A SURVEY OF SOME APPLICATIONS OF CYCLOTRONS

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Introduction

There are several applications of cyclotrons for which no papers were submitted to this conference. It is the purpose of this paper to give a brief description of two of these applications, radiation damage and radioisotope dating, and to discuss some of the considerations involved in large-scale radioisotope production.

Radiation Damage

The various fusion energy programs face the problem of the effect of large fluences of neutrons upon the mechanical integrity of the front wall of the containment vessel. Various accelerators and reactors have been used to generate our present understanding of the neutron damage phenomena.[1] The matters under investigation include: (a) the accumulation of helium within the material - a process that leads to brittle fracture, (b) radiation induced creep and (c) radiation induced swelling. These processes have been investigated extensively for reactor spectra. However, the neutron spectrum for a fusion device has a large number of neutrons present with energy above 10 MeV. These high energy neutrons have much higher helium production rates, and they also produce a larger number of dislocations per neutron, so that experiments will have to be carried out to assess the effect of these differences. In order to perform these experiments in times that will produce engineering data in time for the construction, fluxes of the order of 3×10^{15} n/s over a volume of 100 cm^3 are needed.

The present choice of a source to provide neutrons of this intensity and a reasonable spectrum is a high current (200 mA) 35 MeV cw deuteron linac with either a beryllium or lithium target.[2] Unfortunately, such a source does not exist today. It is scheduled for 1983. To be in a position to use the source, when it does come on line, work must be done with the presently available sources and among these are the cyclotron of the Crocker Nuclear Laboratory at UC-Davis, the Naval Research Laboratory cyclotron and the Oak Ridge Isochronous Cyclotron. These have been used to characterize neutron spectra for the d + Be and d + Li reactions. The most intense source of 14 MeV neutrons currently available is the Rotating Target Neutron Source (RTNS-1)[3] at Lawrence Livermore Laboratory (LLL). Since both RTNS-1 and the cyclotron sources are intensity limited, one tends to put the experimental apparatus as close to the source as possible. Thus, the neutron fluxes in the target volume vary both in energy and number and must be determined experimentally.

I shall now discuss one particular study carried out by personnel at Atomic International (AI) in cooperation with groups at LLL and Argonne National Laboratory (ANL) that involved use of the UC-Davis cyclotron.[4,5] The experimental design is based on the use of radiometric dosimetry foils or wires, helium generation dosimetry wires, and helium generation samples all together in one capsule. These capsules are then irradiated at RTNS-1 and UC-Davis. Figure 1 shows the cross section of the target capsule irradiated at UC-Davis. There are three layers of helium generating materials sandwiched between four layers of dosimetry foils. There are samples of 23 pure elements: the stable isotopes of Li, B, Fe, Ni, Co, and Mo; and 14 samples of various alloys of interest as helium generation samples in addition to helium generation dosimetry wires. The wires are mostly circular so that any one wire will be irradiated with neutrons of a given energy. Figure 2 shows a drawing of the irradiation assembly that permits the mounting of the capsule and a number of additional helium generation samples, helium generation dosimetry wires, and radiometric dosimetry foil stacks at well-defined positions relative to the d + Be source.

Thus we see that the present use of the UC-Davis cyclotron permits the investigators to begin the characterization of the neutron field of a d + Be source and to gather information on helium generation dosimetry materials as well as helium generation rates in several materials - one small, but necessary, part of a big undertaking.

Radioisotope Dating

There has been a flurry of interest recently in the use of accelerators for the dating of samples containing radioactive isotopes.[6] This interest centered around some ideas presented by Richard Muller of the Lawrence Berkeley Laboratory.[7] The suggestion was made to use the accelerators to count atoms and not decays. In the case of cyclotrons, this is done by range discrimination. For ^{14}C, the interfering nuclide is ^{14}N. At typical cyclotron energies of 40 MeV, the range of the carbon is some 30% greater than that of nitrogen. Tandem accelerators perform this discrimination in the ion source, since $^{14}N^-$ is not a stable species. Another discrimination method that could be used is the E-dE/dx detection method. It is possible to combine the two techniques and pass the beam first through an absorber and then E-dE/dx particle identifier.[8,9] These procedures give discrimination levels of one in 10^{16}, which combined with transmission efficiencies of 10^{-5} yield sensitivities on the order of one in 10^{11}.

The measurements are relative measurements; *i.e.*, one measures the number of target nuclides for a known current of stable atoms. For carbon, it is the number of ^{14}C *vs.* the current of ^{14}N. This necessitates the use of two different detectors, a particle-identifier for the ^{14}C and a Faraday cup for the ^{14}N. There are a number of problems that are involved in improving the precision of these measurements.

The isotopes of present interest are:

^{14}C for the dating of archeological samples
^{10}Be for the dating of geochronology (*i.e.*, rock and ice) samples to determine variation of cosmic ray fluxes over geologic time and to date samples on a world-wide basis
^{36}Cl for ground water hydrology and measurements of the isolation time of ground water from the atmosphere

Figure 3 shows results obtained at Berkeley on a

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blind sample.[9] In this case, three samples were alternately fed into the ion source, a blank, the unknown and a sample of known age. Note the change in absolute values with time and the large background (due supposedly to contamination of the ion source and/or acceleration chamber). However, an age value in good agreement with radioactivity determinations was obtained, with uncertainty due principally to the background subtraction.

Large-Scale Radioisotope Production

Back in the early days of the development of cyclotrons, one could put a target of almost any material in the way of the cyclotron beam and produce a new radioactive species. After producing all these new goodies for the nuclear chemists to study, nuclear physicists began to become interested in the accumulation of cross section data; initially on natural targets and, when they became available, on isotopically separated targets. This accumulation of data led to the recognition of the existence of general systematic trends that could be described by the use of the compound nucleus theory of nuclear reactions. An early paper that summarized and projected yields is that by Martin, Livingston, Murray, and Rankin for the production of a great number of isotopes by 22 MeV protons.[10] Another such summary, but giving data for other projectiles, was presented at the Oxford conference.[11] It is true that radioisotopes can be made in almost any cyclotron. The success of the venture depends upon three things: 1) The willingness to use machine time in this way. 2) The development of target techniques, both in bombardment and in handling. 3) Sufficient beam in a suitable location to achieve substantial production.

Willingness to Use Cyclotron for Production

This section could also be subtitled "the necessity for a dedicated machine". The nature of the market for radiopharmaceuticals is such that there is almost no such thing as brand loyalty. Further, the doctor has several procedures at his command for a given problem; so that, in order to establish a radioisotope procedure as standard, the radioactivity must be available on a routine, scheduled basis. Thus, parasitic operation on a large research facility is a risky business except for all but long-lived activities. The accelerator must be ready all the time for isotope production.

Development of Target Techniques

The cyclotron beam is a charged particle beam and, when it interacts with matter, the principal method of energy loss is ionization of the target material. This process leads to a loss of energy with penetration and to a definite range for the particle. Since the cross sections for radioisotope production change with energy, the resultant isotopes will not be uniformly distributed by depth. The yield is proportional to the total number of target nuclei seen by the projectile; thus, it is advantageous to choose the projectile with the greatest range, i.e., protons. There are, of course, cases where the desired product cannot be made by proton bombardment so that other projectiles are occasionally used. The efficiency of medium-energy proton bombardment is such that only one in a thousand protons produces a nuclear reaction; the rest just go to produce heat.

The cyclotron internal beam has a radial distribution upon the edge of a probe that is triangular with the high intensity toward the cyclotron center. If one places a flat plate target tangent to the beam, the resultant pattern will be spread over a larger area than for normal incidence. If one now bends the target plate at a radius larger than the beam radius, a still larger spread can be obtained. For the Oak Ridge National Laboratory (ORNL) 86-inch cyclotron, patterns from 0.25" to as much as 10" have been obtained by this method.

Typical beam dimensions for a flat field, 60-inch cyclotron are 25 mm x 4 mm; the 86-in. has a big beam, 70 mm x 6 mm; and a normal Cyclotron Corporation product produces a beam 4 mm x 1.5 mm. Thus one can easily produce beam power densities of up to 70 kW/cm^2. Burnout tests with a large variety of target materials show that, for normal water cooling, the peak flux is 3 kW/cm^2, so one has to bring a lot of ingenuity to bear upon the problem of spreading the beam. The use of grazing incidence targets is one such procedure, as are oscillating and rotating targets.

Investigations of the heat transfer from cyclotron targets easily indicates that a boiling regime holds - one simply cannot get enough heat out without boiling. Luckily this regime of subcooled-nucleate boiling is also characteristic of nuclear reactors so that there has been a great deal of work done in this field.[12] The requirements for high heat transfer are: high water velocity, a surface well supplied with nucleation sites, turbulence promoters, and high static pressure.

The use of isotopically enriched target material should be considered when looking at the economics of large-scale production. The production rate of the desired product can be increased by a factor of two to five, and the production rate of an undesired product from another stable isotope of the target material can be reduced. The cost of the target material requires that it be recovered and reused for new targets. However the total cost of the two systems, when compared to the net production rates per hour, often leads one to use enriched targets.

A further constraint on the materials of target fabrication is that placed by the requirements of the chemistry. The process by which the irradiated target is reduced to a solution of the desired radioisotope generally involves one or more steps of separation of neighboring elements, and these are generally performed either by extraction or ion-exchange methods. The presence of some other material may make these separations either difficult or impossible, so that this matter should be studied before a final target construction is chosen. Another problem is the generation in some target component of a radioactive isotope of the target material, thus making the recovery and target production processes more difficult if enriched targets are used.

The target fabrication techniques that have been used include most of the normal metallurgical techniques. Usually because of matters of cost, possibility of difficult machining, size of available material, etc., the targets are composite; i.e., the target material is attached in some fashion to a base plate. The methods of attachment include electrodeposition, chemical vapor deposition, vacuum evaporation, soft soldering, brazing, casting and even flame spraying. The plates can be made of the material itself, if it is cheap enough and can be machined. Electroplating can be used for manganese, chromium, nickel, tin, cadmium, and zinc. Chemical deposition has been used for molybdenum[13] and will probably be useful for other refractory materials. The low melting point materials - lead, lithium, sodium, and thallium - can be cast into a depression milled into the target surface. Good success had been obtained in soft soldering nickel-plated zirconium, vanadium, and tantalum. Such targets can be operated at current levels high enough so that the solder is molten in the middle, if the beam is kept away from the edges of the soldered joint.

Figure 4 shows the final form of the target used at the ORNL 86-in. cyclotron for the bombardment of bismuth. The target base is aluminum, water passages were milled in the back, welded closed, the water manifolds welded on, the target material cast and machined smooth, the target bent, and the finish machining done on the water manifolds. It is a rather complex target and involves a number of fabrication steps in its production. The target plate shown in Fig. 5 is the high current target blank currently in use at the 86-in. cyclotron. The 0.5" thick copper plate is drilled on a rifle drilling machine with 3/16" dia. holes on 3/8" centers. The seal between water and vacuum is made by O-rings against the ends of the plate and has proved to be very satisfactory. These represent two levels of complexity in the consumable portion of the target.

The above discussions deal with stationary targets. Rotating targets have also been used. The earliest in my record is one at MIT that was designed in 1943 and used during the war for the production of ^{60}Co.[14] Krasnov's paper at the Oxford conference[15] shows a modern design, not only for a rotating target but also for two versions of static targets. Fig. 6 shows a sketch of the rotating target used at Amersham.[16] The head is beveled to give grazing incidence in addition to the rotation. They mount three complete target assemblies - complete with water supplies, driving motors, and seals - on a large turntable; they are so arranged that, while one target is being irradiated, another is available at a position supplied with a manipulator and lead-glass window for removal and installation. This led me to the final consideration in the design of targets. They must be easy to disassemble with remote handling tools. In fact, as much time and design consideration should be given to methods by which the target is removed from the cyclotron and transferred to the processing cell as to the design of the target itself.

Sufficient Beam in a Suitable Location

The answer to the question, "How much beam do you need?" is dependent upon the half-life or the activity to be produced. For half-lives on the order of one to three days, the desired currents are of the order of 100 to 300 microamperes. Preferably, this beam should be available as an external beam so that the large source of neutrons that the target represents can be separated from the cyclotron and thus the activation of the cyclotron can be kept to a minimum. If the process of extraction is only about 60% efficient, one is really not very far ahead. One possible design that we looked at was a separated-magnet cyclotron with one quadrant devoted to targetry. Such a machine does have the advantage that there is no iron in the way that limits the vertical aperture of the target mechanism - in fact the targets can easily be inserted vertically. Our real decision against this machine was the fact that the cyclotron will still be activated by the target-generated neutrons.

One very interesting idea that has been recently brought to realization is an H^- cyclotron with high currents.[17] There may be problems in getting a stripper for high currents and 500-hour lifetime, but this problem should be solvable.

The ORNL 86-in. cyclotron is the only cyclotron that I know of that was designed from the beginning as an isotope production machine. That this was so is evident in the large dee gap of 15 cm and the large physical size of the internal beam. It is also a high-current machine, being able to produce 2500 microamperes at 18 MeV. For the isotopes that it can produce, it is still the premiere production cyclotron in the world - for all that it is 35-years old.

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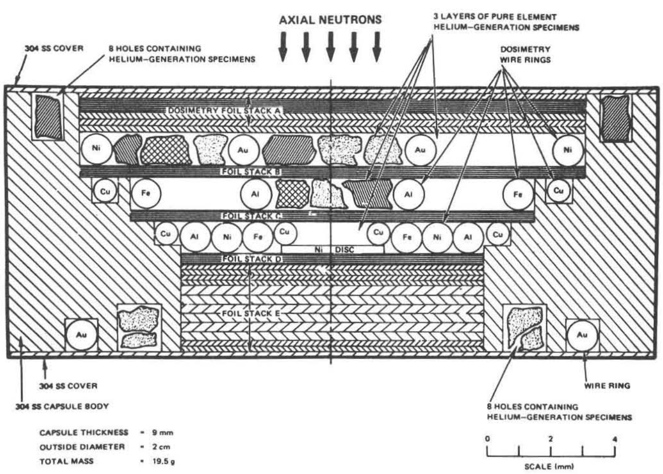

Fig. 1 Helium Generation Capsule for irradiation at UC-Davis cyclotron.

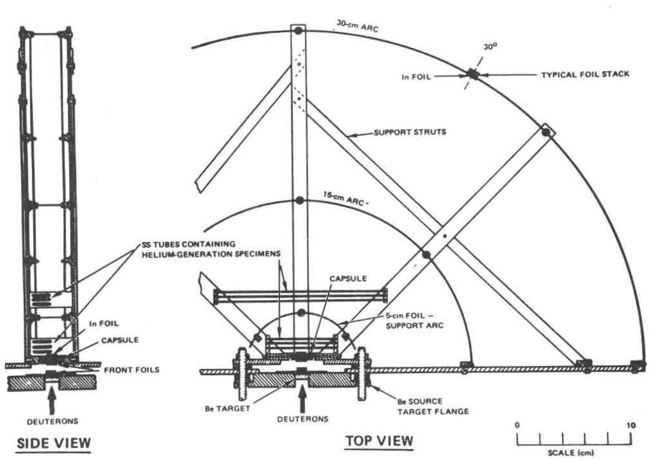

Fig. 2 Irradiation assembly for Foil Dosimeters and helium generation samples for the characterization of the neutron field at UC-Davis.

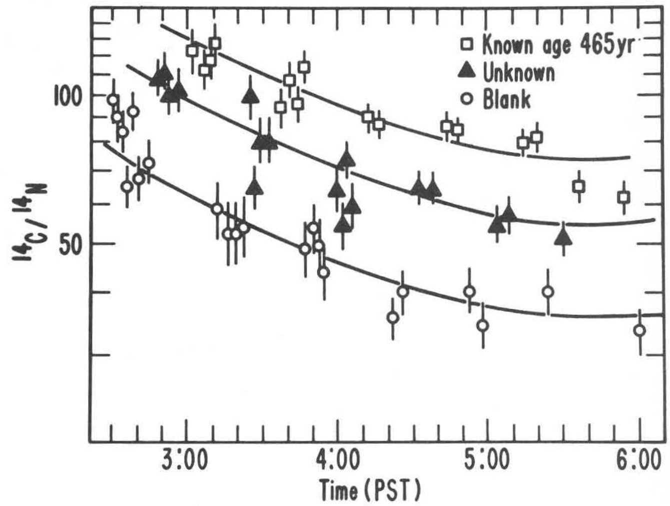

Fig. 3 The $^{14}C/^{14}N$ ratio as a function of time for three samples run at the LBL 88-inch cyclotron.

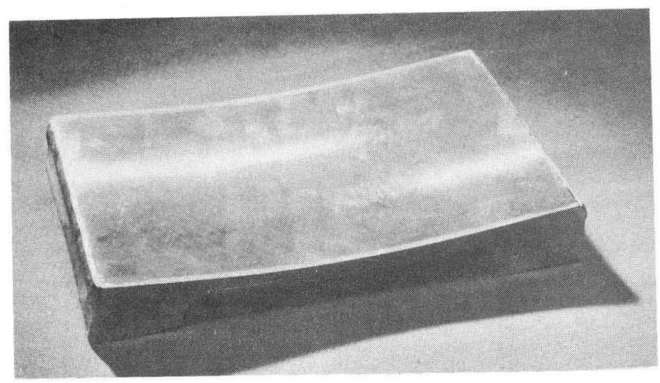

Fig. 4 Final Bi production target; ORNL 86-inch cyclotron.

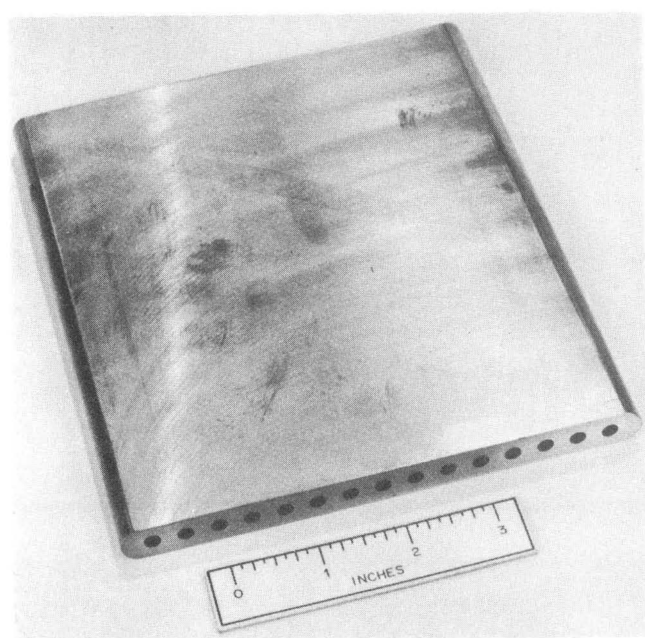

Fig. 5 Copper target blank presently used for high current production runs; ORNL 86-inch cyclotron.

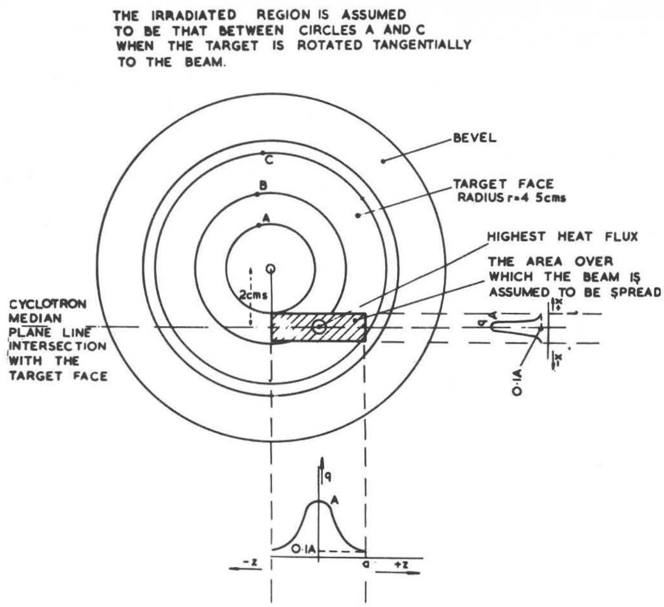

Fig. 6 The Amersham rotating target head.

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