (p,n) excitation function. A graphite degrader foil is placed in front of the magnesium target to reduce the incident proton energy from 42 MeV to the appropriate energy required.

Because of the need for high specific activity, it is anticipated that other future targets will also necessarily be made by light ion bombardment on enriched stable isotopes. Alternatively, the isotope may be made by some less expensive means (e.g. a reactor or the IPF) and later enriched (and implanted) using an isotope separator. Such targets have already been made by using an "on-line" isotope separator [20]. In the end, both techniques may be fairly expensive.

Also, some of the targets we have made by deposition from a water solution have been too nonuniform for successful measurements. To obtain more uniform and robust targets, we are currently assembling our own vacuum evaporation apparatus (We were unable to find one that was available for making non-actinide radioactive targets).

7. Conclusions

With the advent of intense pulsed spallation neutron sources such as LANSCE cross section measurements on small quantities of short-lived radioisotopes has become possible. These measurements have applications in nuclear physics, nuclear astrophysics, and in many applied programs. Obtaining a suitable target is often one of the largest difficulties to be overcome in making a successful measurement. In the past few years, we have made several (relatively inexpensive) radioactive targets and employed them in successful measurements. However, we have also run into difficulties in several cases, both in obtaining suitable raw material, and in fabricating the targets. It has sometimes been our experience that although the technology for the making the desired target exists it is not always available, or it may be too expensive.

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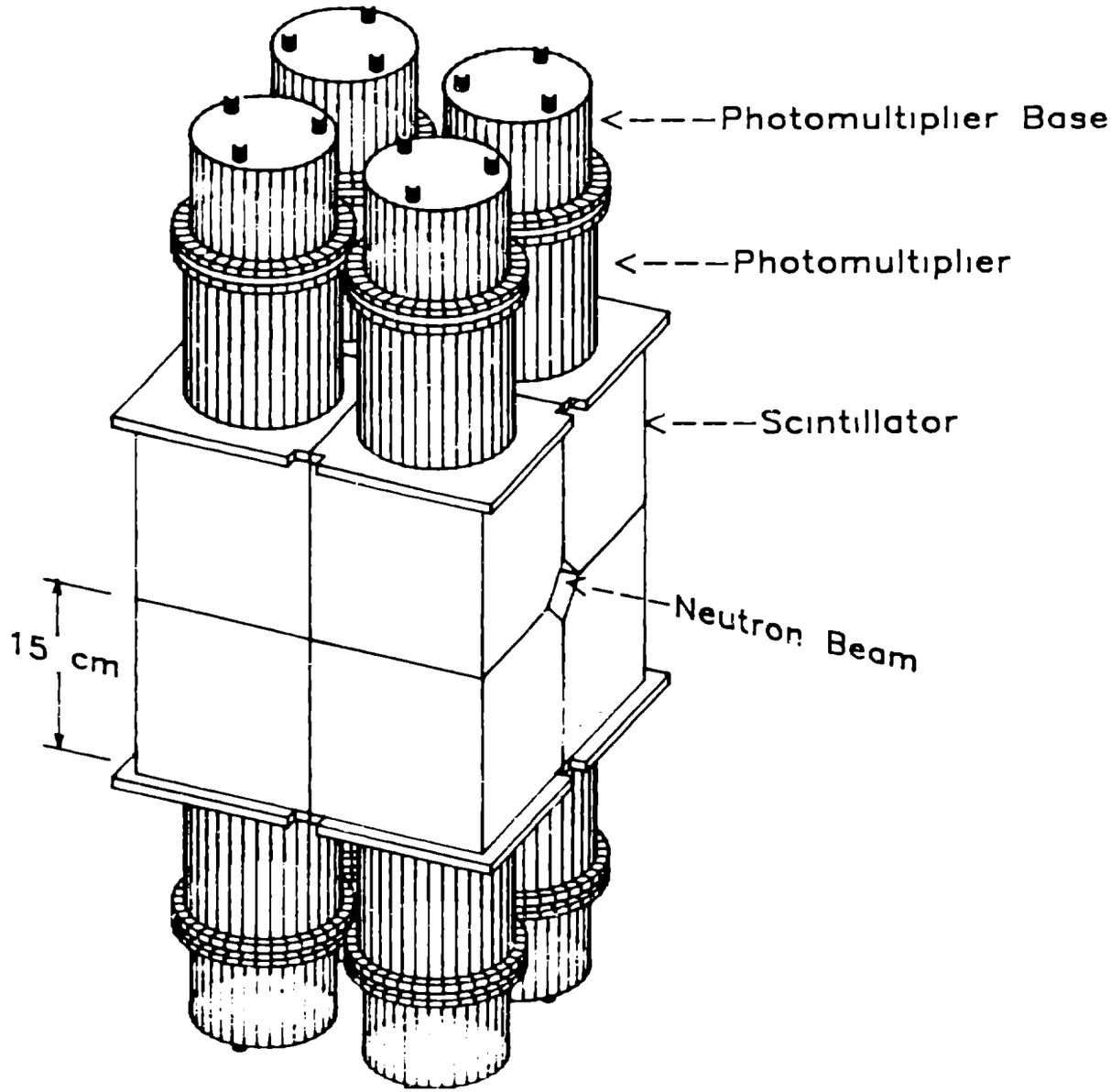
Figure Captions

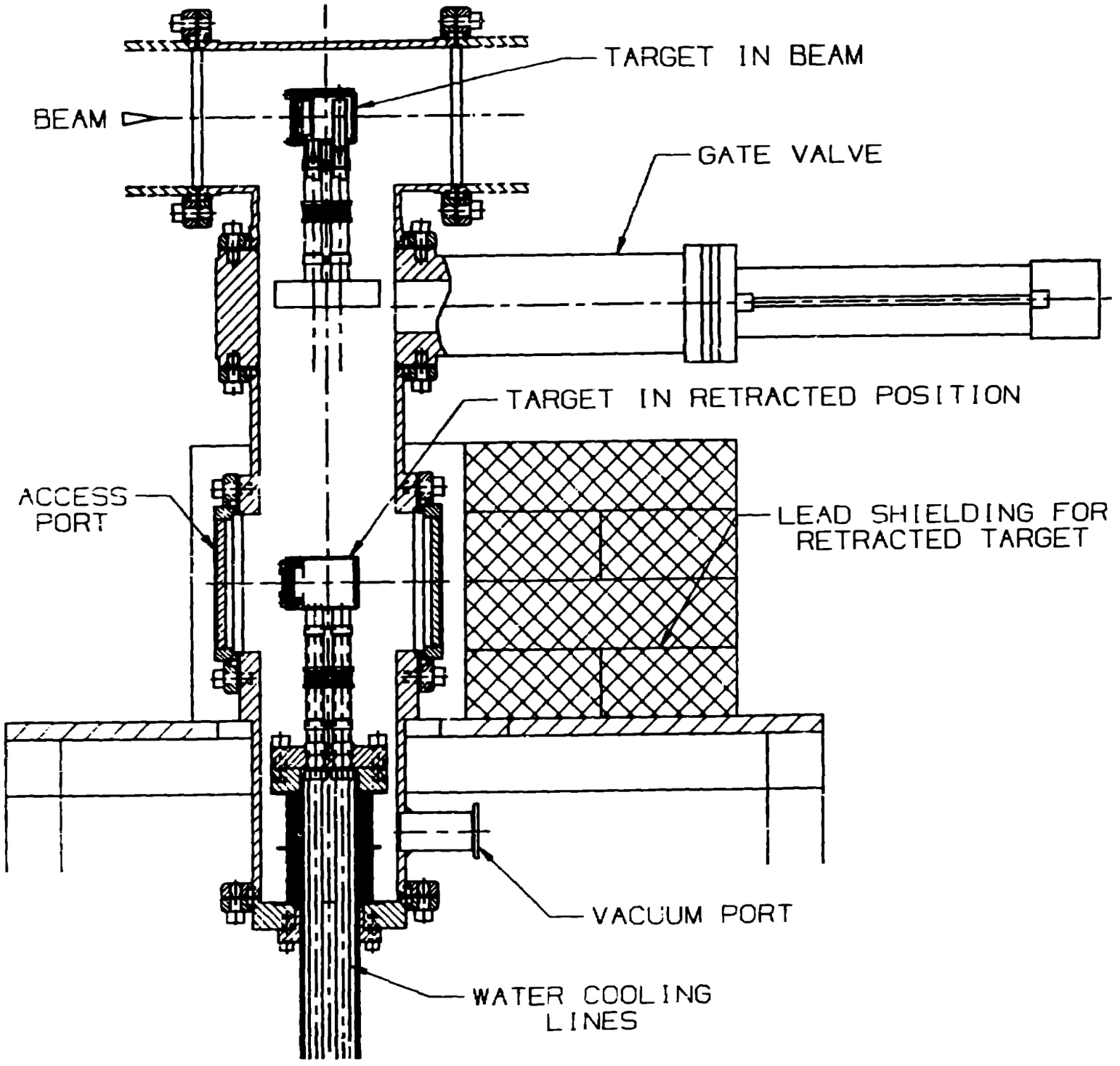
1) Barium fluoride detector for (n,γ) measurements on radioactive samples at LANSCE. The overall detector is a 30 cm cube of BaF_2 assembled from 8 cubes 15 cm on a side. Each cube is beveled on two edges so that when the 8 cubes are assembled there are two 4 cm wide holes through the center of the detector - one to allow the neutron beam to pass through, and the other for a target ladder. The target is placed at the center of the cube. Each BaF_2 scintillator is wrapped in teflon tape, encased in a thin walled aluminum box, and coupled to a 7.6 cm diameter photomultiplier tube. Hence there are 8 separate detectors which are held in place by an aluminum support structure which is not shown.

2) Target irradiation station for making radioactive targets. The station is installed on the neutron therapy beam line of the M.D. Anderson Hospital Cyclotron Facility. The target can be inserted into and removed from the beam remotely. When the target is retracted from the beam, the large gate valve can be closed allowing the target to be removed for processing without disturbing the neutron therapy beam line vacuum.

Barium Fluoride Detector

For $A^*(n,\gamma)$ Measurements at LANSCE




TARGET IN BEAM
BEAM
GATE VALVE
TARGET IN RETRACTED POSITION
ACCESS PORT
LEAD SHIELDING FOR RETRACTED TARGET
VACUUM PORT
WATER COOLING LINES