

USES OF ACCELERATORS IN ENERGY R & D

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Summary

At many laboratories an increasing emphasis is being placed on energy-related research, often at the expense of more basic programs. The effects of this change can be clearly seen at accelerator laboratories, where tools traditionally reserved for nuclear and particle physicists are being applied in areas such as radiation damage, nuclear waste management, and materials science. The success of accelerator-based work in these fields is reflected by the increasing interest in proposals for new facilities devoted entirely to applied programs. Current proposals include various forms of intense neutron sources for fusion-related work, synchrotron x-ray sources for materials studies, and even the use of accelerators for large-scale nuclear waste disposal.

Introduction

In 1973, on the 500th anniversary of the birth of Copernicus, Werner Heisenberg gave a lecture at a joint Smithsonian Institute National Academy of Sciences symposium<sup>1</sup> in which he suggested that scientists do not have a great deal of freedom in the selection of their research. The problems available arise largely from contemporary developments as the result of a historical process. The choice is limited to the decision to participate in their solution or not.

We can see this process acting on our own research. We are faced with considerable technical and social problems associated with the environment and the supply of energy. In the last few years a variety of economic and social pressures have combined to change the emphasis of research away from areas like nuclear and particle physics toward more immediately practical goals. There is ample evidence in the program of this conference of the effects of these pressures on the users and designers of particle accelerators. This is, however, no cause for alarm. Experiments can be fun and beautiful whether they are in areas of basic research or applied research.

The major part of this talk is concerned with the application of accelerators to various energy related endeavors. I shall concentrate on current work at Oak Ridge because I am familiar with it and because it is illustrative of the type of work being pursued at many other laboratories around the world. At the end I shall mention some possibilities for the future, but first it may be instructive to look back two or three decades to some earlier examples of the practical application of accelerators.

Early Applications

During World War II E. P. Wigner made some calculations that indicated that the graphite cores of the Hanford reactors would swell under the influence of neutron irradiation.<sup>2</sup> The "Wigner disease" might seriously curtail their operational life. As reactor test facilities were not available, radiation damage studies were initiated using accelerators. Deuteron beams from the cyclotrons at Chicago and Michigan, and neutrons from d-Be sources based on the cyclotrons at Washington University and Berkeley were employed.<sup>3,4</sup> Although most of the damage studies were transferred to

reactors as soon as they became available, some work continued with accelerators into the 1950's.<sup>5,6</sup>

Another concern voiced during the war years was the possibility of an atomic weapon triggering an atmospheric chain reaction. Calculations, I believe by Teller and Konopinski, showed this to be extremely improbable and the matter was dropped. However, the possibility was again raised before the first fusion device was tested. Further calculations were made by Breit<sup>7</sup>, which confirmed the original estimates provided that no anomalies existed in the relevant cross-sections, in particular the exo-ergic reaction  $^{14}\text{N} + ^{14}\text{N} \rightarrow ^{16}\text{O} + ^{12}\text{C} + 10.5 \text{ MeV}$ . As these reactions had not been extensively studied, a heavy ion accelerator was built for the purpose. This was the Oak Ridge 63" cyclotron. Many of the first experiments in the now popular field of heavy-ion nuclear physics used the 25 MeV  $\text{N}^{3+}$  beam from this machine.<sup>8,9,10</sup>

Finally, there was an ambitious project initiated by E. O. Lawrence. In 1950 the AEC, worried by possible inadequacies in the supply of fissionable materials, approved Lawrence's suggestion of an accelerator to be used for plutonium or tritium breeding<sup>11</sup>. The first phase of this project, known as the Mark I, was a 25 MeV 50 mA proton linac. The final phase, eventually called the Materials Testing Accelerator (MTA) MkII was to have been a machine capable of breeding 467 kg of plutonium each year.<sup>12</sup> The specifications were awesome. It was to be a linac housed in a tank 60 ft. in diameter and 350 ft. long.<sup>11</sup> A 350 MeV deuteron beam with an intensity of 0.5 A was to bombard a 12 ft x 12 ft: uranium target assembly. Remarkable progress toward these goals was made. The target neutronics were studied using a small scale assembly at the Berkeley 184" synchrocyclotron.<sup>13</sup> The injector was built and proved capable of delivering 2 A of protons with a 19% duty factor in a beam diameter  $\approx$  4 inches. However, reactor breeding proved economically more attractive and the project was abandoned.

From these examples it can be seen that the practical application of accelerators is not new. Rather it went out of fashion for a time.

Current Uses of Accelerators

Today it is apparent that nuclear power will be required to provide a significant fraction of the world's electrical generating capacity within the next few decades. We can expect to see the large-scale introduction of Fast Breeder Reactors (FBR) and eventually perhaps Controlled Thermo-nuclear Reactors (CTR). Both of these projects present major technical problems to which accelerators can be applied.

Ion-Induced Radiation Damage

Radiation damage is now one of the most important areas of accelerator applications, and will be discussed in more detail later in these proceedings.<sup>15</sup> The heart of the problem lies in the two principal mechanisms by which neutrons induce radiation damage in metals. They are atomic displacement and nuclear transmutation.

\* Operated by Union Carbide Corp. for the ERDA.

A typical fast neutron scattering event in a metal results in approximately  $10^3$  atoms being knocked from their lattice sites along the path of the primary recoil atom. Nuclear reactions such as  $(n,\alpha)$  or  $(n,2n)$  produce not only displacement damage, but also an accumulation of impurity atoms within the lattice, helium and hydrogen being particularly important in their effects on physical properties.

Both of these mechanisms can be studied using accelerators. A few hours of irradiation with a heavy ion beam can produce displacement damage near the end of the ion's range which can only be achieved after years of irradiation in a fast reactor. Useful concentrations of helium or hydrogen can be achieved in relatively short times by direct injection with a or proton beams. Although the effect of fast neutron irradiation cannot yet be reliably determined from ion bombardments there are two compelling reasons why the effort is worthwhile. First there is the question of speed. It takes many years to irradiate a new alloy to relatively modest fluences in a fast reactor. In fact, as yet no neutron data exist at the highest doses expected in a commercial FBR. Secondly, reactor experiments are difficult to instrument, are expensive, and reactor space is very limited. The easier experimental access possible in an accelerator bombardment is, therefore, a significant advantage. The major disadvantages of accelerator work are beam heating, and the small volume of material that can be irradiated.

The phenomenon which is studied most extensively with accelerators is the swelling resulting from void formation. First discovered<sup>16</sup> in 1966 it is now known to occur in most metals at temperatures in the range of  $(0.35 - 0.55) T_m$ , where  $T_m$  is the absolute melting point. Figure 1 shows the growth of voids in high-purity aluminum irradiated in a reactor.<sup>17</sup> The final picture corresponds to an irradiation time of nine months, and a volume increase  $\sim 7\%$ . The stainless steels to be used in the Clinch River Breeder Reactor are much more resistant to swelling. Nevertheless, some core components are projected to swell by 30% during their lifetimes.

Soon after the discovery of void formation a group at Harwell began studies with 150 keV ions.<sup>18</sup> Today many laboratories are involved in such work using 1 MeV electron microscopes, electrostatic accelerators and cyclotrons. The work at Oak Ridge is fairly typical. Most bombardments use a 4 MeV Ni beam from the CN Van de Graaff in a program which can be divided into two main areas. The more basic studies involve small 3 mm diameter samples which are examined after irradiation by transmission electron microscopy. Beam densities  $\sim 1 \mu\text{A}/\text{cm}^2$  are used at temperatures in the range  $300^\circ\text{C} - 700^\circ\text{C}$ . Figure 2 shows voids produced in this way in an Fe-Ni-Cr alloy exposed in a few hours to a dose equivalent to  $\sim 2 \times 10^{23}$  neutrons/cm<sup>2</sup>.

The second area involves rapid comparisons of the swelling of different alloys in order to select promising materials for further study. A surface profilometry technique<sup>19</sup> is used to measure the swelling directly. Figure 3 shows a typical array of specimens, each approximately 3 mm x 1 mm, after irradiation. During bombardment a portion of the array was masked from the beam. Swelling in the bombardment region caused the surface to expand

outward leaving a depression behind the mask. Figure 4 shows a profilometer trace taken across such a masked region of a stainless steel specimen irradiated to a dose equivalent to  $\sim 3 \times 10^{23} \text{n}/\text{cm}^2$ .

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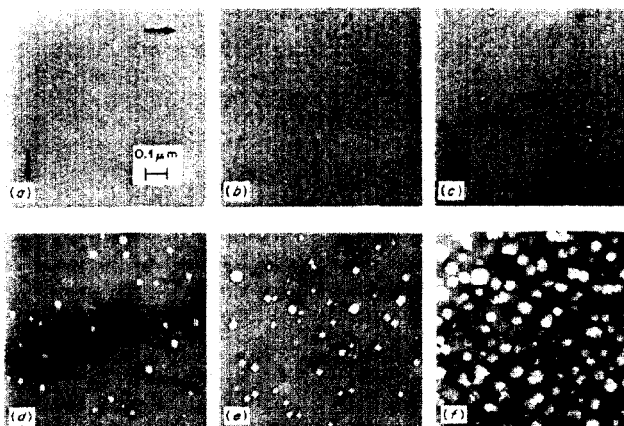


FIG. 1. Voids produced in high-purity Aluminum<sup>17</sup> irradiated to neutron fluences (in  $\text{n}/\text{cm}^2$ ) of (a)  $1 \times 10^{19}$ , (b)  $5 \times 10^{19}$ , (c)  $1 \times 10^{20}$ , (d)  $3 \times 10^{20}$ , (e)  $1 \times 10^{21}$ , (f)  $1 \times 10^{22}$ . The final picture corresponds to a volume increase  $\sim 7\%$ .

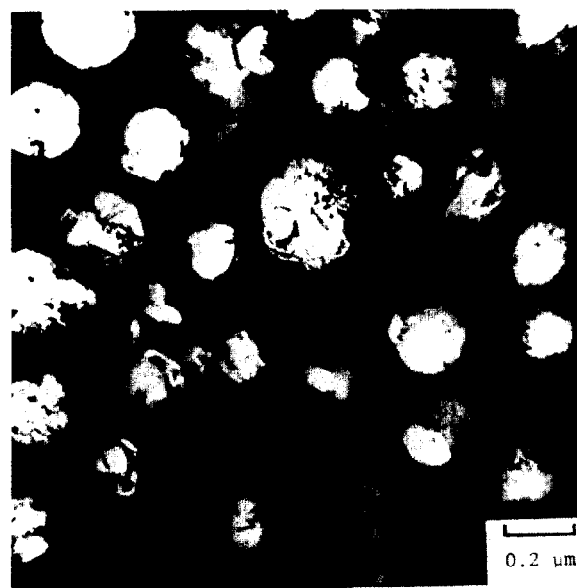


FIG. 2. Voids produced by Ni ion bombardment of an Fe-Ni-Cr alloy. The volume increase is  $\sim 50\%$ .

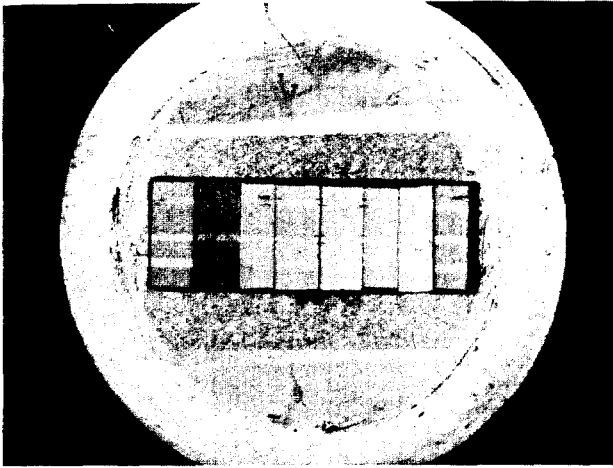


FIG. 3. An array of different alloys after irradiation with a 4 MeV Ni beam. The three different portions of the array which were masked from the beam in each of three separate irradiations can be seen as horizontal lines.

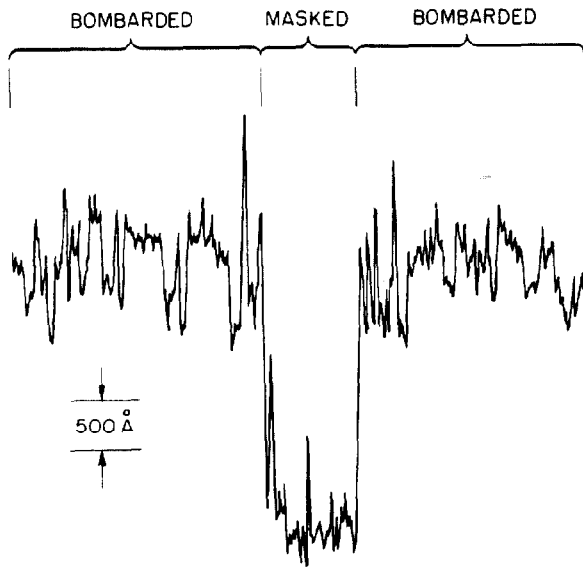


FIG. 4. A profilometer trace from a stainless steel sample irradiated by 4 MeV Ni ions. The measured step height corresponds to a maximum swelling  $\approx$  50%.

This step height corresponds to a swelling  $\sim$  50%. It should be noted that no neutron data exist at this fluence level.

A facility using two accelerators is being implemented at Argonne.<sup>20</sup> A Ni beam from the 4 MV Dynamitron and an  $\alpha$ -beam from a 2 MV Van de Graaff will be used simultaneously to bombard specimens, permitting better simulation of a neutron irradiation than is possible by alternating  $\alpha$ -injection and ion bombardment.

One common requirement of many damage irradiations deserves mention. Specimens are generally examined over a very small area, and often several specimens are irradiated simultaneously. It is, therefore, convenient to work with a very uniform beam density profile. Currently we are using a lens developed by C. H. Johnson<sup>21</sup> which was designed to transform a gaussian profile into a uniform spot. Using this lens we are able to focus 30% of the 4 MeV Ni beam onto a 7 mm x 10 mm rectangular target array with excellent uniformity, as illustrated in the X and Y profiles shown in Figure 5.

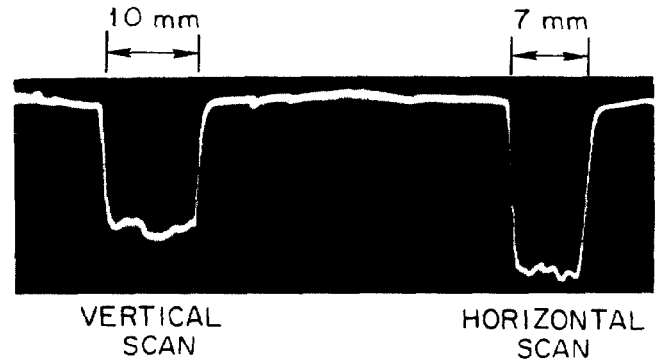


FIG. 5. The beam profile measured near the target position during a Ni ion bombardment in which the "ring lens" was used.<sup>28</sup>

Light ion beams are used to inject hydrogen and helium into samples for subsequent irradiation by ions or in a reactor. In this work beam heating and beam uniformity are the two most troublesome experimental problems. 60 MeV  $\alpha$ 's from the ORIC are also being used in the preliminary stages of an experiment designed to permit in-beam measurements of creep. Beam heating is particularly troublesome in this type of experiment as slight temperature changes of the sample can result in displacements which mask the effect to be measured. Even with temperature control by resistive heating beam stability  $\sim$  2 - 5%

over a 24 hour period will be required, together with good uniformity over the target area. Similar work is being pursued at other laboratories including Harwell, Jülich and NRL.

### Neutron-induced Damage

The CTR program faces many of the same radiation damage problems as does the FBR program. However, no test reactors exist with an appropriate neutron spectrum which will include a large number of neutrons with energies  $\approx$  14 MeV. Existing accelerator-based sources provide fluxes a factor of  $10^2$ - $10^3$  lower than those expected on the first wall. The Rotating Target Facility<sup>22</sup> at Livermore produces 14 MeV neutrons via the d-T reaction with fluxes up to  $\sim 10^{12}$  n/cm<sup>2</sup>/sec. The beam stop facility at LAMPF will give an evaporation spectrum with a useable flux  $\sim 10^{13}$  n/cm<sup>2</sup>/sec when the accelerator is running at full intensity.<sup>23</sup> Quasi 14 MeV sources based on the <sup>9</sup>Be(d,n) reaction are being used for damage studies at Oak Ridge and U.C. at Davis.<sup>24</sup>

A schematic of the Be target geometry used at Oak Ridge is shown in Figure 6. The target is just thick enough to stop the 40 MeV deuteron beam from the ORIC. Normal beam intensity is 20  $\mu$ A with a beam diameter  $\sim$  5 mm (f.w.h.m)<sup>25</sup>, giving a maximum neutron flux  $\sim 2 \times 10^{12}$  n/cm<sup>2</sup>/sec. Figure 7 shows the neutron beam profile measured during a 12 hour irradiation using a Nb dosimetry foil at the upstream end of the sample stack.

With the relatively low fluences that can be achieved with these sources experiments are mainly concerned with the primary effects of radiation. Two experiments are in progress at Oak Ridge. The first is a measurement of neutron sputtering from Nb samples. Kaminsky<sup>26</sup> has reported very high yields for particle emission from the surface of various materials subjected to 14 MeV neutron irradiation. Average yields for Nb as high as 0.25 Nb atoms/neutron have been quoted. Such an effect would cause rapid poisoning of a fusion plasma and significant erosion of the first wall material. Measurements made at Oak Ridge<sup>25</sup> have only been able to place an upper limit on the yield from Nb of  $10^{-4}$  Nb atoms/neutron. As yet there is no satisfactory explanation of the discrepancy between these two experiments.

In the second experiment Cu and Nb single crystals have been irradiated to fluences up to  $2 \times 10^{17}$  n/cm<sup>2</sup>/sec. The resulting defect clusters are being characterized using x-ray diffuse scattering and transmission electron microscopy. The objective is to correlate the 14 MeV neutron damage with that produced in fast reactor irradiations and ion bombardment studies.

### Materials Analysis

Energy related research involves many problems in materials analysis. Generally accelerators are too expensive to be used for this type of work, but there

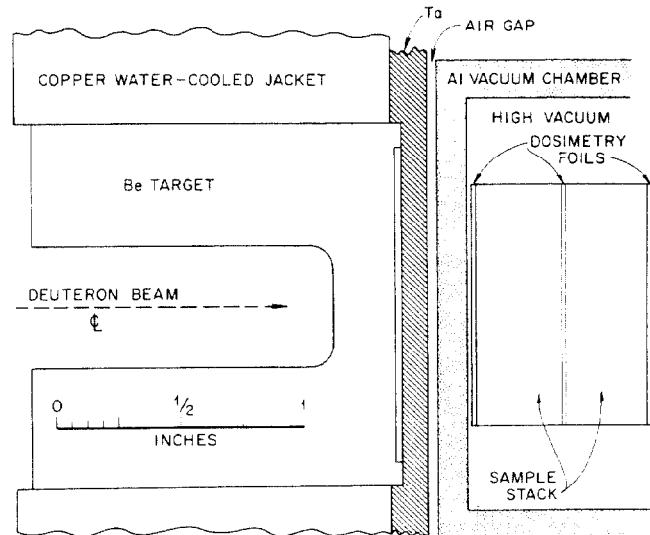


FIG. 6. The target geometry used for the ORIC 14 MeV neutron source. The 40 MeV deuteron beam intensity is normally 20  $\mu$ Amps, with a spot diameter  $\approx$  5 mm f.w.h.m.

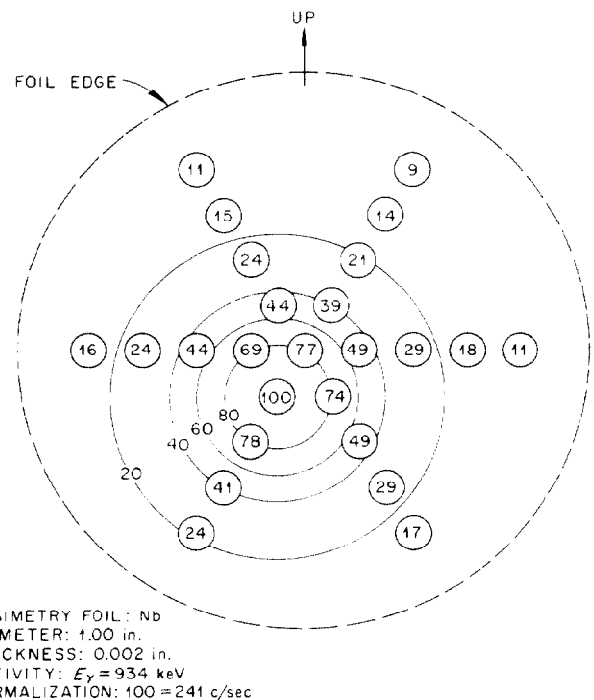


FIG. 7. The neutron flux profile measured at the front of the sample stack using a Nb dosimetry foil. The foil diameter is 1 inch.

are many specific cases where their use is advantageous. Only a few examples will be cited here.

a) The presence of hydrogen and helium in metals has already been mentioned as an important factor in radiation damage work. Depth profiles of these elements in thin foil samples can be quickly and conveniently measured using an identical particle scattering technique reported by Cohen et al.<sup>27</sup> The method as used for hydrogen assays is illustrated in Figure 8. A proton beam is used to produce  $p + p$  scattering from hydrogen in the foil. Both protons are detected in coincidence at  $\pm 45^\circ$  to the beam axis. The sum of the energies deposited in each detector bears an almost linear relationship to the depth at which the scattering occurred. Depth resolution  $\sim 1/10$  of the foil thickness can be achieved with concentrations as low as a few p.p.m.

b) In a hypothetical light water reactor accident involving loss of coolant, water would be injected into the reactor core causing oxidation of the zircaloy fuel cladding. Subsequent oxygen diffusion into the cladding material causes a marked reduction in its ductility. To evaluate this effort, the diffusion of  $^{18}\text{O}$  in zircaloy is being measured by Perkins et al. using a technique described by Condit and Holt<sup>28</sup>. Zircaloy samples are first oxidized using  $^{18}\text{O}$  and then annealed. The sample is then cut perpendicular to the oxidized surface and the fresh surface irradiated with 2.7 MeV protons, producing a 2-hour  $\beta^+$  activity from the  $^{18}\text{O}(p,n)^{18}\text{F}$  reaction. The samples can then be autoradiographed and the distribution of  $^{18}\text{O}$  measured.

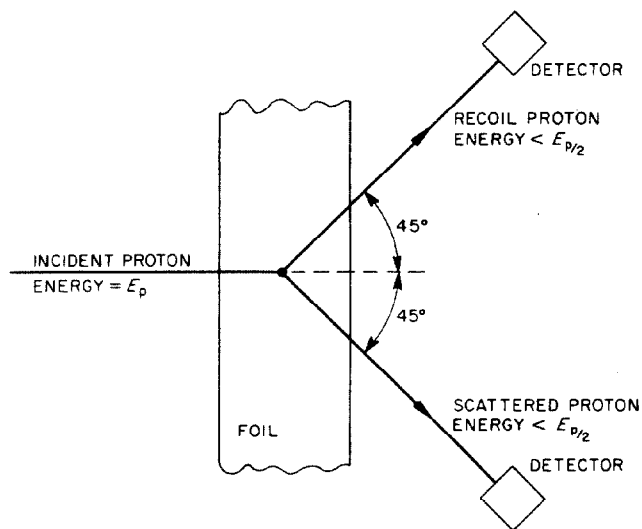


FIG. 8. Scattering geometry used to obtain the depth profile of hydrogen in a thin foil. The same method can be applied to helium assays if an  $\alpha$ -beam is used.

c) Accelerator based pulsed neutron sources can be used for non-destructive assay of fissile materials. The neutrons induce fission events, which then result in delayed neutron emission. By suitably tailoring

the incident spectrum it is possible to distinguish between fissile and fertile materials. Work of this type has been done at Los Alamos in connection with the Nuclear Safeguards Program.<sup>29,30</sup>

d) Intense x-ray sources are extremely useful in characterizing defect structures in materials, such as the voids formed by displacement damage. With sufficiently high quality beams, such as those available from synchrotron sources, it becomes possible to eliminate some of the tedious and costly electron microscopy presently required to examine ion-irradiated specimens.

Many other examples may be found in the literature. The interested reader can consult references 31-35.

### Neutron Cross-Sections

For many years neutron cross-section measurements have been made in support of reactor programs. A relatively new area is concerned with the management of high-level radioactive waste generated<sup>36</sup> in nuclear fuels reprocessing plants. One approach to the disposal of the longer-lived radio-nuclides, particularly the actinides, is to recycle them in a suitable reactor transforming them into shorter lived fission products.<sup>37</sup> Proper evaluation of this scheme requires good neutron cross-section data, especially for neutron-induced fission of the actinides. Experimentally this is a very difficult measurement because of the activity of the target material and the small quantities available. A group working on the ORELA has made such measurements with samples as small as 80  $\mu\text{g}$ .<sup>38</sup>

### Future Applications

In the past few years we have seen an increasing use of existing facilities in applied areas. Now we are beginning to see major new facilities expressly designed for applied work. At least two such machines are to be discussed at this conference. There is a proposal for a 30 MeV 100 mA deuteron linac to provide high energy neutrons for CTR studies via the d-Li reaction<sup>39</sup>, and another for a 2 GeV synchrotron x-ray source.<sup>40</sup> There is a LASL proposal for a d-T neutron source based on a 1 A tritium beam bombarding a supersonic deuterium gas target<sup>41</sup>, and a Canadian proposal for an accelerator-based spallation neutron source for nuclear breeding.<sup>42</sup> One concept that might be new to some of you is an ideal subject to end this talk as it leads directly to the next speaker's topic; Advances in Electrostatic Accelerators. The proposed machine may be truthfully, if sensation-ally, described as an orbiting 10,000 GeV 0.25 A dust accelerator.

The idea is discussed briefly in a report<sup>43</sup> by Dennis O'Keefe associated with the recent Batelle study on radioactive waste management.<sup>36</sup> One alternative to actinide recycle is extra-terrestrial disposal, preferably by ejection out of the solar system. This requires an enormous amount of power if conventional rocketry is used because payloads would be a very small fraction of the rocket's weight. However, the wastes could be placed in a low earth orbit by a space shuttle. They could then be formed into electrically charged particles  $\sim 1 \mu\text{m}$  in diameter, and accelerated to solar escape velocity (42 km/sec) by a 20 MV electrostatic accelerator. 5 MW of beam power would be

sufficient to dispose of the wastes from one hundred 1000 MW capacity nuclear power plants. Such particle accelerators have been built.<sup>44</sup> One based on a 2 MV Van de Graaff was used for micro-meteorite impact studies.<sup>45</sup>

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